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Present issue reports on the works performed in the Institute of Radiation Safety and Ecology under National Nuclear Center of the Republic of Kazakhstan during the years 2007 – 2009. The articles in the Proceedings cover radioecological issues related to former Semipalatinsk nuclear test site and adjacent territories, conventional hazards and general problems of radiation safety assurance at some other sites in Kazakhstan. The Proceedings are intended to provide readers with qualitative, reliable and unbiased information about present situation at a very unique territory called "Semipalatinsk nuclear test site (STS)".

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FOREWORD FROM THE EDITOR

This book presents the outcomes of works mainly performed during 2007-2009 in the Institute of Radiation Safety and Ecology under National Nuclear Center of the Republic of Kazakhstan (IRSE NNC RK). Activities of the Institute are mainly focused on comprehensive studies of ecological situation at former Semipalatinsk Nuclear Test Site (STS) and on its influence on the environment and people's health at adjacent territories.

By now we have accumulated considerable experimental data on STS which allows us to assure radiation safety effectively and completely for any research activities at the site or during remediation activities there. Accumulated knowledge enables us to state with confidence that the major part of lands within STS contour is almost "clean" what has been verified by special thorough investigations at northern part of STS: outcomes of those studies were presented in the first volume of the Proceedings.

Still, one should not mitigate the complexity and scopes of problems in the region taking into account the total amount of radioactivity contained in the bowels. Keeping in mind the scale of the site and a variety of tests performed there we understand that the available information can not be considered as completely comprehensive. In terms of further investigations and practical measures aimed at improvement of radiological situation there, we consider the following as the primary concern:

- realization of additional complex studies at "conditionally clean" lands aimed at further transfer of these lands for commercial use;
- studies of underground migration of radiological contaminants outside the testing grounds – indications for migration of certain radioactive isotopes (tritium) for several tens of kilometers from the original test sites have been revealed;
- inventory of areas with radioactive contamination and development of projects for their rehabilitation or conservation;
- development and implementation of programs aimed to prevent non-authorized access to radioactively contaminated areas at STS;
- preparation of radiation-hygienic passports of inhabited localities on the territories of STS potential impact and establishing of regular radiological monitoring there.

As it can be seen from the papers presented in this issue, the scope and range of National Nuclear Center's activities in these fields is quite large. But I would like to also note that people used to consider Semipalatinsk Test Site as a source of radiological hazard, explaining virtually any and all observed phenomena by influence of radiation, what is scarcely so. Possible influence of other, non-radiological, factors such as heavy metals still needs better studying and understanding as well as deposits of various minerals within STS contour.

Among the primary concerns we also consider studies of places where catastrophic phenomena like gas eruptions or soil collapses are possible: we have revealed some

processes for uncontrolled subsoil gasification, thermal destruction of coals at certain localities of the Site, in particular, at "Balapan" testing ground.

Works presented in the proceedings are quite diverse like in any book of this kind, but I hope that a reader would find here reliable and comprehensive information about present conditions at former Semipalatinsk nuclear test site to better understand this unique place.

Deputy Director General NNC RK for radioecology and Director of the Institute of Radiation Safety and Ecology **Lukashenko S.N.**

CHAPTER: RADIOECOLOGICAL SITUATION AT THE FORMER SEMIPALATINSK TEST SITE AND ADJACENT TERRITORIES

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ECOSYSTEM OF SHAGAN RIVER AND MAJOR MECHANISMS FOR ITS FORMATION

S.A. Aidarkhanov, S.N. Lukashenko, S.B. Subbotin, V.I. Edomin, S.V. Genova, A.V. Toporova, N.V. Larionova, E.Yu. Pestov*

Shagan River is the main water artery of the Semipalatinsk test site and together with the adjacent territory forms the ecosystem that includes groundwater and surface water, bottom sediments and coastal soils, air basin, vegetation and fauna. All these components have been exposed, in varying degrees, to radioactive contamination caused by nuclear tests at the Semipalatinsk test site. In this paper there were analyzed and synthesized the materials on radioecological studies in this area, described the overall picture of the current state of the ecosystem, the principal mechanisms of its formation has been considered. There were identified two major areas with fundamentally different types of radioactive contamination. The first zone is confined to the crest of the crater around the "Atomic" lake; here there is areal radionuclides contamination - fission products and neutron activation, formed during the excavation explosion in the borehole No.1004. The second area is located 4-5 km from the "Atomic" lake. Here, tritium migrates into the groundwater and surface water of Shagan River from the area of "warfare" boreholes of "Balapan" site, and its concentration in river water exceeds the intervention level by 2 orders of magnitude. The water of this river section is actually a liquid radioactive waste. So urgent is the issue of removal of part of Shagan River and adjacent territories out of economic use.

Key words: nuclear tests, "Atomic" lake, ecosystem, hydrogeology, radioactive contamination, radionuclides, boreholes.

INTRODUCTION

The low-water river Shagan (Figure 1-2) with the tributary Ashchisu is the longest surface river and the main waterway in the territory of the Semipalatinsk Test Site. It flows along the STS eastern boundary and is a left-bank tributary of the Irtysh River. The watershed area of the river does not exceed 10,000 km^2, the length of the main gully is 275 km and the length of Ashchisu tributary is 115 km. The river head is situated on the slopes of the mountain range Kanchingiz at a height of 875 m.

In the place where the Shagan River in-flows into the Irtysh, the absolute elevation is 160 m, i.e. the difference in heights is 715 m. As the length of the riverbed is 275 m, the average slope is 0.003. The length of the riverbed on the STS territory is about 50 km and the average slope is 0.002. The watershed area of the left-bank part of the site, where test boreholes are located is about 900 km^2.

The riverbed is characterized by a low speed of the river flow, branching, deadlock creeks and water logging of banks. The sector of the river flow at a distance from 12-15 to 30-40 km from the "Atomic" lake has several places where the riverbed disappears (dries up) and reappears again.

* Institute of Geophysical Research NNC RK, Kurchatov

Figure 1. General view of the Shagan River

1. POTENTIAL SOURCES OF ECOSYSTEM POLLUTION

Radionuclide contamination of the components of the Shagan River ecosystem is mainly caused by the following factors:

- global atmospheric fallouts;
- atmospheric fallouts from surface and atmospheric tests on the test ground "Opytnoye pole";
- atmospheric fallouts from excavation explosion in the borehole 1004;
- transfer of radionuclides by wind and surface waters from the soil heap around the "Atomic" lake;
- inflow of ground waters contaminated with radionuclides from the area of "warfare" boreholes.

1.1. Global atmospheric fallouts

Reference [1] gives theoretical assessment of the density of global fallouts based on the following initial data:

- $5.99*10^{17}$ Bq of ^{90}Sr (from $6.04*10^{17}$Bq released to the atmosphere) and $9.6*10^{17}$ Bq of ^{137}Cs (from released $9.64*10^{17}$ Bq) had precipitated on the earth surface by 1990;
- area of earth surface is 510 Tm^2;
- majority of precipitations (~75 %) was registered in the northern hemisphere;
- uniform distribution of radionuclides on the earth surface;
- depth of radionuclide distribution in lithosphere is 0.05 m;
- soil depth is 1600 kg/m^3.

Based on the above data the specific activity of global fallouts with account for the half-life period in 2009 was 15.2 Bq/kg for ^{137}Cs and 9.4 Bq/kg for ^{90}Sr, which approximately corresponds to the middle of the range of values presented in literature sources (4-29 Bq/kg for ^{137}Cs, 1-19 Bq/kg for ^{90}Sr, 0.02-5 Bq/kg for $^{239+240}Pu$).

Literature data provides no information about such radionuclides as ^{151}Sm and ^{99}Tc. As their concentration is now equal to 8% and 1.6% of that of ^{137}Cs, respectively, based on theoretical calculations with high degree of probability one can suppose that activity of ^{151}Sm and ^{99}Tc in global precipitations amounts to 1.2 and 0.2 Bq/kg, respectively.

1.2. Atmospheric fallouts from surface and atmospheric nuclear explosions on the "Opytnoye pole"

30 surface tests were made on the "Opytnoye pole" in the period from 1949 to 1962. Surface nuclear tests caused high radioactive contamination of rather large areas, formed radioactive traces stretching for hundreds of kilometers from the epicenters of explosions [2]. Some of the traces pass across the area of the Shagan River watershed (Figure 2).

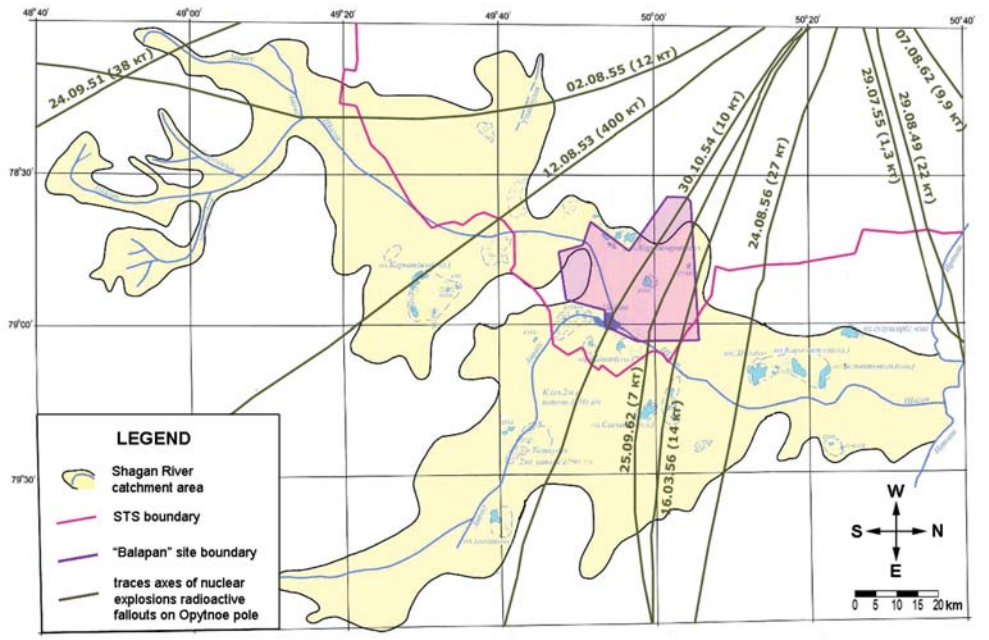


Figure 2. The watershed area of the Shagan River and traces of local atmospheric fallouts from surface explosions on the "Opytnoye pole".

No qualitative data on radioactive fallouts on the ground surface within the test site at the moment of tests are available.

In the 1990s, under the ISTC project K-053, the investigations of the radionuclide contamination of the STS territory were carried out [1]. The works included soil sampling along 8 lines from the epicenter of explosions in the places directly adjacent to the epicenter. The lines mainly passed along the axes of the traces of radioactive fallouts which mainly contributed to the STS contamination. An analysis of the soil samples enabled to determine peculiarities of radionuclides distribution in different directions and at different distances from the "Opytnoye pole". Figure 3 (Figure 3) shows such distribution for radionuclide ^{137}Cs.

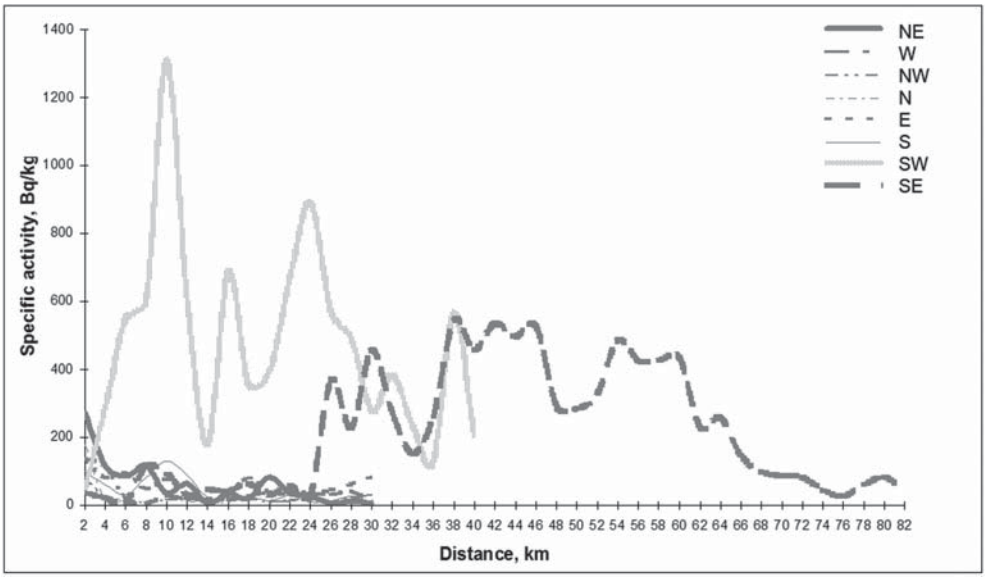


Figure 3. ^{137}Cs concentration in soil along different directions from the "Opytnoye pole".

The studied area is located in the zone of influence of the southeastern trace where maximal detected radionuclide concentrations in soils are 580 Bq/kg for ^{137}Cs, 390 Bq/kg for ^{90}Sr and 270 Bq/kg for $^{239+240}$Pu.

1.3. Atmospheric fallouts from the excavation explosion in the borehole 1004.

The excavation explosion in the borehole 1004 was made on 15.01.1965 in order to test techniques of creation of artificial water reservoirs in the arid areas. The power of the explosion was 140 kg and the charge depth was 178 m. The diameter of the formed crater at the crest level was 408 m. As a result of explosion a water reservoir was formed in the place of confluence of the Shagan and Ashchisu rivers, which was later called the "Atomic" lake.

The cloud after the explosion in the borehole 1004 and the radioactive trace were formed under anomalous distribution of temperature and wind direction in height, which caused formation of local radioactive trace of an intricate shape. For example, radioactive aerosols in the layer from 0 to 750 m were moved in the azimuth direction 330°, in the layer from 750 m to 2500 m the direction was 40-47°, and the upper part of the cloud, at a height over 2500 m, moved to the azimuth of 70°. As a result a branched trace was formed (Figure 4) [1].

The local trace from the explosion did not cause any considerable contamination of the territory. The radionuclide concentration in soil caused by the explosion reduces to almost background values at a distance of 1-2 km from the crest. A specific feature of the local fallout is high concentration of radionuclides – the products of activation of soil substances: ^{60}Co, ^{152}Eu, ^{154}Eu.

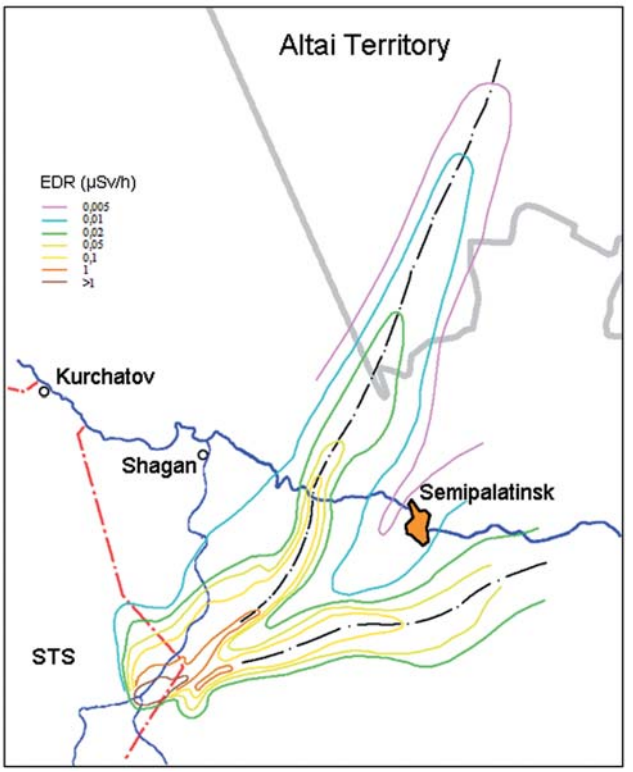


Figure 4. Scheme of the trace of radioactive contamination caused by underground nuclear explosion with soil outburst in the borehole 1004 (24 hours after the explosion)

1.4. Radionuclide transfer by wind and surface waters from the zone of soil heap around the "Atomic" lake

As a result of explosion the following long-lived radionuclides were produced: $^{239+240}$Pu – 8.5 Ci; ^{137}Cs – 800 Ci; ^{60}Co – 80 Ci; ^{152}Eu – 120 Ci; ^{90}Sr – 400 Ci; ^{3}H – $4*10^5$ Ci. Up to 40% of produced amount of radionuclides precipitated in the heaping area.

The explosion caused considerable contamination of the adjacent area. The results of radiometric measurements in the area around the "Atomic" lake gave the EDR values as high as 68 μSv/h and beta-radiation density equal to 350 part./(cm^2*min). In the samples taken in the vicinity of the "Atomic" lake maximal concentrations are 4100 Bq/kg for ^{241}Am, 3400 Bq/kg for $^{239+240}$Pu, 15 000 Bq/kg for ^{137}Cs, 10000 Bq/kg for ^{90}Sr, 21000 Bq/kg for ^{152}Eu and 13000 Bq/kg for ^{154}Eu.

The area of soil heap around the "Atomic" lake is to be considered as a serious potential source of contamination. From this zone radionuclides may be transferred by wind and washed by atmospheric fallouts to the surrounding area or to the Shagan riverbed with further transfer of radionuclides by surface waterways. According to the results of investigations in the place where the Shagan River flows out from the "Atomic" lake the concentration of

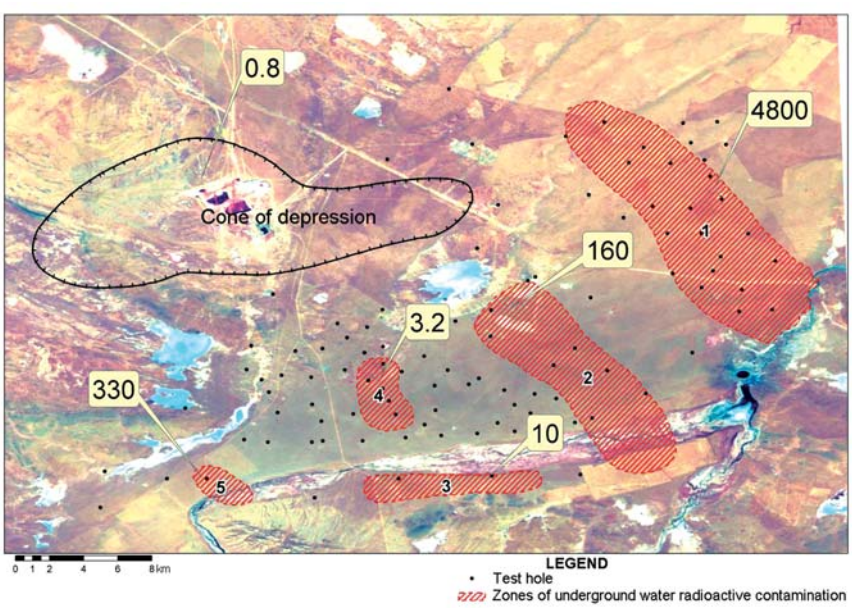
radionuclides is now 4.0 Bq/kg for ^{90}Sr, up to 1.5 Bq/kg for ^{137}Cs, 0.005 Bq/kg for $^{239+240}$Pu and 250 Bq/kg for ^{3}H.

1.5. Transfer of radionuclide-contaminated ground waters from the area of "warfare" boreholes

In the period from 1961 till 1986, 131 underground nuclear explosions of high capacity (up to 150 kilotons in TNT equivalent) were made on the test ground "Balapan" in the vertical mines – boreholes. The average number of boreholes per one square kilometer is 1 borehole per 7 km^2.

Radioecological investigations of ground waters on "Balapan" test ground started after the Test Site closure. A significant role in the investigations was played by 100 hydrological wells drilled in the territory at the time of nuclear tests. The results of investigations showed that high concentrations of ^{137}Cs and ^{90}Sr in ground waters were registered only at a very small distance from the epicenters of nuclear explosions, at a distance of 300 m from "warfare" boreholes the concentration of radionuclides goes down to several mBq/kg. Such low concentrations of radionuclides did not enable to detect any peculiarities in spreading of ^{137}Cs and ^{90}Sr in ground waters.

However, concentration of ^{3}H in ground waters varies in a wide range, from the minimal detectable activity, equal to 7 Bq/kg, to maximal values exceeding 4 million Bq/kg. As a whole, the test ground "Balapan" has a zonal structure of ^{3}H distribution. 5 areas of high ^{3}H level were detected. Figure 5 (Figure 5) shows the scheme of distribution of zones with high concentration of ^{3}H. The numbers give the values of concentrations in kBq/kg.



1 – northeastern zone; 2 – eastern zone; 3 – southern zone; 4 – southwestern zone, 5 – central zone.

Figure 5. Scheme of location of zones with high concentration of ^{3}H in ground waters on the "Balapan" test ground

The highest values of ^{3}H concentration were detected in the northeastern zone, where in the borehole 171 the ^{3}H concentration of 4800 Bq/kg was registered. In terms of geological structure the northeastern area of high ^{3}H values is located in the Zhananskaya contortion zone. It is a tectonic structure of up to 7 km in thickness. In this tectonic structure the rocks are dislocated and intensively layered with slates due to which this territory is easily flooded and affected by ground waters, which creates favorable conditions for intensive washing out of radionuclides from the central zones of nuclear explosions and their inflow into the aquifer.

2. PRESENT-DAY RADIOECOLOGICAL STATE OF THE ECOSYSTEM

The main factors which determined choice of the objects of investigation were the volume of information on the object, available instrumental and methodological base and, primarily, actuality and practical importance. Near the Shagan River there are several winter camps where people permanently live and work, in particular, breed and graze cattle. Hence, of primary importance are the objects determining the level of hazard for people's staying in the territory and possibility of radionuclide transfer from natural objects to the organisms of domestic animals and from it – to the products of cattle-breeding and, as a consequence, to human organisms.

Based on the above factors the following objects were chosen: soils and bottom sediments, surface and ground waters, vegetation, air basin and cattle-breeding products.

2.1. Soils and bottom sediments

2.1.1. Radionuclide distribution along the riverbed

In order to study contamination with artificial radionuclides and assess the state of radioactive contamination of the banks of the Shagan River, samples of bottom sediments (directly in the present-day riverbed) and riverbank soils (in the near-river zone at a distance of 10-15 m from the riverbed) were taken. The spatial distribution of the sampling points is shown on the scheme (Figure 6).

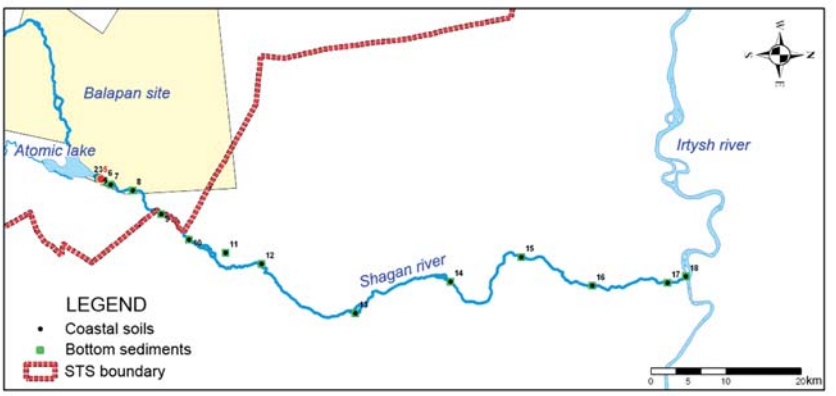


Figure 6. Layout of sampling points

The laboratory analyses showed the presence of artificial radionuclides ^{137}Cs, ^{241}Am, ^{60}Co, ^{152}Eu, ^{154}Eu (Figures 7–10) in bottom sediments and the riverbank areas. They have maximal concentrations in close vicinity of the "Atomic" lake, decrease as the distance from the lake increases and at a distance of 10 km have the values comparable with those typical of the background of global precipitations.

It should be noted that even at very small distances from the "Atomic" lake (50 m) the radionuclide concentrations in the riverbank soils and bottom sediments are much lower than in the soil heap (Para. 1.4). This effect may be caused by redistribution of the fragments by water flow along the riverbed.

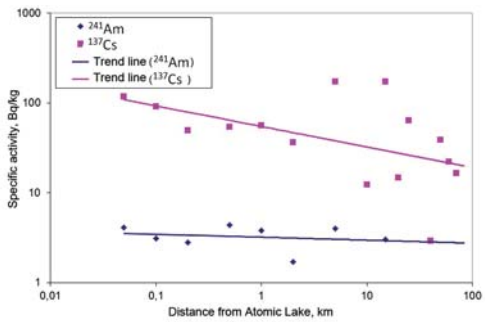


Figure 7. Specific activity of ^{241}Am, ^{137}Cs in the samples from riverbank soils

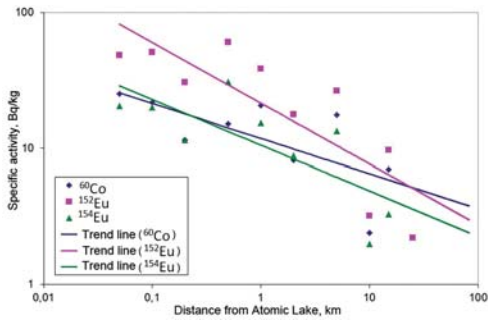


Figure 8. Specific activity of ^{60}Co, ^{152}Eu, ^{154}Eu in the samples from riverbank soils

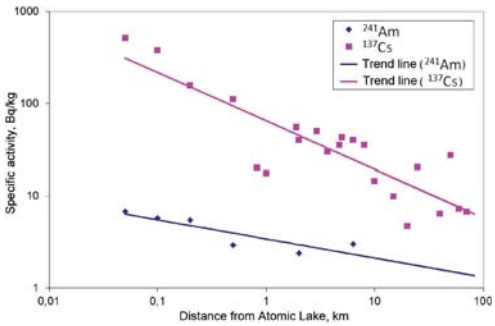


Figure 9. Specific activity of ^{241}Am, ^{137}Cs in the samples of bottom sediments

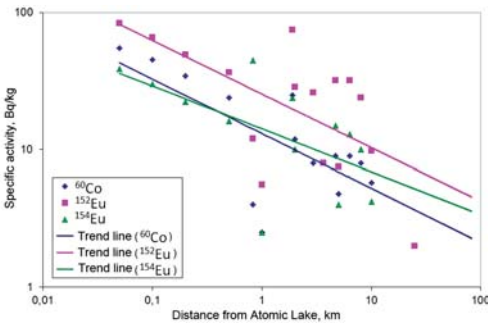


Figure 10. Specific activity of ^{60}Co, ^{152}Eu, ^{154}Eu in the samples of bottom sediments

A clearly pronounced dependence between the radionuclide concentrations, both in bottom sediments and the riverbank soils, and the distance from the "Atomic" lake is registered. The dependence is well approximated by power functions of the type $y = ax^b$, where y is a specific activity of the radionuclide, Bq/kg, x is the distance from the "Atomic" lake, km, a and b are dimensionless coefficients. Table 1 (Table 1) gives parameters of the above functions for all considered radionuclides.

Table 1.

Parameters of power functions approximating dependence of radionuclide concentrations in soil samples as a function of distance from the "Atomic" lake

Radionuclide	Bottom sediments			Riverbank soil		
	a	b	R^2	a	b	R^2
^{241}Am	2.89	-0.20	0.79	2.77	-0.14	0.60
^{137}Cs	64.3	-0.62	0.82	51.8	-0.36	0.37
^{60}Co	10.9	-0.53	0.87	9.59	-0.45	0.82
^{152}Eu	16.6	-0.59	0.89	18.8	-0.57	0.85
^{154}Eu	8.16	-0.51	0.89	8.88	-0.48	0.84

The observed dependencies may be interpreted as follows. The coefficient *a* gives the radionuclide concentration at zero distance from the "Atomic" lake, it must be approximately proportional to the amount of radionuclide produced during the explosion, which is confirmed by the available estimations according to which ~ 80 Ci of ^{60}Co, ~ 120 Ci of ^{152}Eu (which is a little higher) and ~ 800 Ci of ^{137}Cs (which is an order higher) were produced during the explosion. In the diagrams the coefficient *a* corresponds the height of the left edge of the trend line.

The coefficient *b* characterizes the rate of decrease in radionuclide concentration as a function of distance, and in the first approximation it is connected with the migration abilities of the radionuclide. For example, ^{241}Am has the lowest (by module) *b* values which enables to assume that it has higher migration ability than other considered radionuclides. However, an important role in this process may be played by additional factors: chemical processes in the riverbed, flow speed, additional inflow of radionuclides to the places of atmospheric precipitation traces and other factors. In the diagrams the coefficient *b* corresponds to the slope of the trend line.

A comparison of the expressions for the same radionuclide for the riverbank soils and bottom sediments gives the following conclusions:

1. The values of both coefficients for the riverbank soils and bottom sediments are almost the same. It is likely that there is no principle difference between the two types of soils in terms of their formation and history, and division into riverbank soils and bottom sediments shows only the present-day state of the riverbed.
2. The values of coefficient *a* for radionuclides ^{152}Eu and ^{154}Eu are higher in the riverbank soils and the values for ^{241}Am, ^{137}Cs and ^{60}Co are higher in bottom sediments. These differences are most likely within the error caused by poor statistics of points. It is practically impossible to find any cause-effect dependence, and there are no reasons to suppose that it exists, as it was stated before, this coefficient is determined by the activity of the sources i.e. the same object for both types of soils.
3. The values of the coefficient *b* in module are always higher for bottom sediments though the difference is small; it means that the rate of decrease in radionuclide concentrations in the bottom sediments is higher than that in the riverbank soils. This result enables to suppose that the processes of radionuclide washing out

from the bottom sediments are more intensive than the accumulation processes, however, in order to argue with a high degree of certainty one must get statistically significant differences, and the available information is not sufficient to make such a conclusion.

2.1.2. Radionuclide distribution across the river

In order to study the distribution of artificial radionuclides in bottom sediments and riverbank soils across the riverbed two segments of the riverbed with taking of surface and layered samples were examined. The first segment located at a distance of 1.9 km from the "Atomic" lake showed maximal values of radionuclide concentrations (except ^{241}Am). The second segment was chosen in the part of the riverbed with registered high ^{241}Am concentration in bottom sediments. It is located at a distance of 6.35 km from the "Atomic" lake.

The figures (Figures 11–12) show concentration of artificial radionuclides in bottom sediments and riverbank soils.

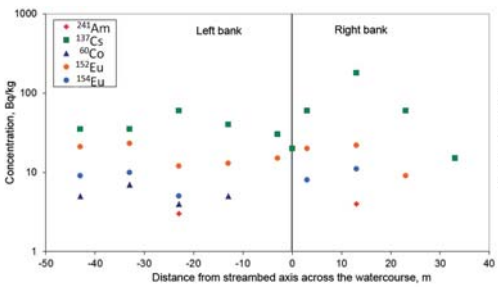


Figure 11. Content of artificial radionuclides in the samples of coastal ground and sediments 1.9 km from "Atomic" lake

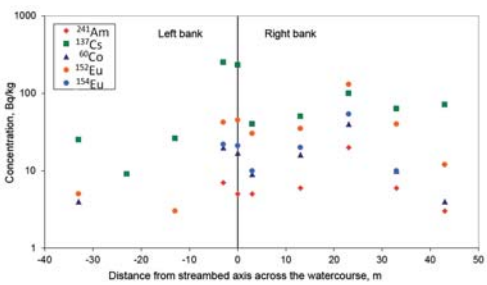


Figure 12. Content of artificial radionuclides in the samples of coastal ground and sediments 6,35 km from "Atomic" lake

The presence of artificial radionuclides was registered in both segments. Of special importance is the presence of activation products ^{60}Co, ^{152}Eu, ^{154}Eu at considerable distances from the "Atomic" lake. This fact requires additional investigations in order to study mechanisms of inflow of isotopes into the riverbed.

2.1.3. In-depth distribution of radionuclides

The figure (Figure 13) shows in-depth distribution of artificial radionuclides in the bottom sediments and riverbank soils.

As it is seen from the figure, the radionuclide concentrations in the second profile are higher than in the first one not only in the surface sediments but also in-depth. The second profile also has a point with extremely high concentrations of radionuclides. It is likely that such high concentration was caused by anomalous conditions for sediments in that place, for example, a sharp cavity in the relief, where surface waters from the surrounding area were washed down.

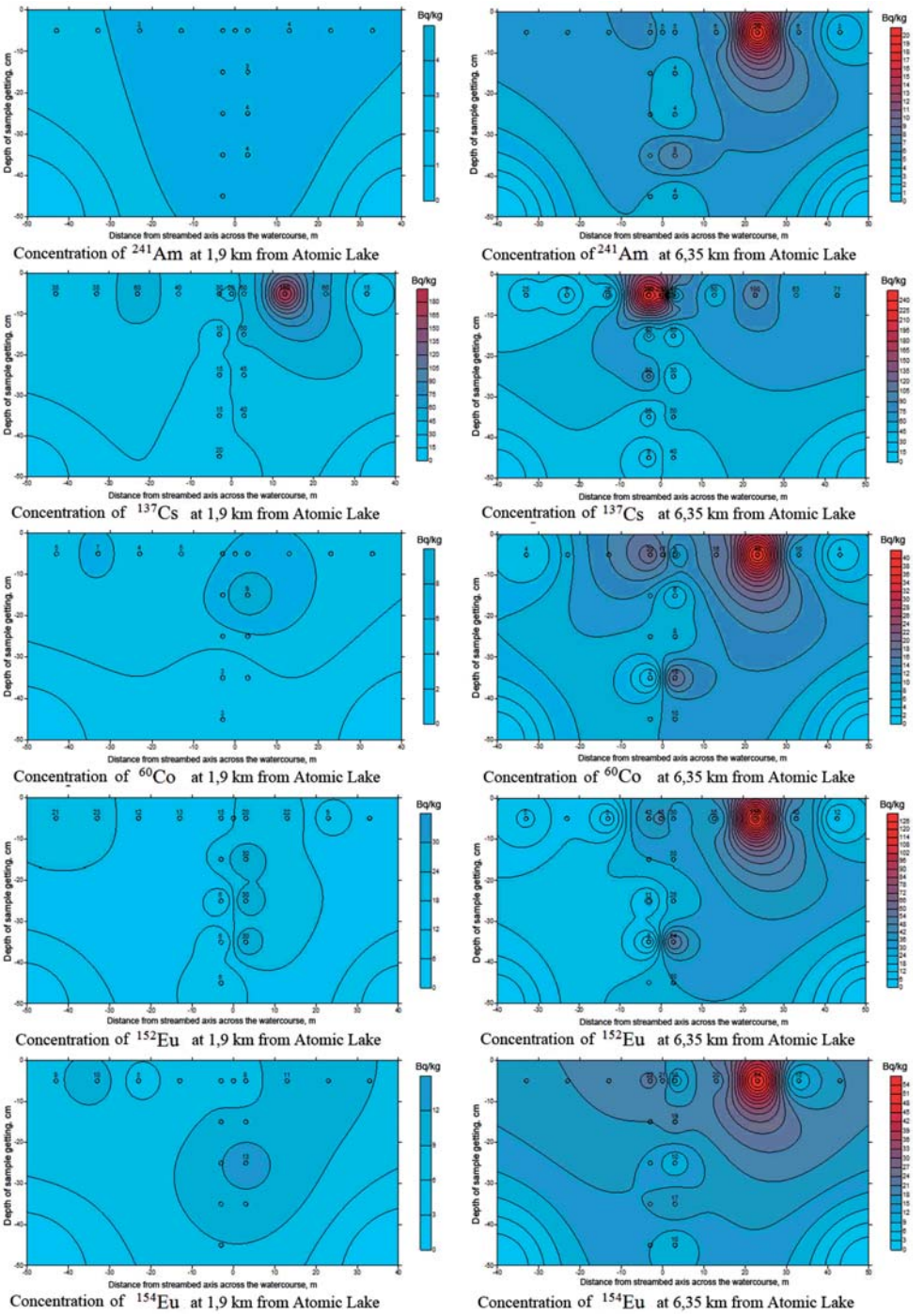


Figure 13. Distribution of artificial radionuclides in depth

2.1.4. Discussion of potential mechanisms responsible for contamination of soils and bottom sediments

Four of five potential sources of ecosystem contamination listed in section 2 may cause soil contamination, they are:

- global atmospheric fallouts;
- atmospheric fallouts from fallouts from surface and atmospheric nuclear explosions on the "Opytnoye pole";
- atmospheric fallouts from the excavation explosion in the borehole 1004;
- transfer of radionuclides by wind and surface waters from the soil heap around the "Atomic" lake.

It is necessary not only to study the role of each of the above sources in soil contamination but also to take into account some additional factors:

- radionuclides are transferred by surface waters in different forms. All possible forms must be subdivided at least into two components – transfer of dissolved radionuclides and transfer of a mechanical suspension of contaminated soil particles, and each component must be considered separately;
- radionuclides are locally redistributed in the landscape with time due to surface washing out and penetration of radionuclides into soil.

It is difficult to give quantitative assessments of all the above factors now. However, it is possible to make some assumptions about the role of each of the above-listed factors at a qualitative level using the data on the example of radionuclide ^{137}Cs given in paras. 2.1.1 – 2.1.3:

Global atmospheric fallouts. All studied objects have points where ^{137}Cs concentration corresponds to the background of global precipitations (15-20 Bq/kg) and points where the concentration is much higher. This result shows that the contribution of global precipitations forms the "base" of radioactive contamination and is observed everywhere, whereas other factors act locally, on some points of the territory.

Atmospheric fallouts from surface and atmospheric nuclear explosions on the "Opytnoye pole". The distribution of radionuclides along the riverbed (Figure 6 – Figure 10) shows that the general tendency of decrease in the radionuclide concentration with increase of the distance from the "Atomic" lake has an irregular structure. In the direction along the riverbed, the areas with low concentrations of artificial radionuclides (further – ARN) alternate with areas with high concentrations. It makes it necessary to consider in detail the assumption of possible additional inflow of radionuclides to some parts of the riverbed from the traces of local fallouts. However, such assumption cannot explain the following results:

1. As it is seen from the figure (Figure 3), maximal ^{137}Cs concentrations in soils in the southeastern trace (up to 580 Bq/kg) were registered at a distance of 38-46 km from the "Opytnoye pole". Investigations of the trace influence were made up to the distance of 80 km, and at this distance ^{137}Cs concentration in soils is much lower – 20-80 Bq/kg. The studied segment of the Shagan River is even farther from "Opytnoye pole", at a distance of about 100 km, and it seems that the factor of trace influence is insufficient to explain ^{137}Cs concentrations in riverbank soils of the order of 180-250 Bq/kg;

2. Fallouts from local traces at rather large distances from the "Opytnoye pole" and on small areas must be rather uniform. Meanwhile, studies of radionuclide distribution across the river showed that profiles of several meters in length have very non-uniform variations in the ARN concentrations, the differences may be as high as the order of magnitude, which does not look like the result of fallout from the trace;
3. The southeastern trace was mainly formed by the fallouts from the 400-kton thermonuclear explosion made on 12.08.1953. [3]; it crosses the riverbed to the south from the "Atomic" lake and does not cross the segment. In the considered segment the riverbed has crossings with the traces of tests of 16.03.1956 and 25.09.1962, but according to the literature data [2] the tests did not refer to the category of radiation-significant tests;
4. Radionuclide composition of contamination of soils and bottom sediments does not correspond to the composition of local fallouts from the "Opytnoye pole", soils and bottom sediments as well as the soil of the crest around the "Atomic" lake are enriched by the products of neutron activation (^{60}Co, ^{152}Eu, ^{154}Eu). In the places of high ARN concentrations no variations in their composition and ratios of elements are registered, and it had to take place if the contamination were formed by the local traces from the "Opytnoye pole".

Atmospheric fallouts from the excavation explosion in the borehole 1004. There is no doubt that these atmospheric fallouts play a role at the initial river segment, but the analysis of soil samples beyond the crater shows that radionuclide concentration at distances of 1-2 km from the "Atomic" lake decreases to almost background values [3].

Radionuclide transfer with wind from the zone of soil heap around the "Atomic" lake. By its physical meaning this process can be considered as a reduced scale, delayed and extended in time process of atmospheric fallouts from the excavation explosion. It may cause short-time increases in the ARN concentrations in air, possibly, to the values hazardous for people, this problem will be considered further in the part describing the air basin. From the point of soil state this factor will hardly produce stronger effect than the above-mentioned local precipitations. High ARN concentrations in soils at a distance of several kilometers cannot be explained by this factor, which is confirmed by high nonuniformity on short segments that does not correspond to the character of wind transfer.

Radionuclide transfer by surface runoff from the zone of soil heap around the "Atomic" lake. Unlike the wind transfer, the ARN transfer by surface runoff can considerably influence the picture of radionuclide contamination of the territory at large distances from the source. It can be explained by the fact that wind transfer creates a two-dimensional areal contamination, so as the distance r from the source increases, the size (area) of influence increases proportionately to r^2. It is also the coefficient determining decrease in potential ARN concentrations caused by wind transfer. The water runoff transfer is in first approximation one-dimensional, and as the distance from the sources increases the size (length) of the influence zone increases to the first power of r. It means that potential concentrations of redeposited radionuclides decrease more slowly than in case of wind transfer. In the ideal case (impermeable and not trapping ARN part of the channel) there is a theoretical possibility of transfer of any amount of substance without losses at any unlimited distance with further sedimentation in any concentrations.

Radionuclides may be transferred by the water flow in different forms. Abstracting from the variety of concrete forms, it is necessary to single out two principally different cases: ARN in solution (real or colloid) and suspension of contaminated soil particles.

Transfer of dissolved radionuclides by surface runoff. Destruction and solution of contaminated mountain rocks and soils in the heap area, leaching of radionuclides and their transfer to solution are durable processes, whose intensity was probably minimal directly after the explosion and became higher with time. Therefore, this process can be observed now better than some years ago. However, the present-day samples from the Shagan River do not contain ^{137}Cs, which makes importance of the above factor doubtful. These doubts are confirmed by the following arguments. On the STS territory there are objects with streams flowing out from the areas of explosions, where tests were made in the same period as the tests in the borehole 1004 – these are water-bearing tunnels of the "Degelen" test ground . As tests in the tunnels were underground camouflet blasts, and the water flowing out from the galleries has to permeate through hermetic elements and sealing structures, therefore the waters cannot carry away radionuclides as a suspension of contaminated particles, they can carry away radionuclides only as a solution. The streams from tunnels are well studied; it is established that on the first kilometer of the riverbed ^{137}Cs concentration in soils decreases by the order of 3 as it is intensively absorbed by soils and therefore has low migration ability. The slope of the area on the near-portal segments of the "Degelen" ground is several degrees, which is tens of times higher than that in the plain conditions of the Shagan River. Hence, in the streams flowing down from the "Degelen" ground the flow speed is much higher, which helps farther transfer of radionuclides. If in the streams of the "Degelen" ground practically all ^{137}Cs sinks on the first kilometer of the riverbed, in the Shagan River it must remain on a much shorter initial segment.

It should be noted that a more mobile radionuclide ^{90}Sr was detected in the water samples from the Shagan River (see further in para. 2.2.2). Therefore, we can conclude that this factor plays a certain role and will play its role in future, which makes it an actual object of further studying. In particular, it is desirable to study forms of various radionuclides in water of the initial part of the Shagan River. However, nowadays we have no evidence to consider this factor as one of the main factors forming the picture of radionuclide contamination.

Radionuclide transfer by surface runoff as a suspension of contaminated particles. The processes of fragments' transfer and re-deposition occurs in any waterway downstream. In the segments with high slopes and high speed of flow only coarse-grain fractions – gravel and sand can deposit, on the slow segments small-grain silt and clay fractions deposit. In the segments where the stream disappears (dries up) all material present in water must deposit. The amount of material transferred by water is determined by the speed of its formation, i.e. rock weathering and destruction in the watershed area.

In January 1965, the excavation explosion in the borehole 1004 instantly formed huge masses of loose, transferable by water material – soil and rock particles. In April 1965, the crater was filled with water during spring flooding and fragments were carried downstream. In spring the Shagan River overflows and water fills the entire riverbed. As there was a huge amount of loose material on the way of river flow, the transfer intensity was very high, the water was probably turbid because of suspended particles. As the soil in the heap area was contaminated with ARN, the transfer of fragments meant transfer of radionuclides.

This mechanism of radionuclide transfer may play role of the main factor determining the picture of radionuclide contamination of the Shagan River valley. The variations of ARN concentrations along the riverbed presented in the figures (Figures 7-10) may be interpreted as follows:

1. General tendency to decrease in radionuclide concentrations in the riverbank soils and bottom sediments with increase in the distance from the "Atomic" lake is caused by decrease in the concentration of suspended material in water as some part of it was left on the previous segments of the riverbed;
2. Local rises and falls in the ARN concentrations can be explained by specific features of the waterway – variations in the relief slope and flow speed, which determine granulometry and amount of precipitable material;
3. Continuous distribution of radionuclides along the riverbed is explained by the fact that the initial moment of transfer, when it was the most intensive, took place in the spring flood period when there was water along all length of the riverbed. If the excavation explosion were made in summer, when at a distance of 12 km or 6 km from the "Atomic" lake (depending on the year) there was no water, the distribution of contamination would be different – in the initial segment soil could be contaminated more intensively but the riverbed would be clean downstream.

The variations in ARN concentrations across the riverbed (Figures 11–12), probably show the picture of water flow in the riverbed in 1965. Points with high radionuclide concentrations correspond to the places where water was most part of the season, and points with maximal concentration must correspond to the position of the riverbed axis at that moment. If these conclusions are correct, one can state that in the second profile (Figure 12) the position of the riverbed has not changed over the last 45 years and in the first profile (Figure 11) the main riverbed axis moved 10-15 m to the left.

A supposed mechanism of ARN spreading with surface water flow explains migration of radionuclides, which do not have high migration ability, at large distances. There is no contradiction between two statements as it is supposed that not ARNs but soil particles containing ARNs migrate.

Local redistribution of radionuclides in landscape due to surface washing-off and penetration into soil. The in-depth ARN distribution (Figure 13) shows that radionuclide concentrations quickly decrease with depth and are mainly concentrated in the upper layer (0–10 cm).

The ARN lateral migration due to surface washing-off has unfavorable conditions on most part of the territory – prevailing of evapouration over precipitation, flat relief, no waterways on the relief (besides the main waterway – the Shagan River). The lateral migration could be pronounced in local segments with steep slopes of the relief but it is unlikely that it could considerably change the picture.

An analysis of all collection of factors responsible for radionuclide contamination of soils and bottom sediments shows that the main factors are global atmospheric fallouts as a background and transfer of suspension of soil particles by the surface waterway as a "picture on the background". For some radionuclides with high migration ability (^{90}Sr and, probably, ^{241}Am) transfer in the dissolved state with surface water flow also gives some contribution, this factor will be considered in more detail in the section devoted to radionuclide contamination of surface waters (paragraph 2.2.2).

2.2. Surface waters

In studying composition of the Shagan River waters, the sampling points were differentiated depending on the point distance from the place where the river flowed out of the outer water reservoir of the "Atomic" lake.

In the summer period not all riverbed of the Shagan River is filled with water. In the period of investigations, in 2009, the water was only present in the following parts of the river: at a distance of 500-800 m from the "Atomic" lake there were water streams of 20-30 m long and 1-3 m wide; at a distance of 2.5 – 6 km from the "Atomic" lake there was a 2-3m-wide stream along all the riverbed; at a distance of 9 km from the "Atomic" lake there was a stream 1000 m long, 11 m wide and 0.8 m deep.

In the summer period, on the area of monitoring in the points located at a distance of 500 m and 4.6 km away the "Atomic" lake the average values of the riverbed water flow rate ranged from 13 to 136 l/min (Figure 14).

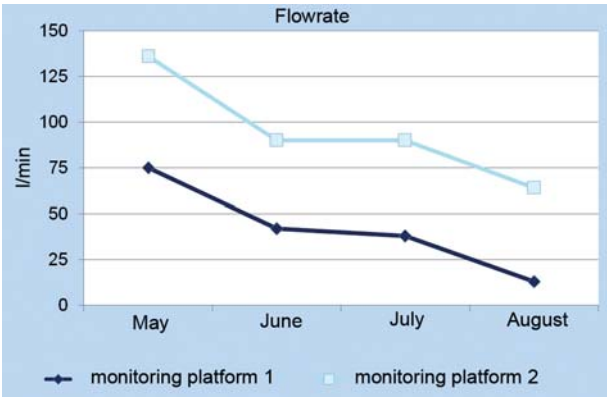


Figure 14. Variations in the flow rate of the Shagan River waters

No surface water flow is observed in the other parts of the riverbed. The area along the riverbed in the sector from the "Atomic" lake to a distance of 12 km is covered with rich vegetation, which indicates presence of ground waters close to the ground surface.

The figure (Figure 15) shows dried-up riverbed of the Shagan River.

Figure 15. Dried-up riverbed of the Shagan River

2.2.1. ^{3}H content in the Shagan River

The 2006–2009 investigations detected and repeatedly confirmed the presence of high ^{3}H concentrations in the Shagan River waters. In 2006, in order to study the ^{3}H distribution in the Shagan River, water samples were taken from the "Atomic" lake downstream to its flowing into the Irtysh. In 2006-2007, in the framework of the preliminary examination, ^{3}H concentration was determined in all surface water samples taken in the river part ranging from the "Atomic" lake to 40 km downstream. The results of investigation of ^{3}H distribution in surface waters are shown in the figure (Figure 16) (the area of sharp increase in ^{3}H specific activity is marked).

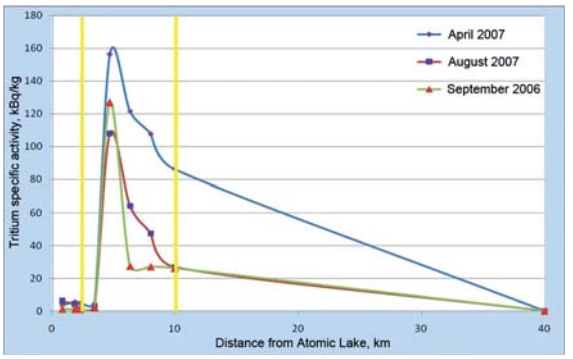


Figure 16. Distribution of ^{3}H specific activity in the Shagan River waters in 2006-2007

^{3}H concentration in the river surface waters along all distance varied from 10 to $1.6*10^{5}$ Bq/kg. Maximal values were registered at a distance of 4.7 km from the "Atomic" lake, minimal values were registered in the point of confluence of the Shagan and Irtysh rivers. In the section from 12 to 38 km water samples were not taken as there was no surface water.

In 2008 the section of the riverbed from the "Atomic" lake to a distance of 6 km was studied in more detail, in 2009 the section up to 9 km was studied. The water was sampled for ^{3}H concentration along the riverbed with interval of 100 m. The figure (Figure 17) shows the distribution of ^{3}H specific activity along the studied part of the riverbed downstream to a distance of 12 km from the "Atomic" lake (the area of sharp variations in the ^{3}H specific activity is marked).

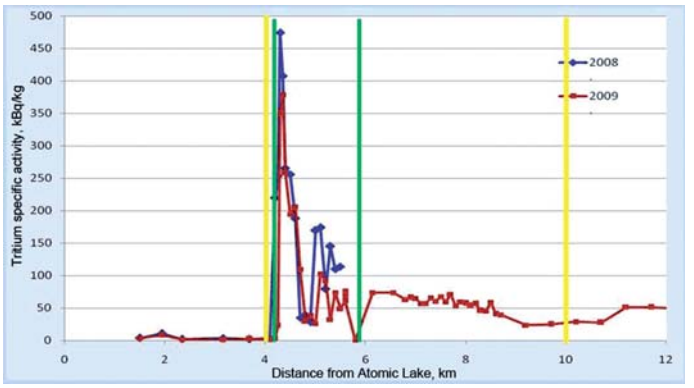


Figure 17. ^{3}H specific activity in the Shagan riverbed

^{3}H specific activity in water varies in a wide range. The highest ^{3}H activity is registered in the interval from 4 to 6 km from the "Atomic" lake, therefore more detailed studies of ^{3}H specific activity were carried out in that interval. Samples of underflow waters were taken, and distance between sampling points was 10 m (Figure 18).

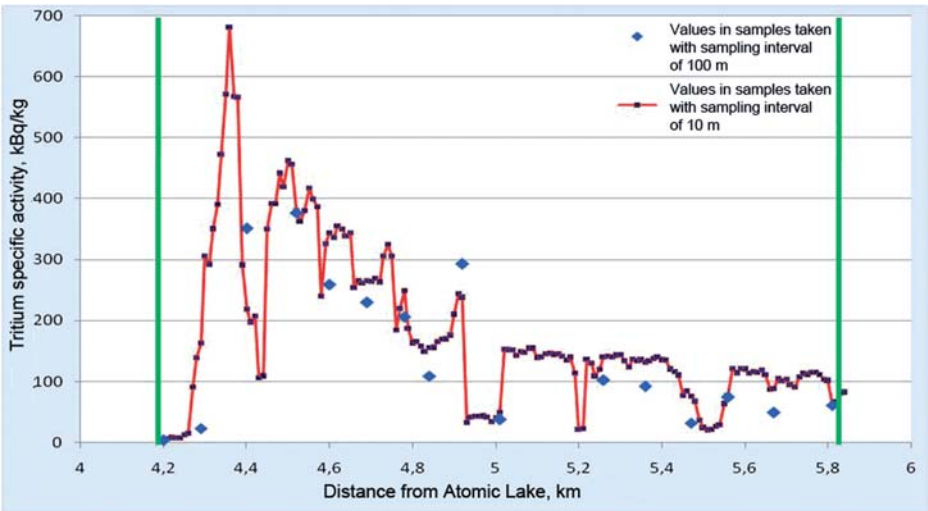


Figure 18. ^{3}H specific activity in the Shagan riverbed in the section 4-6 km from the "Atomic" lake

The investigations detected maximum of ^{3}H specific activity, 680 kBq/kg, in the point 4.4 km from the "Atomic" lake.

^{3}H specific activity in the studied section varies step-wise with pronounced maximum and minimum points. Such distribution may be a result of a complicated system of water exchange between subsoil and riverbed waters with several points of inflow of subsoil waters. As a whole, the specific activity decreases to the point of 6 km. In that point ^{3}H concentration in water is an order of magnitude lower that the maximal concentration but it still remains very high, several times above the intervention level according to NRB-99, equal to 7700 Bq/kg. The sections with sharp decrease in ^{3}H specific activity indicate probable presence of either area of subsoil water discharge or inflow of subsoil waters not contaminated with ^{3}H. The increase in ^{3}H specific activity following minimal values (particularly, at the 6-th km from the "Atomic" lake) also indicates ^{3}H migration with subsoil waters going out to the surface along the riverbed.

2.2.2. Concentration of ^{90}Sr and ^{241}Am radionuclides in the Shagan River water

In 2006-2008, along all the riverbed samples were taken, and ^{90}Sr specific activity was determined. In order to get preliminary assessment of the level of riverbed contamination the samples for determining strontium specific activity were taken with an interval of 10 m downstream to the place where the river inflows into the Irtysh. In 5 water samples the concentration of ^{241}Am was determined. The values of specific activity of ^{90}Sr and ^{241}Am are given in the figure (Figure 19).

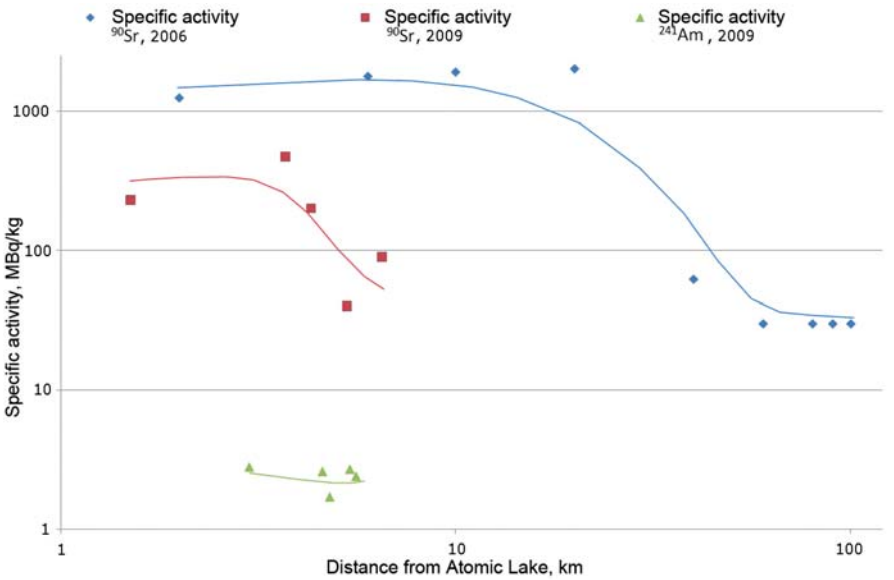


Figure 19. Specific activities of ^{90}Sr (2006, 2009) and ^{241}Am (2009).

^{90}Sr specific activity varied from 30 mBq/kg to 2000 mBq/kg. Maximal values of ^{90}Sr specific activity were registered in the interval 2 – 20 km from the "Atomic" lake. However, observations in 2009 showed lower ^{90}Sr concentrations in that section. A more detailed study of ^{90}Sr concentration in the Shagan waters in 2009 was impossible due to the absence of sufficient for analysis amount of water in the river. It is necessary to carry out further studies of strontium contamination of the riverbed.

The investigations showed the presence of ^{241}Am isotope in all water samples. Specific activity of ^{241}Am is on average 2.5 mBq/kg. The presence of ^{241}Am indicates its ability to migrate with subsoil and surface waters at large distances from the sources of contamination. ^{241}Am can penetrate into water by washing out from bottom sediments which contain a considerable amount of this radionuclide, however, it is not impossible that ^{241}Am is brought to the riverbed with subsoil waters. The presence of ^{241}Am in the riverbed requires further investigations in order to predict spreading of contamination with transuranium radionuclides.

2.2.3. Concentration of radionuclides ^{137}Cs, ^{152}Eu, $^{239+240}$Pu in the Shagan River water

^{137}Cs concentration in the studied section of the river is below the registration level, 30 mBq/kg, of measurement devices. The specific activity of $^{239+240}$Pu in the studied section is also below the registration level of measurement devices equal to 5 mBq/kg. ^{152}Eu concentration was measured in 5 samples. In most tested samples ^{152}Eu specific activity was below the registration level of measurement devices – 0.8 mBq/kg. In two sampling points (3 km and 5.6 km from the "Atomic" lake) ^{152}Eu specific activity was 1.8 mBq/kg and 2.8 mBq/kg.

2.2.4. Concentration of macro- and micro-components in the Shagan River waters

The samples of the Shagan River waters were analyzed not only for detecting radionuclides but also for determining cation-anion composition, mineralization and presence of microelements.

The chemical composition of the river water was determined to a distance of 7 km downstream from the point 1.5 km away the "Atomic" lake. The sampling interval was 100 m. The variations in the mineralization level are shown in the figure (Figure 20).

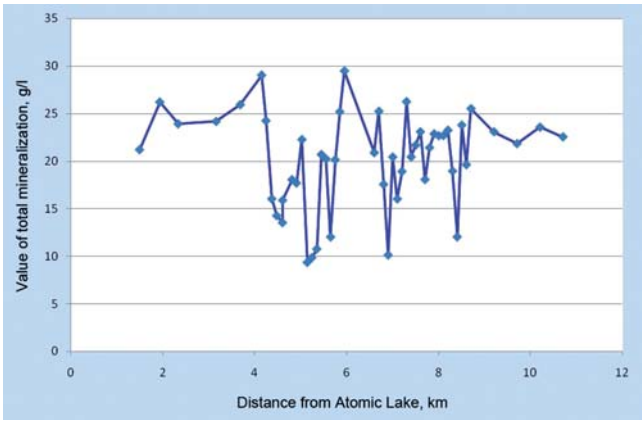


Figure 20. General mineralization of the Shagan River waters

The Shagan River waters along all studied section have high salt content. Water mineralization ranges from 10 g/l to 30 g/l. According to V.I. Vernadsky's classification the waters refer to salt waters (10-50 g/l).

The character of variations of the main macro-elements corresponds to the changes in general mineralization (Figure 21).

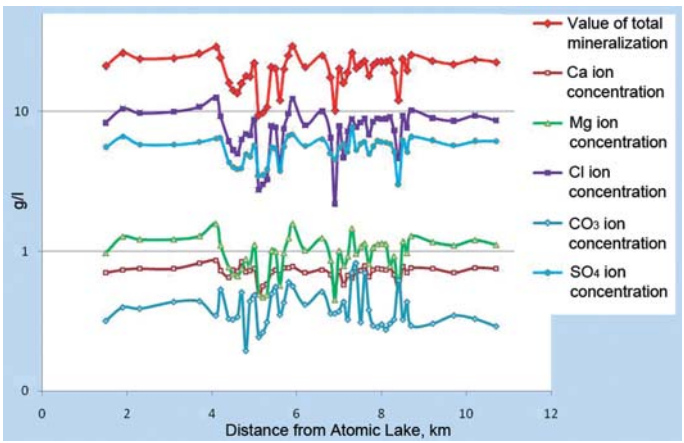


Figure 21. Concentration of the main macro-elements in the Shagan River waters along the riverbed

Concentration of the main macro-elements in the river waters changes with changes in general mineralization. According to the cation-anion composition the waters are subdivided into chloride-sulphate and sodium-magnesium.

High magnesium concentrations are observed along the entire riverbed. The ratio of calcium and magnesium ions is on average 1:1.5, which is not typical of natural waters. This phenomenon may have a regional character as in the subsoil waters of the test ground "Balapan" concentration of magnesium ions is also much higher than that of calcium ions. Variations in the ratio Mg/Ca along the Shagan River are shown in the figure (Figure 22).

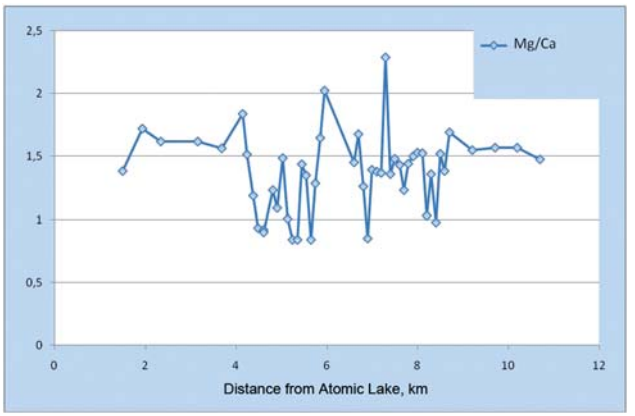


Figure 22. Ratio of magnesium and calcium ions concentrations in the Shagan River waters

The other factor studied in the Shagan River waters was their microelement composition. To determine the microelement composition 67 water samples were taken and concentrations of 18 microelements: Al, Cr, Ni, Cu, Zn, Cd, Ba, Pb, Co, Mn, U, Fe, Hg, Be, V, Se, As, Sr Se were measured. Concentrations of Se, As, Sr, Hg, Cs, Be, Cd are below the registration level of measuring devices $C_{(Se, Sr, Hg, Cs, Be, Cd)}$<0.5 µg/l, C_{As}<5 µg/l. The results for other 11 microelements are presented in the table (Table 2).

Table 2.

Concentration of micro-components in the Shagan River waters, µg/l

Microelement	Minimal value	Average value	Maximal value	Average content in nature	MPC for drinking water
Al	< 5	42.4	593	100-300	500
Cr	<1	3.75	83	n*10	500 (Cr^{3+}). 50 (Cr^{6+})
Ni	17.8	23.7	32.8	2-10	100
Cu	11.5	15.1	22.9	5-25	1000
Zn	<1	4.7	27.8	1-10	5000 (Zn^{2+})
Ba	7.0	15.4	28.4	~ 15	100
Co	<1	1.8	9.3	1-10	100
Mn	1.7	363	2500	~ 100	100
Fe	25.0	132	1430	~ 500	300
V	2.6	21.7	50	1-5	100

Sections with high concentration of some elements are detected along all the riverbed. In some sections concentrations of iron and magnesium many times exceed average values for natural waters [1] (Figures 23–24). Variations in concentrations of some elements are shown in the figure (Figure 25) (a dotted line shows the range of average concentrations for natural waters).

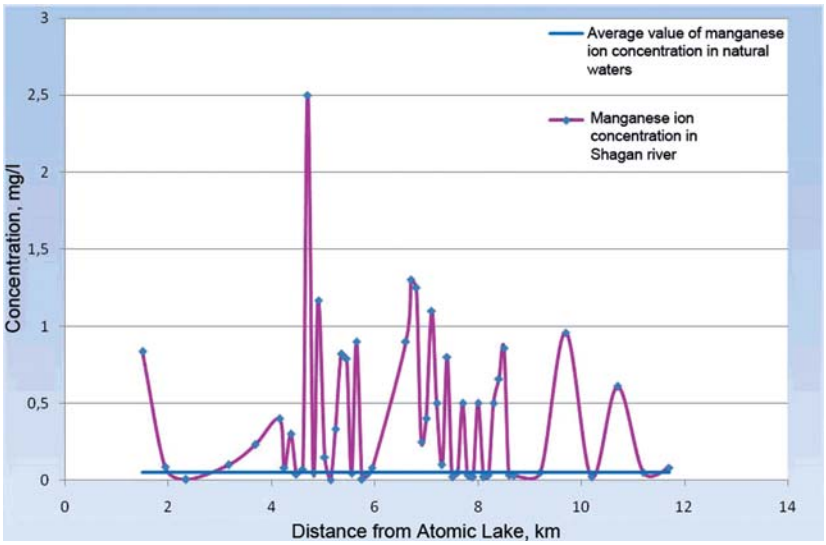


Figure 23. Magnesium concentration in the Shagan River waters

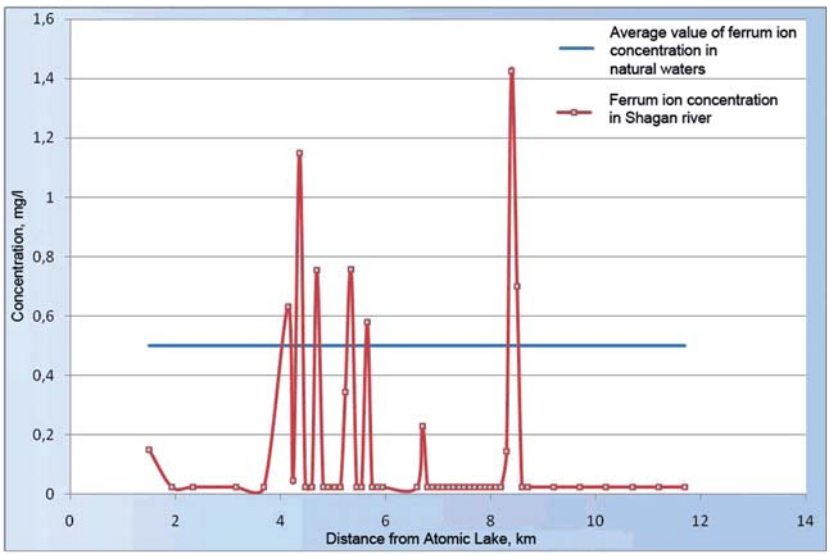


Figure 24. Iron concentration in the Shagan River waters

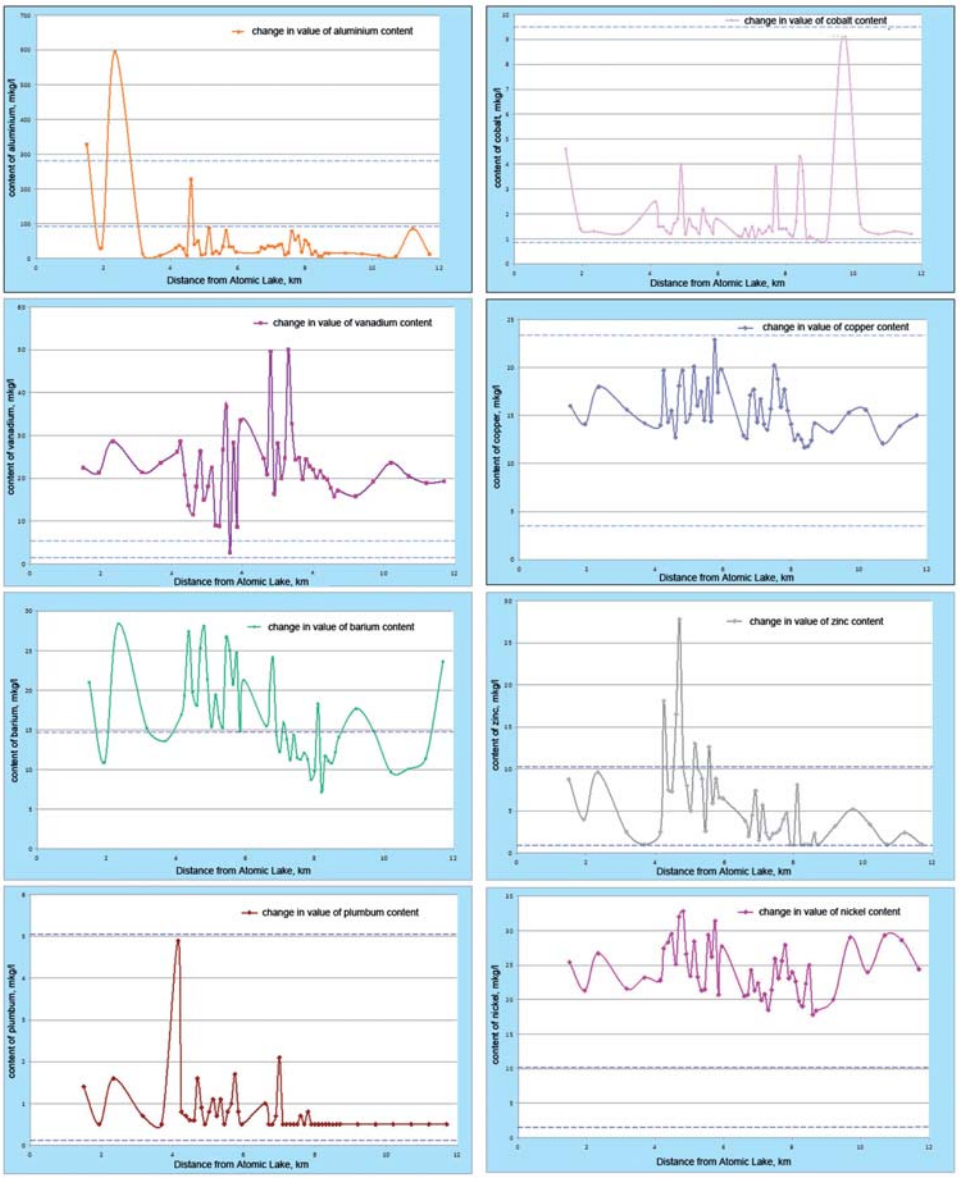


Figure 25. Concentration of microelements in the Shagan River waters

Of special attention is uranium concentration the average concentration of which in the river waters is about 100 μg/kg (Figure 26).

In units of specific activity of natural mixture of isotopes (^{238}U – 99.284%, ^{235}U – 0.711%, ^{234}U – 0.005 %) such concentration corresponds to 2.5 Bq/kg. Therefore the presence of uranium in the river water makes a considerable contribution to the radiation background of the Shagan River.

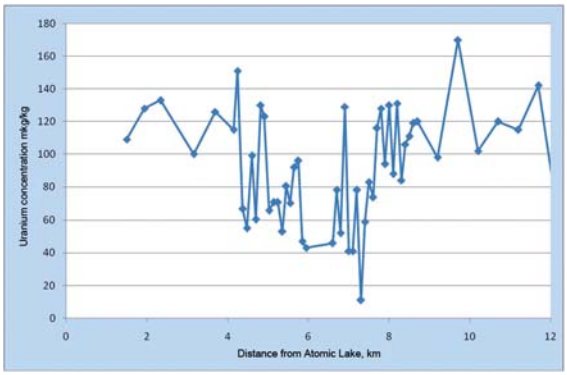


Figure 26. Uranium concentration in the Shagan River waters

It can be stated that the Shagan River has a specific multi-component "portrait". A typical feature of the river is high concentration of such elements as magnesium, vanadium, barium, nickel considerably exceeding their average concentrations in natural water and aluminum concentration much lower than that in natural waters.

As a whole, the concentration of microelements in the Shagan River waters does not vary uniformly. The points with maximal concentrations of some microelements coincide with the points of maximal specific activity of ^{3}H, which indicates that they came to the river from the same source (Figure 27).

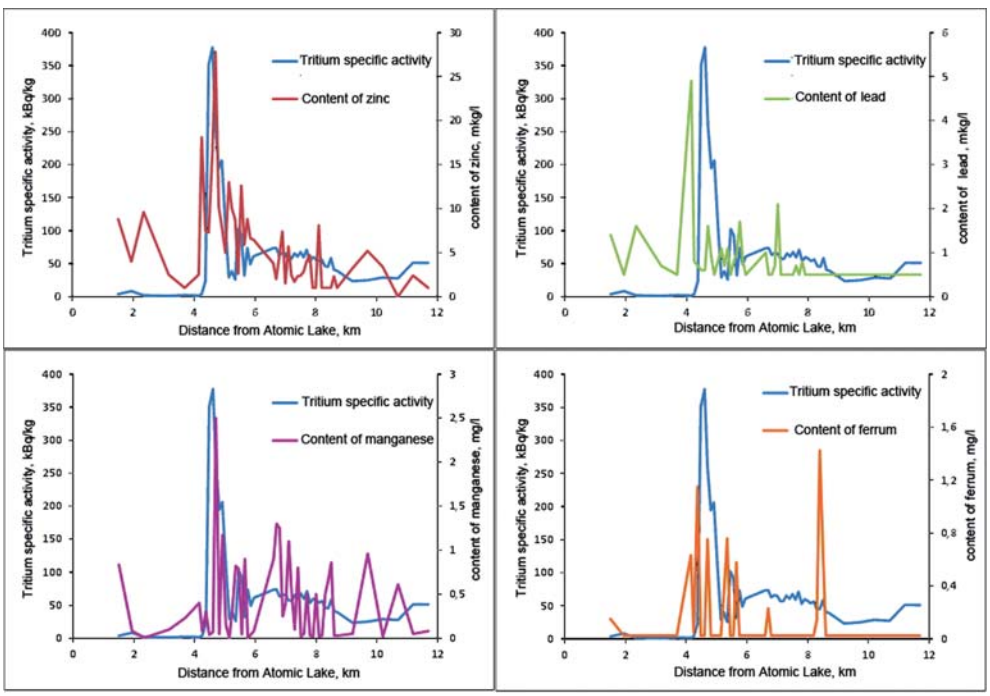


Figure 27. Variations in concentrations of ^{3}H and some microelements in the Shagan River waters

2.2.5. Mechanism responsible for radionuclide contamination of the Shagan River surface waters

The most probable way of radionuclide contamination of the Shagan River waters is ARN washing out from riverbank soils and bottom sediments as well as inflow with ground waters. The main sector of groundwater discharge characterized by maximal values of ^{3}H specific activity is supposedly located at a distance of 4 – 5 km from the "Atomic" lake. This sector has a minimum value of general water mineralization. It should be noted that sharp changes in mineralization and ^{3}H specific activity along the riverbed are indicative of a complicated water exchange system between subsoil and surface waters (Figure 28).

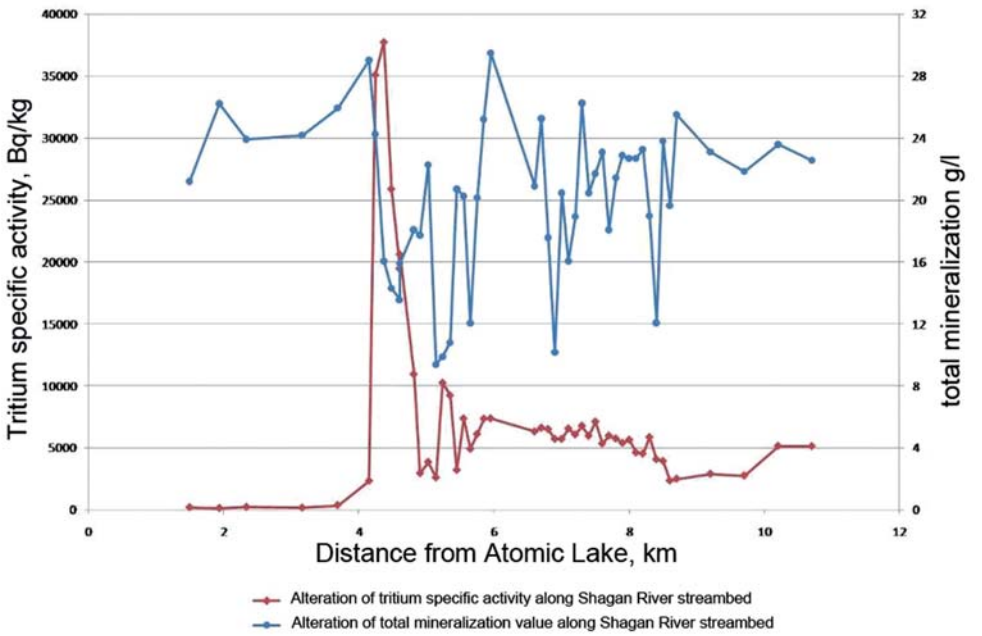


Figure 28. Variations in concentrations of ^{3}H and some microelements in the Shagan River waters

Variations in concentrations of the main microelements in the river waters correspond to the variations in general mineralization and confirm the presence of several zones of ground water discharge in the riverbed. The points with minimal values of general mineralization are detected along all studied part of the river (to 9 km downstream from the "Atomic" lake), which confirms the assumption about the existence of several zones of ground water discharge.

A detailed examination of the riverbed at a distance of 4 – 6 km from the "Atomic" lake revealed clear correlation between ^{3}H concentration and mineralization – sometimes it is positive, sometimes – negative but well expressed in both cases. Projections of variations of ^{3}H specific activity and general mineralization at a distance of 4-6 km from the "Atomic" lake are given in the figure (Figure 29).

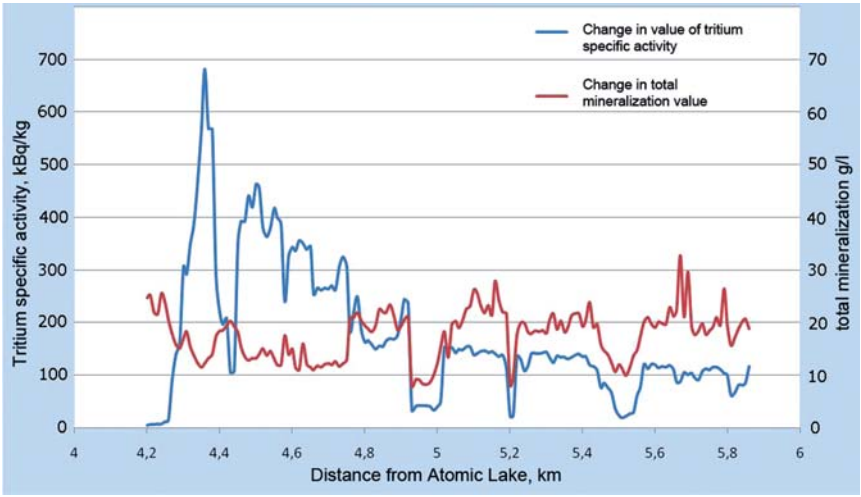


Figure 29. Variations in ^{3}H concentration and general mineralization on the 4-6 km from the "Atomic" lake

It may be assumed that there are several sources of ground waters inflow into the riverbed, there are at least three:

1. fresh waters, not containing ^{3}H, formed by permeation of atmospheric fallouts from the surface;
2. waters with interim mineralization (~10 g/l), hence, waters coming from depths of several hundreds of meters, they contain ^{3}H and are likely to flow along the fracture zones passing near "warfare" boreholes;
3. highly mineralized waters coming from even bigger depths. They do not contain ^{3}H; they are likely to flow along the other system of fractures.

It is likely that in point 4.2 km the waters of the second type inflow as mineralization and ^{3}H concentration increase step-wise.

The presence of sections where mineralization is much lower than the average along the riverbed indicates discharge of subsoil waters with lower mineralization. In particular, the results of observations showed that the subsoil waters from the test ground "Balapan" have average mineralization of about 10 g/l, so their inflow in the Shagan riverbed may cause the effect of sharp decrease in mineralization. It is quite probable taking into account the fact that general mineralization as well as ions concentration in the Shagan River is rather high and in some parts is as high as 30 g/l. Such salt concentrations are not typical of river waters. Most rivers have mineralization ranging in the interval 0.5-0.9 g/l.

The distribution of microelement concentrations enables to obtain a more complete picture of the distribution of tritium contamination along the riverbed. The distribution of micro-components of chemical composition in the Shagan River is indicative of the existence of a complicated water exchange system along all studied part of the riverbed. It is quite likely that different parts of the riverbed experience discharge of ground waters of different chemical composition; it is also not unlikely that the river has sections with outflow of riverbed waters, which gives sharp decrease in the ^{3}H activity and concentrations of macro- and microelements

in some points. One cannot exclude a possibility that sharp decrease in concentrations of microelements may be caused by chemical processes. For example, waters with high oxygen concentration can oxidize ferrous minerals. As a result, ferrous iron converts into ferric iron forming oxides and hydroxides. Oxygen-free waters often contain reduced iron. If on the ways of their migration they meet oxygen-containing waters or air oxygen, an oxygen barrier is created where iron, manganese, sulphur and other elements can concentrate.

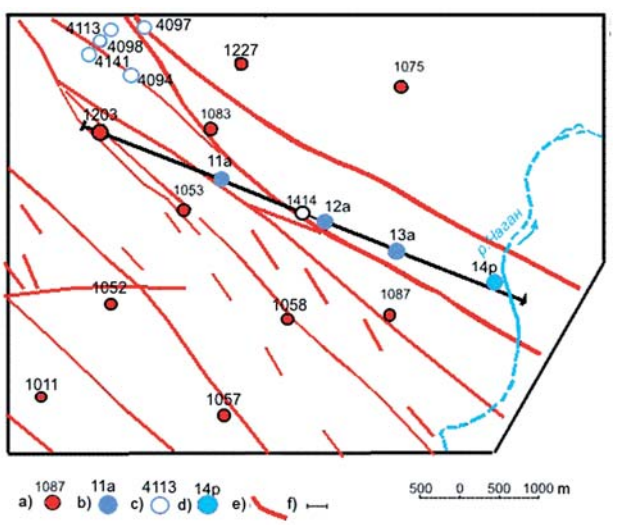
In order to give prognosis of spreading of radionuclide contamination it is necessary to carry out further investigation of the Shagan River along all its riverbed including the section from 12 to 38 km, which is insufficiently studied because it does not have water most part of the season.

2.2.6. Additional investigations: search for a channel discharging ^{3}H contaminated ground waters into the Shagan River

The most important feature of the aquifer of the test ground "Balapan" is its location under Neogene waterproof clays on most part of the area. The clay layer is not uniform in thickness as the erosion surface of Paleozoic bedding has a very complicated configuration. Maximal thickness of the waterproof clays is 70 m.

The studied part of the aquifer has a hollow with the centre in the place of the "warfare" borehole 1414 (Figure 34a). In 1969-1974, 7 underground nuclear explosions were made in that area, and the hollow may be a "storehouse" for preserved artificial radionuclides. The flood-plain of the Shagan River has some areas without Neogene waterproof clays. In those areas fracture waters directly contact with surface waters and may be sources of radioactive contamination of the Shagan River.

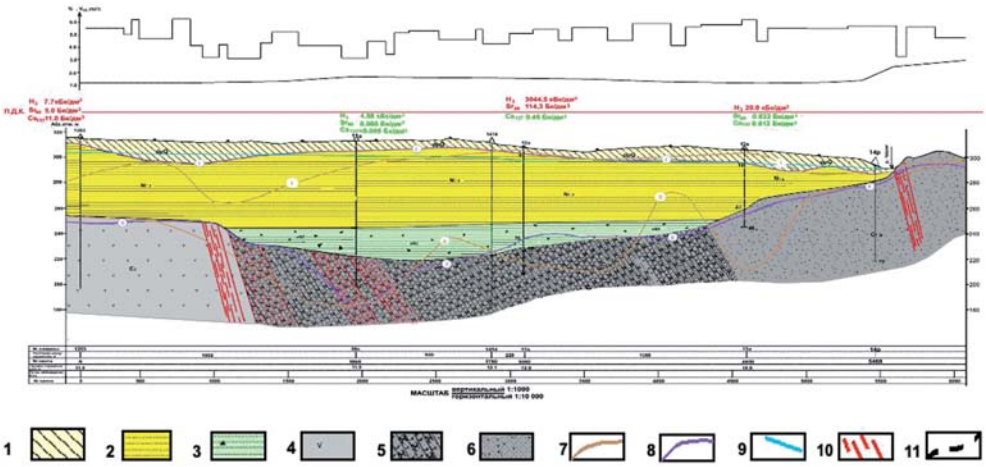
In the vicinity of the borehole, in the Paleozoic bedding electric and seismic prospecting profiles were made, and several hydrological boreholes were drilled (Figure 30).



a-d: boreholes (a – warfare, b, c – hydrogeological, d – operating);
e – tectonic dislocations; i – line of geological-geophysical cross-section

Figure 30. Geological-geophysical prospecting in the vicinity of potential contamination source

The obtained data were used to construct the geological-geophysical section showing the geological situation in the studied part of the "Balapan" test ground (Figure 31).



1 – Quaternary sediments, 2 – Neogene clays, 3 – Mesozoic weathering crust, 4 – black coal andesites, 5 black coal and silt sandstones, 6 – black coal tuff-sandstones, 7 – resistance boundaries: I – surface of Quaternary sediments (30-600 Ohm·m), II – boundaries between Neogene clays (2-12 Ohm·m) and roof of formation (over 1000 Ohm·m)); 8 – boundaries for speed characteristics: I – surface of Quaternary sediments (up to 1 km/s), II – roof of formation (over 2.5 km/s), 9 – subsoil water level; 10 – zones of tectonic fracturing; 11 – supposed surface of exogenous fracturing zone. In the upper part of the figure boundary and average speeds (km/s) are given.

Figure 31. Geological-geophysical prospecting in the vicinity of potential contamination source

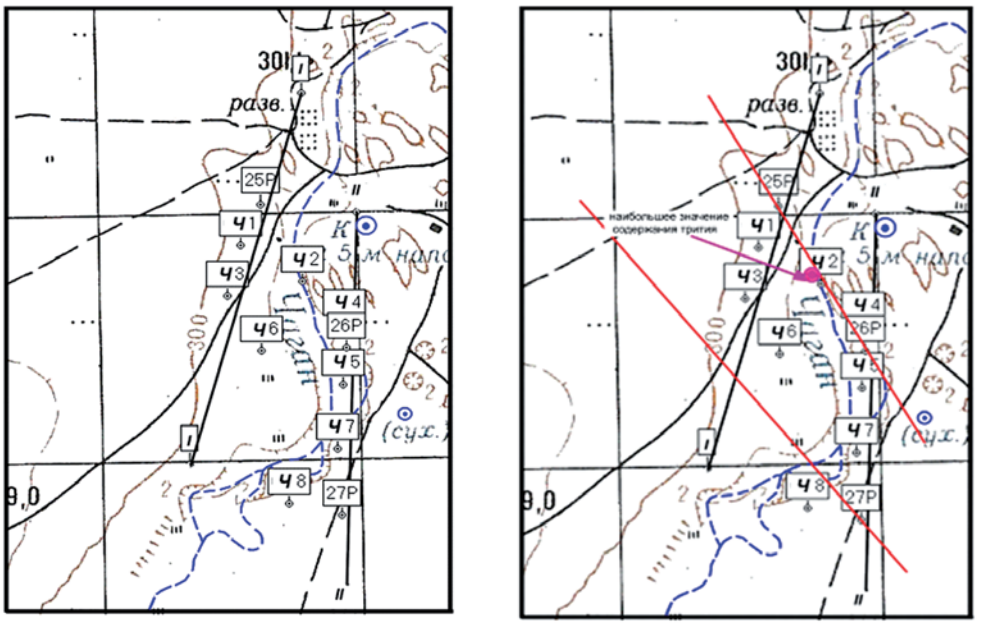
As feathering-out of Neogene clays in the Shagan River valley is confirmed, and waters in the valley are pressure waters with piezometric level inclined to the Shagan River, it is quite likely that radionuclide contaminated waters can permeate from the above-mentioned hollow to the underflow and surface waters of the Shagan River. The water samples taken from the drilled boreholes confirm this version (Table 3). In the borehole 12a ^{3}H concentration exceeds 3 MBq/kg, which is several times higher than its concentration in the Shagan River water, and one can state that this borehole crosses the channel of radionuclide migration. However, further the traces of migration channel are lost: in the next borehole – 13p – ^{3}H concentration falls by two orders of magnitude and is much lower than its concentration in the Shagan River. Hence, if the supposed channel exists in that place, its direction does not coincide with that of the geological-geographical section.

Table 3.

Results of borehole testing

Borehole	Flow rate, m^3/day	Water permeability, m^2/day	Aquifer thickness, m	Filtration coefficient, m/day	^{3}H, kBq/kg	^{90}Sr, Bq/kg	^{137}Cs, Bq/kg
11a	86.40	3.00	50	0.060	4.98 ± 0.04	0.008 ± 0.003	< 0.005
12a	43.20	3.40	40	0.085	3045 ± 1	114 ± 6	0.46 ± 0.11
13a	3.12	1.96	40	0.049	20.0 ± 0.1	32 ± 13	0.012 ± 0.004

Besides the area of an expected source, geological-geophysical prospecting was made directly in the vicinity of the Shagan River (Figure 32). Measurements with refracted and curved waves (prospecting depth up to 50-100 m) and vertical electrical sounding (prospecting depth up to 70 m) were made along two 3km-long profiles I (along the left bank) and II (along the right bank) [1, 2, 3].



a b

Figure 32. Geological-geophysical prospecting in the area of ^{3}H inflow in the Shagan River:
a – scheme of profiles (I, II) with boreholes drilled in 2009;
b – scheme of tectonic dislocations according to the data of seismic exploration (red lines)

The results of prospecting were used to construct engineering geological sections (Figure 33).

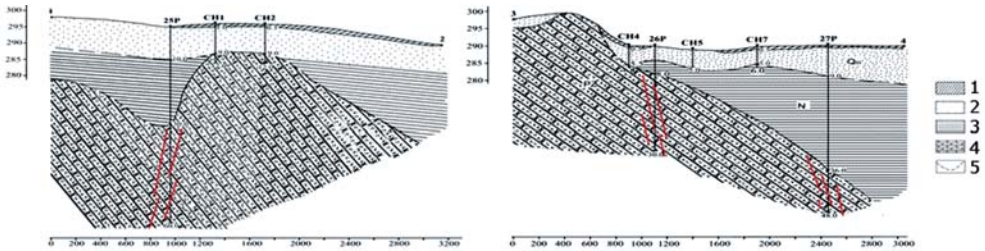


Figure 33. Engineering geological sections in the area of ^{3}H inflow in the Shagan riverbed:
a – I -I (left bank); b– II - II (right bank), 1 - loam; 2 - sands; 3 - clay; 4 – tuff-sandstone;
5 – boundary of Neogene clays according to the data of seismic prospecting

The results of experimental-filtration works in the boreholes 25P, 26P and 27P enabled to determine in more detail the relief of Paleozoic bedding within the three-kilometer section of the Shagan riverbed and to construct a more precise map of confining bed isohypses (Figure 34b).

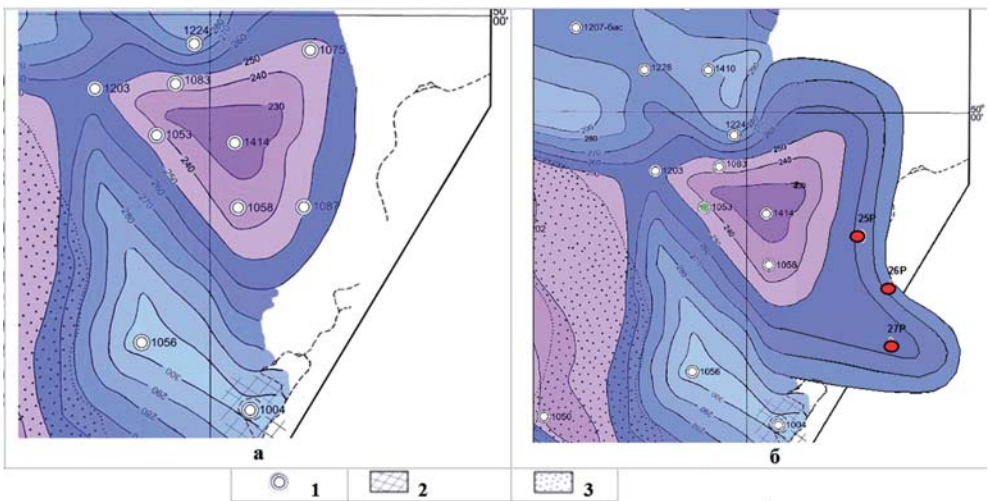


Figure 34. "Balapan" test ground. Fragment of the map of confining bed isohypses:
a – the results of 2007; b is a fragment with additional data for 2009.
1 – "warfare" boreholes; 2 is the area without clays (potential drainage area);
3 – water-saturated sands in the clay bedding.

In the water samples taken from the drilled boreholes high ^{3}H concentrations (up to 140 kBq/kg) comparable with concentration in the Shagan River were registered only in the second profile, beyond the river, on the other side from the source. In the first profile, on the side of the source, ^{3}H concentrations are not higher than 0.8 kBq/kg. It indicates that either the channel of radionuclide inflow is located in the other place (most probably it is located to the south from the areas of drilling) or it has very non-uniform filtration properties and ^{3}H concentrations, and boreholes were drilled in the places with low permeability.

Of special interest are the results of analysis of subsoil waters sampled in the boreholes drilled in one of the sections (length ~60 m) of potential discharge of subsoil waters with high ^{3}H concentration to the Shagan River, 4.3 km downstream from the "Atomic" lake. In the territory of the "old" riverbed located a little higher than the present-day riverbed, 2 profiles (5 points in each profile) parallel to the riverbed at a distance of 10 m from each other were made. The task of the profiles was to sample vegetation, air and subsoil waters (Figure 35).

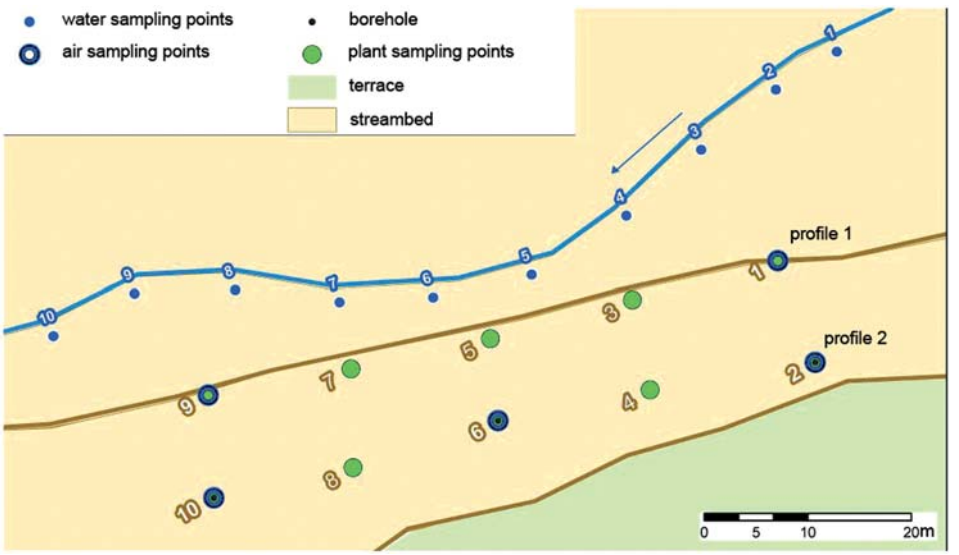


Figure 35. Scheme of profile location

In the boreholes C-2, C-6 and C-10 drilled in the second profile tritium concentrations in the subsoil waters were determined, the results were 14, 165 and 420 kBq/kg, respectively. The concentrations were higher than ^{3}H concentrations in the first profile (closer to the present-day riverbed), which enables to assume the existence of a channel through which tritium contaminated ground waters get into the Shagan River in that place.

An additional data on the section hydrology were obtained with radioactive indicator ^{131}I. The indicator was injected in the stream and its level was detected at the observation stations downstream. The results of such experiments enable to assess water losses or water inflow to the riverbed and to localize such points.

In 2009, a pilot experiment was made on the Shagan River. The starting station was located 4.7 km downstream from the "Atomic" lake, the first observation station was 68m and the second one was 128 m downstream.

At the starting station 0.9 MBq of indicator ^{131}I were injected in the stream. The indicator was detected at the first observation station 40 minutes after injection, it was being registered for 2 hours and 10 min, and about 60% of injected indicator passed through the station. The indicator was detected at the second station 1 hour 40 minutes after injection, its presence in water was registered for 3.50 hours, and about 15% of the injected indicator passed through the second station.

The specific activity of ^{131}I in water samples was determined by gamma-spectroscopic measurements. The results of measurements gave the indicator curves shown in the figure (Figure 36).

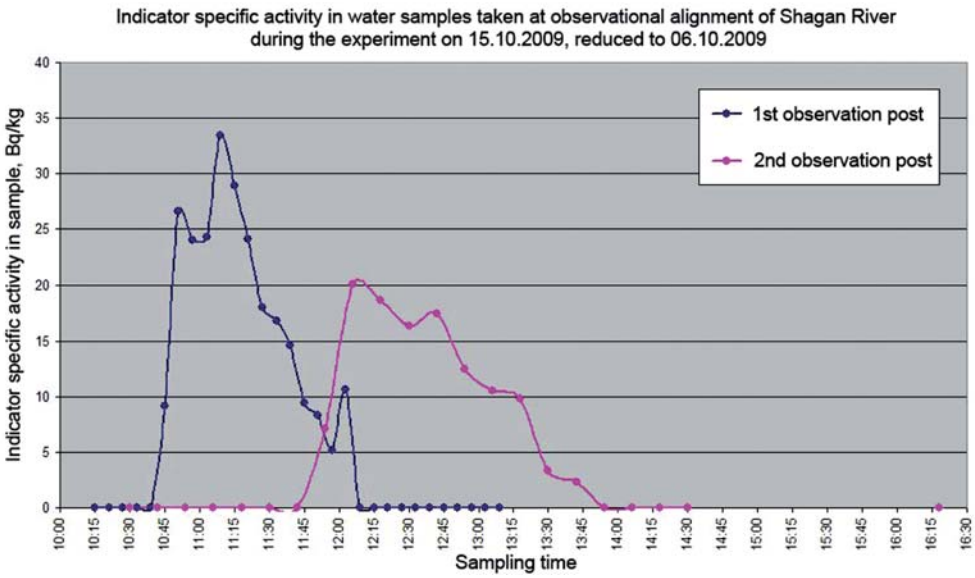


Figure 36. Resulting curves of the indicator experiment in the Shagan River

The data of the experiment enabled to obtain the main hydrological characteristics of the waterway. The results of calculation are presented in the table (Table 4).

Table 4.

Results of indicator experiment in the Shagan River

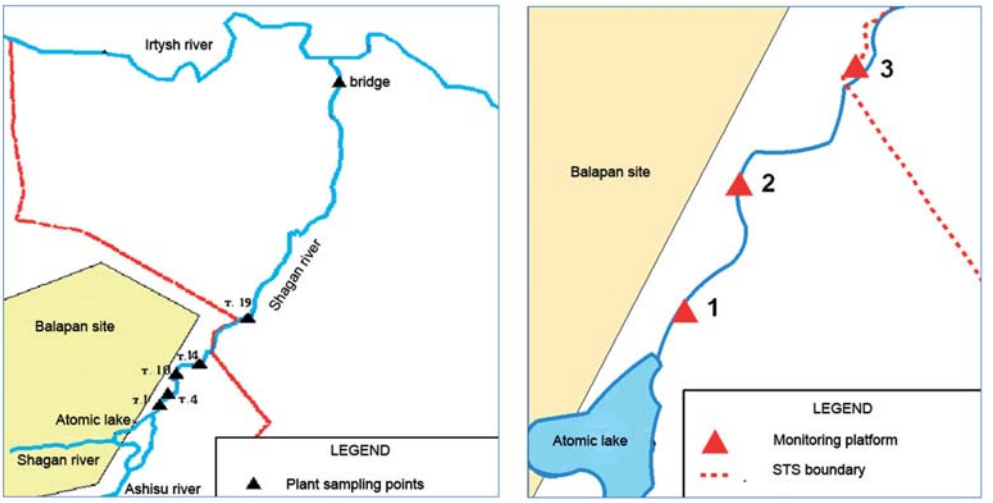
Section	Amount of indicator passed, MBq	Indicator front propagation rate, m/min	Water loss in the riverbed, l/min	Water inflow to the riverbed, l/min
Origin	0.90	-	-	-
1st observational	0.55	1.74	127	163
2nd observational	0.14	1.25	270	6

The results of the experiment show that the second section only has losses from the riverbed, and hence, the channel of ^{3}H migration cannot be located there. The first section has both water inflow and losses, so one cannot exclude the presence of a migration channel in the section. Further development of the above method and accumulation of experimental material will enable to examine all initial part of the Shagan River, to discover points of water inflow, and to consider each of such points as a potential place where ^{3}H can inflow into the river. There is no doubt that it must be done in cooperation with other works (sampling, geophysical investigations, etc.).

2.3. Vegetation

2.3.1. State of vegetation cover

To study parameters and dynamics of ^{3}H concentration in vegetation along the Shagan riverbed, 6 monitoring stations were organized, each of which has an area of ~ 100 m^2 including part of the Shagan riverbed, bench and territory of adjacent terrace (Figure 37a).



a b

Figure 37. Scheme of location: a) points of plant sampling (2007–2008), b) monitoring stations

In 2007–2008, vegetation samples were taken regularly in each monitoring ground. As experimental plants the following species were chosen: reed (*Phragmites australis*) growing in water, wild rye (*Leymus angustus*), spread in the territory of the riverbed and sometimes on the bench and cheegrass (*Achnaterum splendens*) growing on the terrace. ^{3}H concentration was determined in the free water of the top part of plants extracted during sublimation (t up to 200 °C). The results of analysis are presented in the histogram (Figure 38).

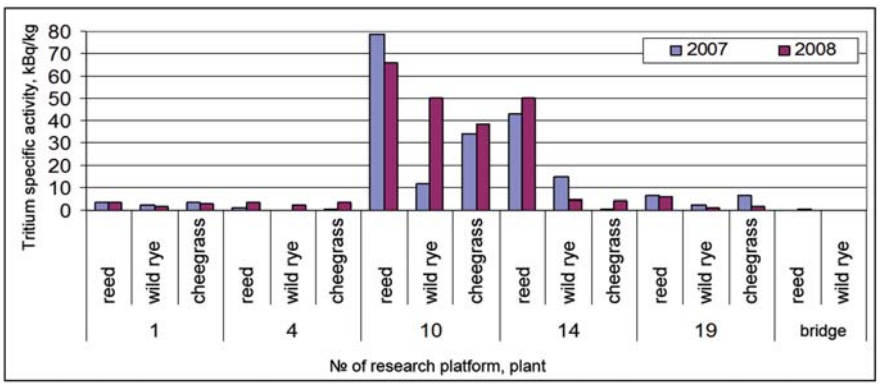


Figure 38. ^{3}H concentration in the free water in vegetation (2007–2008)

Based on the above results 3 monitoring grounds were chosen to further study dynamics of ^{3}H concentration in vegetation (Figure 37, b). Every month from May to August, samples of studied species were taken on the left riverbank (monitoring grounds Nos. 1 and 2) and right riverbank (monitoring ground No.3). Altogether 36 samples were taken (4 times 3 samples from each ground). The results of ^{3}H analysis in surface and underflow waters of monitoring grounds and in the studied plant species are presented in the (Table 4).

Table 5.

^{3}H specific activity in surface and underflow waters of monitoring stations and in free water of studied plant species

Sampling month	Monitoring site	Specific activity ^{3}H, Bq/kg				
		plants			water	
		reed	wild rye	cheegrass	surface	Sub-riverbed
May	1	$(5.5\pm0.1)*10^{3}$	$(2.5\pm0.1)*10^{3}$	$(0.7\pm0.1)*10^{3}$	$(7.7\pm0.1)*10^{3}$	$(7.8\pm0.1)*10^{3}$
	2	$(1.0\pm0.1)*10^{5}$	$(0.6\pm0.1)*10^{5}$	$(0.6\pm0.1)*10^{5}$	$(1.9\pm0.1)*10^{5}$	$(2.6\pm0.1)*10^{5}$
	3	$(1.0\pm0.1)*10^{4}$	$(0.2\pm0.1)*10^{4}$	$(0.2\pm0.1)*10^{4}$	$(4.1\pm0.1)*10^{4}$	$(5.7\pm0.1)*10^{4}$
June	1	$(3.8\pm0.4)*10^{3}$	$(3.3\pm0.3)*10^{3}$	$(2.7\pm0.3)*10^{3}$	$(4.3\pm0.1)*10^{3}$	$(4.1\pm0.1)*10^{3}$
	2	$(1.0\pm0.1)*10^{5}$	$(0.6\pm0.1)*10^{5}$	$(0.7\pm0.1)*10^{5}$	$(1.4\pm0.1)*10^{5}$	$(1.2\pm0.1)*10^{5}$
	3	$(0.7\pm0.1)*10^{4}$	$(0.3\pm0.1)*10^{4}$	$(0.6\pm0.1)*10^{4}$	$(2.0\pm0.1)*10^{4}$	$(1.9\pm0.1)*10^{4}$
July	1	$(3.5\pm0.3)*10^{3}$	$(2.0\pm0.2)*10^{3}$	$(2.7\pm0.1)*10^{3}$	$(6.5\pm0.1)*10^{3}$	$(7.0\pm0.1)*10^{3}$
	2	$(0.7\pm0.1)*10^{5}$	$(0.5\pm0.1)*10^{5}$	$(0.4\pm0.1)*10^{5}$	$(2.0\pm0.1)*10^{5}$	$(1.4\pm0.1)*10^{5}$
	3	$(0.9\pm0.1)*10^{4}$	$(0.2\pm0.1)*10^{4}$	$(0.3\pm0.1)*10^{4}$	$(2.8\pm0.1)*10^{4}$	$(2.8\pm0.1)*10^{4}$
August	1	$(4.0\pm0.4)*10^{3}$	$(2.6\pm0.3)*10^{3}$	$(2.9\pm0.3)*10^{3}$	$(6.0\pm0.1)*10^{3}$	$(6.5\pm0.1)*10^{3}$
	2	$(0.9\pm0.1)*10^{5}$	$(0.6\pm0.1)*10^{5}$	$(0.5\pm0.1)*10^{5}$	$(1.9\pm0.1)*10^{5}$	$(1.8\pm0.1)*10^{5}$
	3	$(0.5\pm0.1)*10^{4}$	$(0.1\pm0.1)*10^{4}$	$(0.1\pm0.1)*10^{4}$	$(3.2\pm0.1)*10^{4}$	$(3.2\pm0.1)*10^{4}$

The data obtained show that maximal values of ^{3}H specific activity in vegetation are registered on the monitoring ground No.2, which is caused by higher radionuclide concentration in the main sources of contamination – surface and subsoil waters (with respect to two other grounds). Of studied plant species the highest concentration is observed in reed (*Phragmites australis*), which can be explained by its growth directly in water.

In order to assess the spatial ^{3}H distribution in vegetation in the vicinity of the Shagan River, field investigations were carried out on the adjacent territory. 19 observation points were organized along the former riverbed of the Shagan River and on its right riverbank perpendicular to the part of the riverbed with maximal contamination. In each of these points vegetation, vegetation communities and plant species were studied. In some points samples of vegetation were taken to determine the ^{3}H concentration (12 plant samples were taken). The main object of investigations in the area was cheegrass (*Achnaterum splendens*). The figure shows the scheme of location of observation points and values of ^{3}H specific activity in the free water of plants (Figure 39).

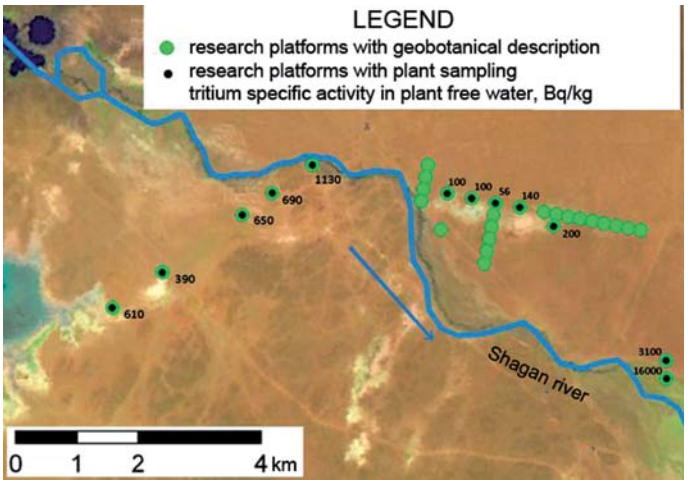


Figure 39. Scheme of location of observation points in the vicinity of the Shagan River and values of ^{3}H specific activity in the free water of plants

The distribution of ^{3}H specific activity in the vegetation in the vicinity of the Shagan River enables to conclude that maximal ^{3}H concentration is registered in plants near the main surface riverbed and considerably reduces with the distance from it. A little increased values of ^{3}H specific activity are registered on the right riverbank of the Shagan River, which is indicative of high concentrations of the radionuclide (more than 1 kBq/kg) in subsoil wasters of the area. A noticeable increase in ^{3}H specific activity is observed at a distance of 4 km from the Shagan riverbed, in the deep hollow, which may be caused by hydrogeological characteristics of the area and is of interest in terms of spatial ^{3}H distribution in the territory with subsoil waters.

2.3.2. Mechanism of vegetation contamination

In order to estimate the mechanisms of ^{3}H redistribution between water and vegetation based on the data obtained on the monitoring stations (Table 4), the ratios of ^{3}H specific activity in the plant free water to the ^{3}H specific activity in surface and underflow waters were calculated (Table 5).

Table 6.

Ratios of ^{3}H specific activity in the free water in vegetation to the ^{3}H specific activity in surface and underflow waters

Sampling month	Expected ^{3}H sources for plants	Monitoring sites									Average
		1	2	3	1	2	3	1	2	3	
		reed			wild rye			cheegrass			
May	Surface water	0.69	0.53	0.22	0.32	0.29	0.04	0.08	0.31	0.04	0,28
	Sub-riverbed water	0.68	0.38	0.16	0.32	0.22	0.03	0.08	0.22	0.03	0,24
June	Surface water	0.88	0.68	0.35	0.77	0.41	0.16	0.63	0.51	0.28	0,52
	Sub-riverbed water	0.93	0.79	0.37	0.80	0.48	0.16	0.66	0.59	0.29	0,56

Sampling month	Expected ^{3}H sources for plants	Monitoring sites									Average
		1	2	3	1	2	3	1	2	3	
		reed			wild rye			cheegrass			
July	Surface water	0.54	0.34	0.31	0.31	0.26	0.08	0.42	0.19	0.10	0,28
	Sub-riverbed water	0.50	0.49	0.31	0.29	0.37	0.08	0.39	0.28	0.10	0,31
August	Surface water	0.67	0.48	0.15	0.43	0.35	0.03	0.48	0.29	0.02	0,32
	Sub-riverbed water	0.62	0.49	0.15	0.40	0.36	0.03	0.45	0.29	0.02	0,31
Average		0,69	0.52	0.25	0.46	0.34	0.08	0.40	0.34	0.11	0.35

The results of calculation show that during all period of observations (from May to August) in all monitoring stations the surface and underflow waters give practically equal ^{3}H contribution to the studied plant species.

A relative stability in the ^{3}H redistribution between water and vegetation does not depend on ^{3}H concentration in surface and underflow waters. For example, for monitoring stations No.1 and No.2 the ratios of ^{3}H amount in water and vegetation are 0.51 and 0.40, respectively. A little decrease in the ^{3}H inflow into vegetation is observed at the monitoring station No.3 (the average calculated value is 0.15), which is probably explained by additional inflow of pure subsoil waters from the right riverbank of the Shagan River. The increase in the intensity of ^{3}H inflow into vegetation is observed in June, which may be caused by meteorological conditions, for example, atmospheric fallouts.

The results of calculations confirm the fact of the highest ^{3}H concentration in reed (*Phragmites australis*) (the average calculated value is 0.49). No considerable difference in ^{3}H concentration in the cheegrass (*Achnaterum splendens* and wild rye (*Leymus angustus*) was registered (average values are 0.29 and 0.28, respectively).

The average ^{3}H quantity in plants in the Shagan River ecosystem amounts to about one third of its quantity in surface and underflow waters.

2.4. Air basin

2.4.1. Assessment of the concentration of artificial radionuclides ^{137}Cs, ^{241}Am, ^{60}Co, ^{152}Eu, ^{154}Eu, ^{90}Sr, $^{239+240}Pu$ in the air basin

As the main pollutant of the air environment in the Shagan River valley is ^{3}H, no direct measurements of ^{137}Cs, ^{241}Am, ^{60}Co, ^{152}Eu, ^{154}Eu, ^{90}Sr, $^{239+240}Pu$ were made. The concentration of the above radionuclides in the air environment was estimated theoretically, based on modeling of the processes of wind transfer of radionuclides from the heap zone around the "Atomic" lake and riverbank soils of the Shagan River. The programs simulate the worst scenario when the wind is directed from the "Atomic" lake, which gives higher ARN concentrations than for any other wind direction.

The dust formed by the wind on the soil surface rises to the atmosphere with different intensity [1]. The fist stage in the process of secondary precipitation is rise of particles from the underlying soil. This process is estimated by the wind rise coefficient. The intensity of wind rise of the long-lived radionuclides from the underlying surface depends on a number of parameters: the level of radioactive contamination of the soil surface layer, soil physical-chemical characteristics, vegetation cover and micrometeorological parameters. The depen-

dence of wind rise intensity on the above-listed parameters has not been well studied. In our case the wind rise coefficient was estimated as [9]:

$$\alpha = \frac{2 \cdot 10^{-10}}{z_0^{1,4}}; \qquad \text{where}$$

α – wind rise intensity, 1/s;

z_0 – height of the underlying surface roughness, cm.

The bottom volume activity (VA) of radionuclides caused by wind rise and further admixture transfer was calculated based on the turbulent diffusion theory using a semi-empirical equation: [9].

$$q(x,y,z) = \frac{aA}{\sqrt{2\pi}\,(1+m)k_1 b_y x^2}\, exp\left(-\frac{y^2}{2b_y^2 x^2} - \frac{u_1(z^{1+m}+z_0^{1+m})}{(1+m)^2 k_1 x}\right) I_0\left[\frac{2u_1(zz_0)^{\frac{1+m}{2}}}{(1+m)^2 k_1 x}\right], \text{ where} \quad (1)\ [9]$$

α – wind rise intensity, 1/s;

A – surface contamination density, Bq/m^2;

m – parameter characterizing atmosphere stability;

κ_1 – turbulent diffusion coefficient for $z=z_1$, m^2/s;

by=0.08, an empirical constant;

u_1 – wind speed at a height z_1=1 m;

I_0 – Bessel function;

z_0 – height of the underlying surface roughness, m;

z – source height, m;

x – distance from the source, m.

The initial data used to determine bottom (average annual) volume activity (VA) of artificial radionuclides in air are presented in the tables (Table 6 – Table 7).

Table 7.

Maximal values of radiation parameters in the heap zone around the "Atomic" lake

Radionuclide specific activity at soil surface, Bq/kg						
^{241}Am	^{137}Cs	^{60}Co	^{152}Eu	^{154}Eu	^{90}Sr	$^{239+240}$Pu
1000	12000	15000	21000	13000	10000	3500

Table 8.

Initial data used to determine calculated average annual volume activity

m	k_1, м^2/с	u_1, м/с	I_0	z_0, м	z, м	α, 1/с	b_y
2.1	0.2	4	1	0.1	1	$7.5 \cdot 10^{-11}$	0.08

The tables below present the results of calculations of the average annual volume activity of artificial radionuclides in air at a height of 1m at different distances from the "Atomic" lake caused by wind transfer from the heap zone around the "Atomic" lake (Table 8), wind transfer from the upstream part of the Shagan River (Table 9) and the resulting volume activity of both components (Table 10).

Table 9.

Average annual volume activity of artificial radionuclides at a height of 1m caused by wind transfer from the heap zone around the "Atomic" lake

x, km	Average annual volume activity, Bq/m^3						
	^{241}Am	^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{60}Co	^{152}Eu	^{154}Eu
0.05	$2.41\cdot10^{-3}$	$3.67\cdot10^{-2}$	$2.45\cdot10^{-2}$	$8.22\cdot10^{-3}$	$3.57\cdot10^{-2}$	$5.08\cdot10^{-2}$	$3.23\cdot10^{-2}$
0.1	$1.20\cdot10^{-3}$	$1.83\cdot10^{-3}$	$1.23\cdot10^{-2}$	$4.11\cdot10^{-3}$	$1.79\cdot10^{-2}$	$2.54\cdot10^{-2}$	$1.61\cdot10^{-2}$
0.2	$6.02\cdot10^{-4}$	$9.17\cdot10^{-3}$	$6.13\cdot10^{-3}$	$2.06\cdot10^{-3}$	$8.93\cdot10^{-3}$	$1.27\cdot10^{-2}$	$8.06\cdot10^{-3}$
0.5	$2.41\cdot10^{-4}$	$3.67\cdot10^{-3}$	$2.45\cdot10^{-3}$	$8.22\cdot10^{-4}$	$3.57\cdot10^{-3}$	$5.08\cdot10^{-3}$	$3.23\cdot10^{-3}$
1	$1.20\cdot10^{-4}$	$1.83\cdot10^{-3}$	$1.23\cdot10^{-3}$	$4.11\cdot10^{-4}$	$1.79\cdot10^{-3}$	$2.54\cdot10^{-3}$	$1.61\cdot10^{-3}$
2	$6.02\cdot10^{-5}$	$9.17\cdot10^{-4}$	$6.13\cdot10^{-4}$	$2.06\cdot10^{-4}$	$8.93\cdot10^{-4}$	$1.27\cdot10^{-3}$	$8.06\cdot10^{-4}$
5	$2.41\cdot10^{-5}$	$3.67\cdot10^{-4}$	$2.45\cdot10^{-4}$	$8.22\cdot10^{-5}$	$3.57\cdot10^{-4}$	$5.08\cdot10^{-4}$	$3.23\cdot10^{-4}$
10	$1.20\cdot10^{-5}$	$1.83\cdot10^{-4}$	$1.23\cdot10^{-4}$	$4.11\cdot10^{-5}$	$1.79\cdot10^{-4}$	$2.54\cdot10^{-4}$	$1.61\cdot10^{-4}$
15	$8.02\cdot10^{-6}$	$1.22\cdot10^{-4}$	$8.17\cdot10^{-5}$	$2.74\cdot10^{-5}$	$1.19\cdot10^{-4}$	$1.69\cdot10^{-4}$	$1.08\cdot10^{-4}$
20	$6.02\cdot10^{-6}$	$9.17\cdot10^{-5}$	$6.13\cdot10^{-5}$	$2.06\cdot10^{-5}$	$8.93\cdot10^{-5}$	$1.27\cdot10^{-4}$	$8.06\cdot10^{-5}$
AVA, Bq/m^3	$2.1\cdot10^{-1}$	$1.7\cdot10^{3}$	$3.3\cdot10^{2}$	$3.2\cdot10^{-2}$	$8.3\cdot10^{2}$	$2.1\cdot10^{2}$	$1.6\cdot10^{2}$

Table 10.

Average annual volume activity of artificial radionuclides at a height of 1m caused by wind transfer from the upstream part of the Shagan River

x, km	Average annual volume activity, Bq/m^3				
	^{241}Am	^{137}Cs	^{60}Co	^{152}Eu	^{154}Eu
0.05	$3.42\cdot10^{-3}$	$9.81\cdot10^{-2}$	$2.10\cdot10^{-2}$	$4.04\cdot10^{-2}$	$1.71\cdot10^{-2}$
0.1	$2.58\cdot10^{-3}$	$7.62\cdot10^{-2}$	$1.81\cdot10^{-2}$	$4.25\cdot10^{-2}$	$1.67\cdot10^{-2}$
0.2	$2.33\cdot10^{-3}$	$4.13\cdot10^{-2}$	$9.59\cdot10^{-3}$	$2.54\cdot10^{-2}$	$9.59\cdot10^{-3}$
0.5	$3.67\cdot10^{-3}$	$4.51\cdot10^{-2}$	$1.26\cdot10^{-2}$	$5.0\cdot10^{-2}$	$2.57\cdot10^{-2}$
1	$3.17\cdot10^{-3}$	$4.67\cdot10^{-2}$	$1.72\cdot10^{-2}$	$3.21\cdot10^{-2}$	$1.28\cdot10^{-2}$
2	$1.42\cdot10^{-3}$	$3.02\cdot10^{-2}$	$6.84\cdot10^{-3}$	$1.48\cdot10^{-2}$	$7.34\cdot10^{-2}$
5	$3.34\cdot10^{-3}$	$1.43\cdot10^{-1}$	$1.46\cdot10^{-2}$	$2.21\cdot10^{-2}$	$1.12\cdot10^{-2}$
10	$1.0\cdot10^{-3}$	$1.03\cdot10^{-2}$	$2.0\cdot10^{-3}$	$2.67\cdot10^{-3}$	$1.67\cdot10^{-3}$
15	$2.50\cdot10^{-3}$	$1.44\cdot10^{-1}$	$5.84\cdot10^{-3}$	$8.09\cdot10^{-3}$	$2.75\cdot10^{-3}$
20	$1.25\cdot10^{-3}$	$1.22\cdot10^{-2}$	$1.25\cdot10^{-3}$	$1.33\cdot10^{-3}$	$1.0\cdot10^{-3}$
AVA, Bq/m^3	$2.1\cdot10^{-1}$	$1.7\cdot10^{3}$	$8.3\cdot10^{2}$	$2.1\cdot10^{2}$	$1.6\cdot10^{2}$

Table 11.

Average annual volume activity of artificial radionuclides at a height of 1m caused by wind transfer from the heap zone around the "Atomic" lake and the upstream part of the Shagan River

x, km	Average annual volume activity, Bq/m^3				
	^{241}Am	^{137}Cs	^{60}Co	^{152}Eu	^{154}Eu
0.05	$5.83\cdot10^{-3}$	$1.35\cdot10^{-1}$	$5.67\cdot10^{-2}$	$9.12\cdot10^{-2}$	$4.94\cdot10^{-2}$
0.1	$3.79\cdot10^{-3}$	$9.46\cdot10^{-2}$	$3.60\cdot10^{-2}$	$6.79\cdot10^{-2}$	$3.28\cdot10^{-2}$
0.2	$2.94\cdot10^{-3}$	$5.04\cdot10^{-2}$	$1.84\cdot10^{-2}$	$3.81\cdot10^{-2}$	$1.77\cdot10^{-2}$

x, km	Average annual volume activity, Bq/m^3				
	^{241}Am	^{137}Cs	^{60}Co	^{152}Eu	^{154}Eu
0.5	$3.91 \cdot 10^{-3}$	$4.88 \cdot 10^{-2}$	$1.62 \cdot 10^{-2}$	$5.51 \cdot 10^{-2}$	$2.89 \cdot 10^{-2}$
1	$3.29 \cdot 10^{-3}$	$4.84 \cdot 10^{-2}$	$1.90 \cdot 10^{-2}$	$3.46 \cdot 10^{-2}$	$1.45 \cdot 10^{-2}$
2	$1.48 \cdot 10^{-3}$	$3.11 \cdot 10^{-2}$	$7.73 \cdot 10^{-3}$	$1.60 \cdot 10^{-2}$	$8.14 \cdot 10^{-3}$
5	$3.36 \cdot 10^{-3}$	$1.44 \cdot 10^{-1}$	$1.49 \cdot 10^{-2}$	$2.26 \cdot 10^{-2}$	$1.15 \cdot 10^{-2}$
10	$1.01 \cdot 10^{-3}$	$1.04 \cdot 10^{-2}$	$2.18 \cdot 10^{-3}$	$2.92 \cdot 10^{-3}$	$1.83 \cdot 10^{-3}$
15	$2.51 \cdot 10^{-3}$	$1.44 \cdot 10^{-1}$	$5.96 \cdot 10^{-3}$	$8.26 \cdot 10^{-3}$	$2.86 \cdot 10^{-3}$
20	$1.26 \cdot 10^{-3}$	$1.23 \cdot 10^{-2}$	$1.34 \cdot 10^{-3}$	$1.46 \cdot 10^{-3}$	$1.08 \cdot 10^{-3}$
AVA, Bq/m^3	$2.1 \cdot 10^{-1}$	$1.7 \cdot 10^{3}$	$8.3 \cdot 10^{2}$	$2.1 \cdot 10^{2}$	$1.6 \cdot 10^{2}$

The results of calculations show that the data of tables 9 and 10 are practically identical except for a small section located at a distance of a few hundreds of meters from the "Atomic" lake. The calculated average annual volume activity of artificial radionuclides along the Shagan River is much lower than maximal permissible volume activities in NRB-99.

2.4.2. Concentration of ^{3}H radionuclide in the air basin

To study dynamics of atmospheric air contamination with ^{3}H, in the period May-August 2008, sampling of water vapour was carried out on the monitoring stations of the hydrological sections Nos. 1, 2, 3 located at a distance of 0.8, 4.7 and 11 km from the "Atomic" lake. The results of measurements of ^{3}H concentration in the water vapour are shown in the diagram (Figure 40). The activity is given in Bq/kg for frozen water.

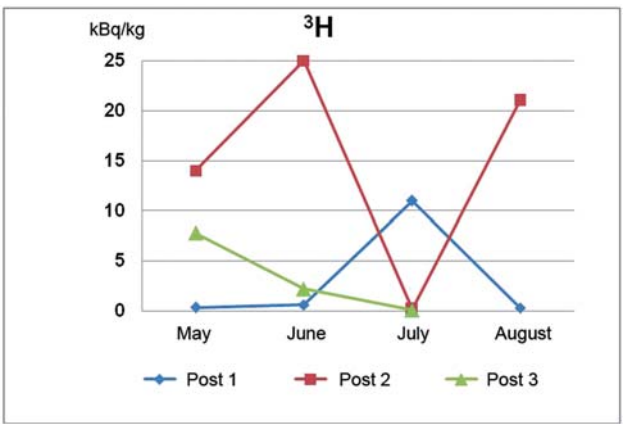


Figure 40. ^{3}H distribution in water vapour samples

The results of laboratory analyses showed that maximal values of ^{3}H concentration in water vapour, 2.5 x 10^4 Bq/kg, were registered at the monitoring station (point) No. 2 at the end of June. It is a predictable result as in this point maximal ^{3}H concentrations were registered both in surface and ground waters. What is unexpected is the fact that in the same point in a month period the ^{3}H concentration becomes tens of times lower and a month later returns to the previous level > 2 x 10^4 Bq/kg. The monitoring stations Nos. 1, 3 also registered sharp changes in the ^{3}H concentrations.

The above data enable to conclude that ^{3}H concentration in the atmospheric air in the part of the Shagan River flowing across the STS territory can vary in a wide range.

It is also of interest to study the dependence of ^{3}H concentration as a function of distance from the riverbed. For this purpose, in 2007 water vapour samples were taken on the left bank of the Shagan River, in the point located 7.3 km away from the "Atomic" lake and water vapours were samples along the line perpendicular to the riverbed axis. The results of laboratory studies are presented in the figure (Figure 41).

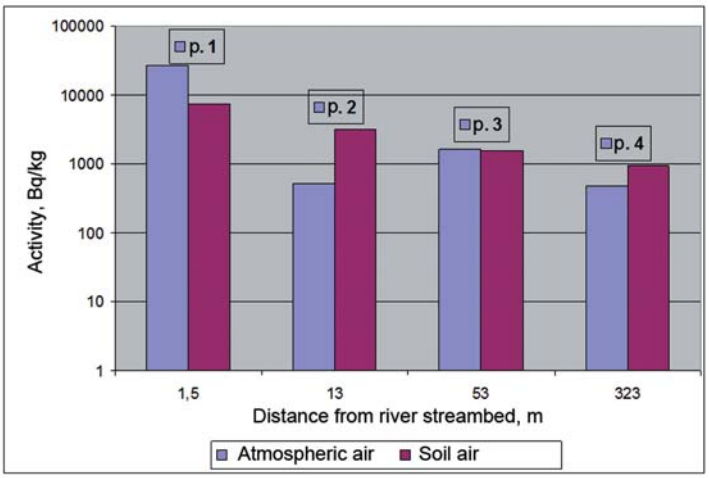


Figure 41. ^{3}H distribution in soil and atmospheric air

The above figure shows that the highest ^{3}H amount, 7470 Bq/kg, rises to the atmospheric air from the flood-plain part of the riverbank at a distance of 1.5 km from the riverbed. The depth of underflow waters in that point is 0.2 m. In the other sampling points gradual decrease in ^{3}H concentration in the soil air, to 940 Bq/kg, was registered, which is probably caused by the decrease in the level of ground waters.

2.4.3. Mechanisms responsible for atmospheric air pollution and its prognosis

Section 2.4.1 gives theoretical estimation of concentration of radionuclides ^{137}Cs, ^{241}Am, ^{60}Co, ^{152}Eu, ^{154}Eu, ^{90}Sr, $^{239+240}Pu$ in the atmospheric air and describes the mechanism responsible for this type of pollution.

The experimental data on ^{3}H concentration in the Shagan River waters and the initial data were used to estimate zoning of the territory in terms of ^{3}H concentration in the atmospheric air. The zoning was made in two directions:

- Along the Shagan riverbed from the "Atomic" lake to the inflow into the Irtysh;
- In the direction perpendicular to the Shagan riverbed.

^{3}H concentration in the atmospheric air for territory zoning was estimated using ^{3}H concentration in the river waters based on the condition that the average concentration of water vapours in the atmosphere was about 10 g/m^{3}. Territory zoning in the perpendicular direction was made using the experimental dependence of ^{3}H concentration on the distance

from the source. The experiments were made on *"Degelen" test ground*, in streams of tunnels Nos. 176 and 177. The calculations did not take into account emanation from the soil surface and transpiration of vegetation.

According to the estimations the ^{3}H intake with inhaled air is the main way of radionuclide penetration into human organism. This fact can be used for preliminary zoning of the riverbank zone of the Shagan River by the level of ^{3}H concentration in the air basin. It is suggested thin the territory should be divided into three zones according to the expected annual effective dose from ^{3}H intake with inhaled air:

- Zone of satisfactory radiation situation (<0.3 mSv/year);
- Zone of limited presence of people (0.3 –1 mSv/year);
- Zone of hazardous radiation (>1 mSv/year).

As the initial data for zoning one must use values of ^{3}H average annual volume activity in air equal to <140, 140–460, >460 Bq/kg, respectively.

Based on the values of ^{3}H average annual volume activity in air, the Shagan riverbed can be subdivided into three zones along the river flow starting from the "Atomic" lake to the inflow to the Irtysh River (Table 11).

Table 12.

Zoning of the territory along the riverbed of the Shagan River according to the ^{3}H concentration in air

Distance, km	Zone		
	Radiation hazard	Restricted public access	Relatively good radiological conditions
0 – 27	>460 Bq/m^3	-	-
27 – 30	-	140 – 460 Bq/m^3	-
>30	-	-	<140 Bq/m^3

In the direction perpendicular to the riverbed on the left and right side of the river three zones are also determined according to the level of ^{3}H concentration in the atmospheric air (Table 12).

Table 13.

Zoning of the territory perpendicular to the riverbed of the Shagan River according to the ^{3}H concentration in air

Distance from the riverbed, m	Zone		
	Radiation hazard	Restricted public access	Relatively good radiological conditions
0 – 5 km away the "Atomic" lake			
0 – 45	>460 Bq/m^3	-	-
45 – 50	-	140 – 460 Bq/m^3	-
>50	-	-	<140 Bq/m^3

Distance from the riverbed, m	Zone		
	Radiation hazard	Restricted public access	Relatively good radiological conditions
5 – 10 km away the "Atomic" lake			
0 – 25	>460 Bq/m^3	-	-
25 – 42	-	140 – 460 Bq/m^3	-
>42	-	-	<140 Bq/m^3
10 – 20 km away the "Atomic" lake			
0 – 20	>460 Bq/m^3	-	-
20 – 40	-	140 – 460 Bq/m^3	-
>40	-	-	<140 Bq/m^3
20 – 25 km away the "Atomic" lake			
0 – 10	>460 Bq/m^3	-	-
10 – 35	-	140 – 460 Bq/m^3	-
>35	-	-	<140 Bq/m^3
27 – 30 km away the "Atomic" lake			
10 – 20	-	140 – 460 Bq/m^3	-
>20	-	-	<140 Bq/m^3
40 km away the "Atomic" lake			
	-	-	<140 Bq/m^3

Therefore only at a distance of 40 km from the "Atomic" lake along the riverbed is the boundary which divides the zone of limited presence of people and the zone of relatively satisfactory radiation situation, where ^{3}H concentration in air is < 140 Bq/kg. It should be noted that zones of limited public presence and hazardous radiation are rather narrow – several tens of meters. For example, at a distance of 50 m from the riverbed ^{3}H concentration in air is safe for people along all length of the Shagan River.

2.5. Production of livestock farming

In the period from May to August, in winter camps *Atomkol*, *Yubileynaya* and *Sarapan* located close to the Shagan River, 9 samples of livestock farming products were taken for analysis. The results of analysis are presented in the table (Table 13).

Table 14.

Winter camp	Sample #	Sample	Sampling date	Specific activity, Bq/kg		
				^{3}H	^{137}Cs	^{90}Sr
Atomkol	1	kumiss	29.05.2008	(1±0.1)*10^2	0.9±0.2	0.84±0.24
Yubileynaya	2	milk	30.05.2008	(53±0.5)*10^2	1.7±0.3	<0.16
	3	milk	25.06.2008	(14±0.2)*10^2	-	-
	4	milk	24.07.2008	(14±0.2)*10^2	-	-
	5*	milk	5.09.2008	(43±4)*10^2	-	-
	6	Pulmonary tissue	24.06.2008	(90±0.5)*10^2	-	-

Winter camp	Sample #	Sample	Sampling date	Specific activity, Bq/kg		
				^{3}H	^{137}Cs	^{90}Sr
Sarapan	7	milk	26.06.2008	(38±0.3)*10^{2}	-	-
	8	milk	25.07.2008	(27±0.3)*10^{2}	-	-
	9*	milk	5.09.2008	(9±0.9)*10^{2}	-	-

Specific activity of radionuclides ^{137}Cs and ^{90}Sr in milk and kumiss samples taken in May was low which excluded necessity to determine their concentration in later months. ^{3}H specific activity in milk does not exceed the interference level for public, however, the data refer only to summer months, and the data for winter months may differ significantly. ^{3}H concentration determined in the lung tissue of small cattle (winter camp *Yubileynaya*) was higher than the interference level for public, which is an additional evidence of necessity of further investigations.

CONCLUSION

To study the present-day state of the main components of the Shagan River ecosystem it was necessary to analyze materials of radioecological surveys in the region. The mechanisms responsible for formation of radioecological situation in the vicinity of the Shagan River were studied.

It has been established that radioactive contamination of the ecosystem components is mainly caused by excavation explosion in the borehole 1004 and underground nuclear tests in the "warfare" boreholes on the test ground "Balapan". A small contribution to the radioactive contamination was made by the tests on the test ground "Opytnoye pole" and global precipitations.

The results of investigations enabled to detect two large zones of radioactive contamination.

The first zone is located in close vicinity of the "Atomic" lake crater and is connected with the test in the borehole 1004. The radioactive contamination in that area is areal contamination. The total area of contaminated space is limited by a distance of 1-2 km from the crest of the crater. The field radiometric measurements detected high values of radiation parameters only in the vicinity of the "Atomic" lake where a sector of the area of 20 km^{2} with EDR exceeding 0.3 μSv/h was contoured. Gamma-spectrometric and radiochemical analyses of soil samples from the "Atomic" lake area showed that practically all the examined area had concentrations of ^{137}Cs, ^{90}Sr and $^{239+240}Pu$ exceeding background of global precipitations.

The concentrations of radionuclides $^{239+240}Pu$, ^{137}Cs and ^{90}Sr (besides ^{90}Sr in the place where the Shagan River flows out of the "Atomic" lake) do not exceed the intervention levels for people when they intake the radionuclide with water and food, the levels presented in NRB-99 (Appendix P-2) are equal to 0.56, 11 and 5 Bq/kg, respectively [1].

The second zone is located 4.5 km from the "Atomic" lake. It is the area where the fact of discharge of ^{3}H contaminated ground waters into the surface flow of the Shagan River was discovered and repeatedly confirmed. ^{3}H specific activity in water in this section of the river is as high as 680 kBq/kg. This area is the source of secondary ^{3}H contamination of ecosystem components, such as plants, livestock products and air. The ^{3}H presence was registered in all

plant species sampled on the Shagan banks. Maximal ^{3}H concentration in plants 65 x10^{3} Bq/kg was registered in reed, it is 8 times higher than the intervention level for people in case of radionuclide intake with water or food (NRB-99). Tritium concentration in milk samples taken in winter camps located on the Shagan banks does not exceed intervention level (NRB-99). Nevertheless, as tritium concentration in the Shagan water can vary in a wide range, the fact of detection of high tritium concentrations in milk samples makes it necessary to take the following measures: 1) to continue studying of radionuclide concentrations in the livestock products, 2) to implement obligatory radiation control of products going to the market. The necessity of such measures is supported by the fact of exceeding of tritium intervention level in the lung tissue sample.

The values of radiometric parameters (EDR, density of beta-particles) in the region do not exceed the background values for the STS territory.

The concentrations of artificial radionuclides in the riverbank soils and bottom sediments sampled along the riverbed do not exceed minimal-significant values for specific activity stated in NRB-99. A maximal value of ^{137}Cs activity different from the average value was discovered at distances up to 200 m from the "Atomic" lake i.e. within the first zone. The soils sampled in the points could be classified as limited usage materials (LUM, from 300 Bq/kg to MVSA).

Therefore the main potential hazard is contained in ground and surface waters contaminated with tritium. At the present time we have part of the river with liquid radioactive wastes. This fact makes it necessary to implement a complex of measures aimed at exemption of potentially hazardous lands from land use.

It is difficult to determine the boundaries of the exempted zone now. Tritium concentration in the Shagan River varies not only within the season but also from year to year depending on precipitations, water flow rate and other climatic and hydrogeological factors. To establish boundaries of exempted lands it is necessary to carry out a complex of investigations, which must include studying of ways of inflow and outflow of tritium contaminated ground waters, as well as hydrological regime of the Shagan River with further modeling of the worst scenario for climatic and hydrogeological conditions, which may give maximal possible tritium contamination of the Shagan River covering the largest area. Carrying out of such research and further exemption of the part of the area from land use cannot be delayed, as the problem of tritium contamination of the part of the Shagan River is really pressing.

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ШАҒАН ӨЗ. ЭКОЖҮЙЕСІНІҢ ЖАЙ-КҮЙІ ЖӘНЕ ОНЫҢ ҚАЛЫПТАСУЫНЫҢ НЕГІЗГІ МЕХАНИЗМДЕРІ

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Шаған өзені Семей сынақ полигонының басты су күретамыры болып саналады, және өзіне іргелес жатқан аумақтармен бірге экожүйені құрайды, экожүйеге жерасты және жерүсті сулары, түп шөгінділері және жағалау топырақтары, ауа бассейні, өсімдік және жануарлар әлемі кіреді. Бұл құраушылардың барлығы қандай дәрежеде болмасын Семей сынақ полигонындағы ядролық жарылыстардың салдарынан радиоактивті ластануға ұшыраған. Аталған жұмыста осы ауданда жүргізілген радиоэкологиялық зерттеу материалдары талданған және қорытындыланған, экожүйенің қазіргі жалпы қалпы баяндалған, оның қалыптасуының басты механизмдері

қарастырылған. Радиоактивті ластанудың әр түрлі типі бар екі ірі аймақ анықталды. Бірінші аймақ "Атом" көлі маңайы шұңқырының қырына орайластырылған, мұнда радионуклидпен - 1004 ұңғымадағы экскавационды жарылыс кезінде пайда болған бөлу өнімі мен нейтронды белсендірумен - алаңдық ластану бар. Екінші облыс "Атом" көлінен 4-5 км қашық орналасқан. Мұнда "Балапан" алаңының "әскери" ұңғымалар аудандарынан Шаған өзенінің жерасты және жерүсті суларына тритий түседі, бұл кезде оның өзен суындағы шоғырлануының араласу деңгейі 2 ретке жоғарылайды. Аталған телімнің суы шындығында сұйық радиоактивті қалдық ретінде байқалады. Сондықтан Шаған өзенінің бөлігі мен оған іргелес аумақтарды шаруашылық айналымнан алып тастау мәселесі өте өзекті болып табылады.

Түйін сөздер: ядролық сынақтар, "Атом" көлі, экожүйе, гидрогеология, радиоактивті ластану, радионуклидтер, ұңғымалар.

УДК 577.4:504.4.054: 539.16

СОСТОЯНИЕ ЭКОСИСТЕМЫ Р. ШАГАН И ОСНОВНЫЕ МЕХАНИЗМЫ ЕГО ФОРМИРОВАНИЯ

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Река Шаган является главной водной артерией Семипалатинского испытательного полигона, и вместе с прилегающей к ней территорией образует экосистему, включающую подземные и поверхностные воды, донные отложения и береговые грунты, воздушный бассейн, растительность и животный мир. Все эти компоненты подверглись, в той или иной степени, радиоактивному загрязнению вследствие ядерных испытаний на Семипалатинском полигоне. В данной работе проанализированы и обобщены материалы радиоэкологических исследований, проведенных в данном районе, изложена общая картина современного состояния экосистемы, рассмотрены основные механизмы ее формирования. Установлены две крупные зоны с принципиально разным типом радиоактивного загрязнения. Первая зона приурочена к гребню воронки вокруг "Атомного" озера, здесь присутствует площадное загрязнение радионуклидами – продуктами деления и нейтронной активации, образовавшимися при экскавационном взрыве в скважине 1004. Вторая область расположена в 4-5 км от "Атомного" озера. Здесь в подземные и поверхностные воды р. Шаган из района "боевых" скважин площадки "Балапан" поступает тритий, при этом его концентрация в воде реки превышает уровень вмешательства на 2 порядка. Вода данного участка реки фактически представляет собой жидкие радиоактивные отходы. Поэтому остро актуальным является вопрос изъятия части р. Шаган и прилегающей к ней территории из хозяйственного оборота.

Ключевые слова: ядерные испытания, "Атомное" озеро, экосистема, гидрогеология, радиоактивное загрязнение, радионуклиды, скважины.

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PECULIARITIES OF RADIOACTIVE CONTAMINATION OF ENVIRONMENT IN WATER FLOW ECOSYSTEM OF THE TUNNELS AT DEGELEN MOUNTAIN Massif

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The paper presents data on the contamination of the ecosystem components caused by migration of radionuclides with water. The studies provided data on the nature of radioactive contamination of soil, surface waters and water ecosystems underflow conduit of flow streams from the tunnels of Degelen Massif. There has been showed the character of spatial distribution of radionuclides in these components of the environment. There were determined series of mobility, both vertical and horizontal proliferation of radionuclides in soils along and across the bed of the watercourse. Also, there has been shown the potential for forecasts of radionuclides in the soil by the specific activity of these radionuclides in water. Different quantities of radionuclides in the conjugate components of ecosystems (surface water - groundwater - soil) showed the disequilibrium of their condition in this system. Research of tunnel watercourses with water seepage, chosen as objects of research for this work, allows predicting the behaviour of radionuclides in the meadow soils of other ecosystems of water flows from the tunnels of "Degelen" Massif. Based on analysis of the curves describing the behaviour of radionuclides in the horizontal direction, one can predict that at this point of time ^{137}Cs and $^{239+240}$ Pu would not extend beyond 1.5 km from the outlet, ^{90}Sr - not more than 2 km.

Key words: nuclear tests, radioactive contamination, radionuclides, tunnels, meadow soils, distribution of radionuclides.

INTRODUCTION

The underground nuclear explosions made in 1961–1989 in adits of the Degelen mountain massif caused radioactive contamination not only of adit cavities but also of daylight surface. In spite of demilitarizing measures in the territory, carry-over of radioactive substances by water flows from adits does not stop. As a result, a large amount of radioactive substances has been carried over and is still being carried over to the environment, where they are accumulated in soil taking part in geochemical processes.

After ceasing nuclear tests on "Degelen" test ground, radioecological investigations have been carried out in the framework of national budget projects and international contracts (DSWA, ISTC, INTAS, HDTRA, etc.). The first radioecological projects fulfilled in the framework of the contract DSWA 0027/5 of 1 June 1997 included field gamma-survey of the near-portal areas of studied tunnels, gamma-spectrometric analysis of soil samples in laboratory and estimation of $^{239+240}$Pu and ^{90}Sr concentrations. The investigations enabled to reveal the most contaminated areas with concentrations of $^{239+240}$Pu, ^{90}Sr and ^{137}Cs up to $9*10^6$, $4.5*10^5$, and $6.9*10^6$ Bq/kg, respectively. Observations detected migration of artificial radionuclides along the water flow in the riverbed soils from water outcomes in tunnels, with migration abilities of radionuclides forming the following series: ^{90}Sr>$^{239+240}$Pu>^{137}Cs. As a result of investigations the following conclusion was made: it is necessary to seal tunnels with concrete seals, which to some extent can suppress release of radionuclides from tunnel cavities to the valleys of Degelen mountains.

Under the ISTC project the radiation contamination of surface waters and soils was studied in the near-portal areas around tunnels and stream riverbeds. The most contaminated

zones were detected, and their areas were schematically outlined. It was established that nowadays the most intensive sources of contamination are water streams from tunnels. The first theories about horizontal and vertical migration of artificial radionuclides in soils of the streams of the Degelen mountain massif were developed and rehabilitation measures – use of natural zeolite modified by titanium hydroxide as a sorbent of radionuclides – were suggested.

The monitoring of tunnel streams carried out in the frames of the national budget programs gave the data on the amount of radionuclides carried away by tunnel streams of the Degelen mountain massif from cavities of the tunnels [1, 2, 3].

The above-listed projects gave a certain amount of information on radioecological situation on the "Degelen" test ground . However, it was necessary to carry out investigations to outline in more detail boundaries of stream valleys in soils and to determine spreading of artificial radionuclides ^{137}Cs, ^{90}Sr and especially $^{239+240}Pu$ whose distribution was not revealed in the previous works. The investigations were carried out in the valleys of the stream Baitles (tunnel No.176) and tributary of the stream Uzynbulak (tunnel No. 177). The works also included studying of the character of spatial (both horizontal and vertical) distribution of artificial radionuclides in soils of stream valleys of the Degelen mountain massif.

1. GENERAL CHARACTERISTIC OF THE "DEGELEN" TEST GROUND

Administrative and geographical position of the studied area. The "Degelen" test ground is located in southern part of the Semipalatinsk Test Site in the territory of Eastern-Kazakhstanskaya oblast (former Semipalatinskaya). In geographical terms it is a Degelen mountain massif, the north-eastern part of the Kazakh fold structure. Its total area is about 220 sq. km.

Relief. The prevailing type of relief of the studied area is a hilly landscape with heights of 300-500 m and prevalence of dome-shaped and cone-shaped hills. There are also rocky mountains formed by granites with heights over 1000 m (1084 and 1,126 m) looking like islands on the low-hilly landscape (Figure 1). They mainly form massifs with relicts of smoothed ancient surfaces and fold structures. Among dense rocks there are sediment structures – slates, sandstones, conglomerates, limestone and effusive structures, both bases and acids [4].

Hydrography. Degelen hydrographic network consists of several streams, the largest of which are the Uzynbulak, Baitles, Toktakushik and Karabulak (Figure 1). The largest watershed belongs to the valley of the stream Uzynbulak dividing the mountain massif along the line south-east – north-west. The streams Baitles and Toktakushik flow to the south and south-east, whereas the stream Karabulak flows to the north, north-east. A typical feature of all streams is the absence of continuous surface runoff. A continuous flow may be formed only in spring, during intensive snow melting, later the stream divides into separate ponds – river-reaches which quickly disappear due to evapouration and permeation into subsoil waters.

Hydrogeology. The Degelen mountain massif is part of regional hydrogeological system of the Irtysh left bank [5]. Occupying a dominating position in the relief, it is a source replenishing ground waters due to atmospheric fallouts. The aquifer partially discharges at the beginning of summers in the sides of stream valleys. In the other part of the year

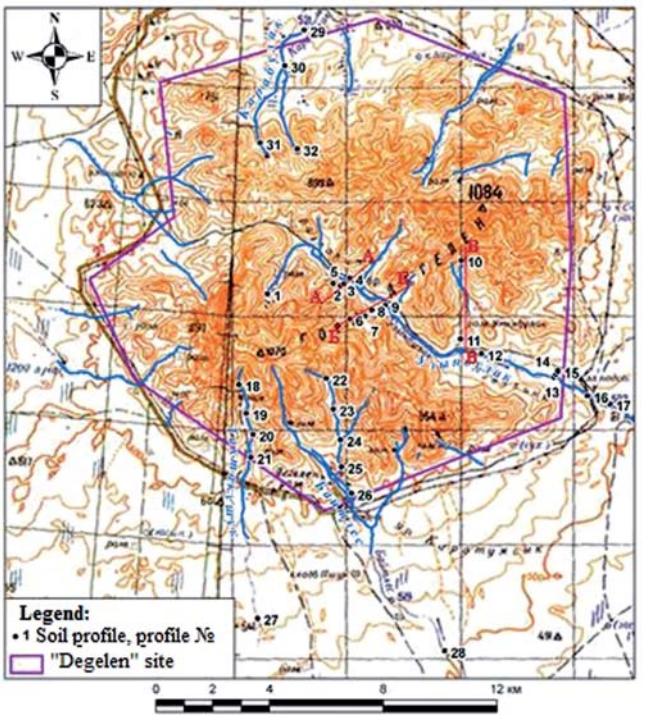


Figure 1. Location of streams, research sites and profiles on the "Degelen" test ground

discharges of springs feeding streams are very small. The underground explosions caused increase in the rock disintegration which, on the one hand, led to greater accumulation of atmospheric fallouts in fracture zones and, on the other hand, increased the part of surface waters permeating to ground waters through the system of fractures. The waters on the massif territory are divided into porous waters of diluvial – proluvial, alluvial – proluvial deposits and fracture waters of Paleozoic rocks.

Soils and vegetation. In terms of soils and geography the territory of the Degelen mountain massif is located in the sub-zone of desertified steppes with light-chestnut soils [6]. Nonuniformity of soil cover is determined by relief and, hence, by moistening and composition of soil-forming rocks.

Most widely spread soils in the territory of Degelen massif are light-chestnut low-developed rubbly soils (thickness up to 40 cm) on light rocks. They are formed in the low-hilly relief where they mainly occupy tops and crests of hills or slopes of rocky mountain massifs. Undeveloped soils are observed near soils of higher thickness – not fully developed soils of fine soil thickness up to 80 cm. Soil-forming structures are eluvial crushed rock or eluvial-dealluvial crushed loam stones. The vegetation cover is rare presented by wormwood – feather grass – fescue species (fescue, feather grass, eastern mat-grass, wormwood sage) with rare islands of bushes (bushy elm, John's wood spiraea, etc.). The soil cover is rather thin, composed of loamy lumpy light chestnut soils with crushed stones. Very often it consists of one horizon "A". Such soils are well washed and not saline.

All undeveloped and not fully developed soils are characterized by the absence of developed carbonate horizon and specific water regime depending on water retention in underlying dense layers or rough skeletal product of their weathering. Therefore changes in weather cause sharp variations in moisture and temperature of soil profile. The content of stony particles even in the upper humus layer is sometimes as high as 50 %. High concentration of rough particles in soils changes their physical-chemical properties such as volumetric and specific weight and porosity. Stony soils with high admixture of broken stones have high filtration ability maintaining dry water regime all time except the moments of atmospheric fallouts.

Light chestnut normal soils lie only in the wide parts of the valley, more often on the outer slopes of hills surrounding the Degelen mountain massif. They have a stable formed profile with thickness of humus horizon ("A" + "B") not more than 35 cm and clearly outlined division into horizons. On the bottom of the profile is a dense carbonate-illuvial horizon turning into soil-forming rock. In the humus horizon, under the action of HCl soil effervescence is observed. The humus concentration in the upper horizons is as high as 2.5–3.0%.

The elevated areas have places with mountain chestnut soils which mainly have porous sediments of small thickness. They differ from light chestnut soils by well pronounced brown colour of the profile and higher humus content up to 4%.

Zonal automorphic light chestnut soils are formed in the conditions of high moisture shortage. The main source of moistening is atmospheric fallouts whose amount in the subzone of desertified steppes is not more than 250 mm per year. Light chestnut meadow soils unlike automorphic soils refer to soils of semi-hydromorphic group which get additional moistening due to surface runoff and subsoil waters. They are formed under the meadow-steppe vegetation. Vegetation is presented by widely spread species of wormwood, feather grass and fescue growing in desertified steppes (wormwood, feather grass, fescue) with rather large amount of bushes, motley grass and some meadow plants (sedge, ricegrass, licorice). Meadow chestnut soils are observed on the studied territory in depressions surrounding the Degelen mountain massif, in the lower parts of hills and mountain massifs, however, they are not widely spread. As they develop in more favorable conditions than zonal soils, they have higher thickness of humus horizon, higher content of organic substance and, hence, darker colour. Such soils are often saline and solonetzic.

The riverbeds of numerous streams mainly have meadow soils and very rarely meadow-swampy soils. Some meadow soils are drying-up and desertified, in some places meadow soils are saline but they are mainly spread on flattened parts of wide valleys at the edge of the Degelen mountain massif where slopes and drainage decrease. The meadow soils of the upper and lower flows of valleys are, as a rule, well washed, hence, not saline and rubbly as all soils of the mountain massif. They have dark-grey colour of profile with high thickness of humus horizon. The content of organic substance in soils varies from 20 % in the upper reaches to 4–5 % in the lower reaches depending on moisture and, hence, richness of vegetation, which is the source of humus. The meadow soils in valleys were subjected to the highest radiation contamination during tests as tunnels are located in the stream valleys. However, contamination did not stop with cessation of nuclear tests, it continues due to carry-over of the explosion products with subsoil waters from tunnels.

The vegetation in stream valleys is mainly presented by meadow species adapted to steppe conditions (woodreed (*Calamagrostis epigeios),* Ural licorice *(Glycyrrhiza uralensis)*, fowl grass (*Poa trivialis),* solonetz biflorous *(Galatella biflora)*. In some areas there are bushes of grey willow *(Salix cinerea)* and naked-leaved dog-rose *(Rosa glabrifolia)*.

As ecosystems of stream valleys: meadow soils – subsoil waters – vegetation are subjected to continuous radiation contamination, they were chosen as an object of more detailed investigations. The main attention in the system is drawn to soils as they are the depot for all pollutants including artificial radionuclides which enter ecosystem and can transfer into other components of the environment.

2. STUDIES OF RADIOECOLOGICAL CHARACTERISTICS OF ECOSYSTEM OF TUNNEL STREAMS ON THE "DEGELEN" TEST GROUND

2.1. Materials and methods of research

As the object of further investigations, the ecosystems of streams flowing out from tunnels No.176 in the valley of the Baitles stream and tunnel No.177 in the valley of the Uzynbulak tributary were chosen. The choice was made on the base of reconnaissance studies, data on discharges of outcomes from the tunnels of the "Degelen" test ground , radionuclide characteristic of tunnel waters and contamination of the near-port areas around tunnels. An important factor for choosing the above tunnel was different conditions. Firstly, the tunnel No. 176 is located in the valley of the Baitles stream whose runoff goes beyond the Degelen mountain massif. The tunnel No. 177 is located in the right tributary of the Uzynbulak stream, so all its runoff remains in the valley. Secondly, the water from the tunnel No. 176 carries away large amounts of ^{137}Cs and ^{90}Sr, whereas the water from the tunnel No. 177, in addition to the above radionuclides, contains $^{239+240}$Pu (up to 12 Bq/l) and ^{241}Am (up to 2.6 Bq/l). As the objects of investigations on the studied territories, scientists chose ecosystem components subjected to the influence of radiation-contaminated waterways – subsoil and surface waters and soils. A specific feature of the ecosystems is the fact that their location excludes impact of other tunnels on the radiation situation.

Field works included measurement of radiation parameters, sampling of environment components, pedestrian gamma-survey and determination of geographical coordinates [7, 8, 9, 10, 11, 12, 13].

The laboratory analyses of radionuclide concentration were made according to the standard methodical instructions on the tested equipment. Concentrations of ^{137}Cs, ^{241}Am, ^{60}Co, ^{152}Eu, ^{154}Eu, ^{40}K, ^{238}U, ^{232}Th were determined as in [14], concentration of ^{90}Sr and $^{239+240}$Pu was determined using radiochemical technique [15, 16] and concentration of ^{3}H was measured on beta-spectrometer TriCarb [17].

Physical-chemical properties of soils were determined by the methods generally used in soil sciences: humus concentration in soil was determined by Tyurin's method [18], acidity – by potentiometric method [19], concentration of exchangeable calcium and magnesium ions was determined by the method based on displacement of cation 1N by NaCl solution [20], the mechanical analysis was made using Kachinskiy's technique [21].

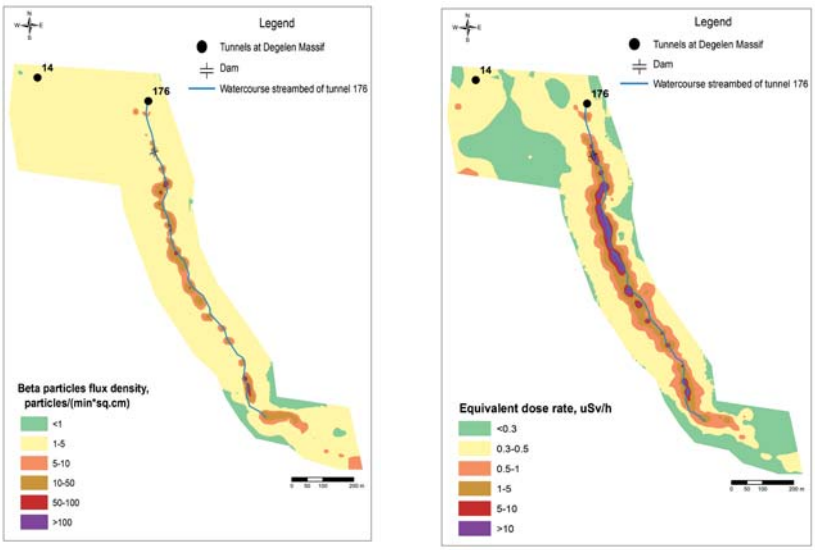
2.2. Description of the system of waterways of tunnels Nos. 176 and 177 as objects of investigation

Ecosystem of tunnel No. 176. The investigations carried out earlier [3] established that the average annual flow rate from the tunnel No. 176 was more than 500 l/min. The average annual specific activity of radionuclides at the stream outcome to the surface was 89 Bq/kg for ^{137}Cs, 275 Bq/kg for ^{90}Sr and 341 Bq/kg for ^{3}H. The data of radiation monitoring show that annual carry-over of artificial radionuclides with tunnel water was not constant over the period of observations and primarily depended on the amount of atmospheric fallouts. The ^{90}Sr content in waters was always 2-3 times higher than that of ^{137}Cs [2]. The specific activity of $^{239+240}$Pu was below the detection limit equal to 0.02 Bq/kg. The results of field radiometry made at the moment of closure showed that EDR on the near-port area was less than 0.50 μSv/h, the density of α- and β-particle fluxes was below the detection limit (<0.5 and <10 part./min*cm^2, respectively) [22].

The area studied is the left-bank side of the Baitles valley separated from the main riverbed by a low elevation of naked rocks stretching along the riverbed. The riverbed width from the tunnel does not exceed 1 m. In some places the water flow has to overcome lumps and rock plates. At a distance of 700-750 m from the dam in the flat riverbed formed by granite plats there is no water. Further, the riverbed goes down with a bench of a height of 3 m, where in case of flood, water goes down as a waterfall.

Brief radiation characteristic. To assess current radioactive contamination of the studied ecosystem more detailed measurements of the equivalent dose rates (EDR) and the beta-flux density on the soil surface with step 25x25 m were made.

The examination showed the highest radioactive contamination in the riverbed of the stream of tunnel No. 176 (Figure 2). The density of β-particles flux varied from background values of 10 part./(min*cm^2) to 2700 part./(min*cm^2). The EDR value varied within 0.13–23.9 μSv/h.



a b

Figure 2. Schematic map of distribution of β-flux density (a) and EDR (b) at the head of tunnel No.176

Ecosystem of tunnel No. 177. The studied territory of the left-bank side of the Uzynbulak tributary is the ecosystem of the waterway of tunnel No. 177. It is separated from the main riverbed by a low elevation stretching along the riverbed and the road embankment. The stream width is mainly not more than 1 m.

On its way the stream runs into heaps of debris where the flow disappears or its riverbed changes. Besides the main waterflow, at the foot of the massif, where the crater of the studied tunnel is located, there appear small springs forming a common stream. The distance from the source of the stream of tunnel No. 177 to the main riverbed of the stream Uzynbulak is about 650 m (Figure 3).

Tunnel No. 177 is now characterized by the presence of a constant stream (in the first years of observations water appeared seasonally). During the years of observations, 2001–2005, the average annual water discharge was estimated as $4.4*10^8$ (more than 500 l/min) [3]. In that period the average annual concentration of radionuclides in the tunnel water varied from 2 to 6 Bq/kg for ^{137}Cs, 630 to 1100 Bq/kg for 2 ^{90}Sr, 550 to 630 Bq/kg for ^{3}H. The earlier analyses of water samples registered almost 12 times excess of the intervention level $^{239+240}$Pu (0.56 and 6.6 Bq/kg).

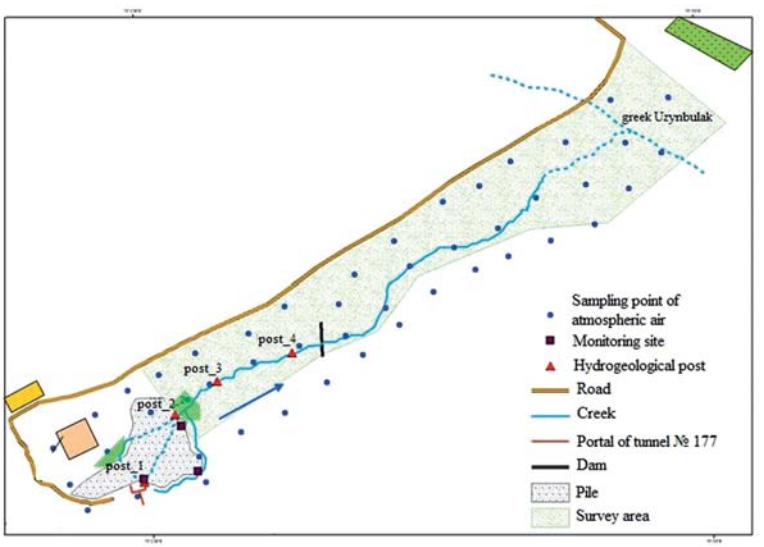


Figure 3. Scheme of the surveyed territory of stream from tunnel No. 177

Brief radiation characteristic. In order to assess current radiation contamination of the studied ecosystem, more detailed measurements of the equivalent dose rates (EDR) and the beta-flux density on the soil surface with step 10x10 m were made in the vicinity of tunnel No. 177.

The survey showed the highest radioactive contamination along the central riverbed of the stream of tunnel No. 177 (Figure 4). The density of β-particles flux varied from background values of 10 part./(min*cm^2) to 1200 part./(min*cm^2). The EDR value varied within 0.18–3.7 μSv/h. The values of α-flux density did not exceed the detection limit of measurement devices (0.06 part./(min*cm^2)).

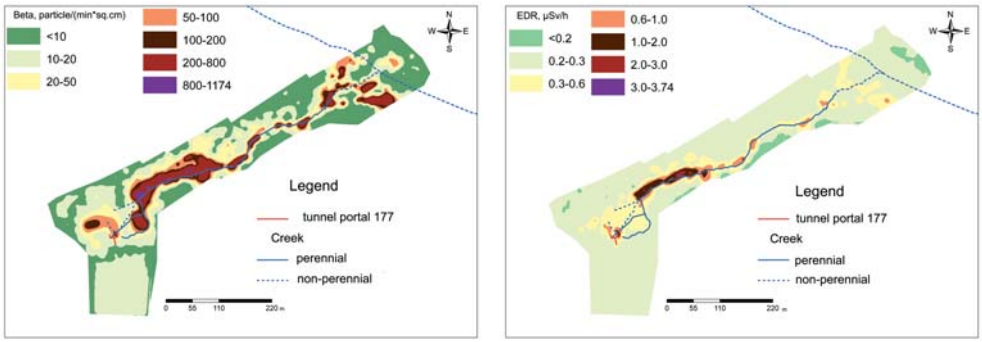


Figure 4. Schematic map of distribution of β-flux density (a) and EDR (b) in the stream from the tunnel No.177

2.3. Character of redistribution of γ- and β-emitting radionuclides

As the main γ-emitting radionuclide is ^{137}Cs and β-emitting radionuclide is ^{90}Sr, the ratio of β-contamination to EDR enables to estimate the ratio of ^{90}Sr amount to that of ^{137}Cs. The distribution of ratios of β-flux density to EDR shows that at the outcome of stream from tunnel No. 176 the main source of contamination of the daylight surface is gamma-emitting radionuclides (Figure 5). Downstream the ratio of their amount to beta-emitting radionuclides decreases. The same distribution is observed in the direction from the riverbed center to the banks. In the ecosystem of tunnel No. 177 stream, the main sources of contamination along the riverbed are beta-emitting radionuclides.

The above ratios show that at the stream outlet from tunnel No. 176 the prevailing radionuclide is ^{137}Cs giving the main contribution to gamma radiation, whereas in the lower part of the stream the prevailing radionuclide is ^{90}Sr – the main beta-emitter. In the ecosystem of tunnel No. 177 ^{90}Sr may prevail along the entire riverbed. These preliminary conclusions are confirmed by quantitative data characterizing radionuclide distribution in the riverbed soils of studied streams given below.

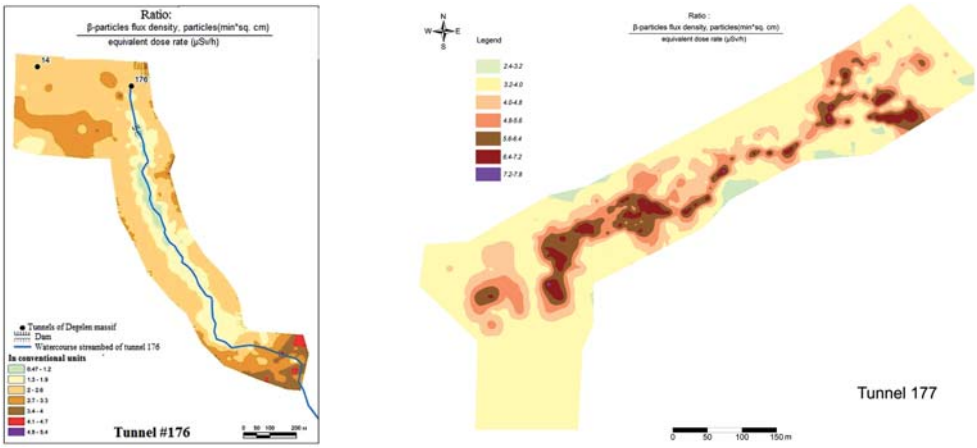


Figure 5. Redistribution of γ- and β-emitting radionuclides at the heads of tunnels Nos.176 and 177

2.4. Investigation of radioactive contamination of components of ecosystems of streams from tunnels of the Degelen mountain massif

2.4.1. General methodology

In order to obtain quantitative factors characterizing the level of radioactive contamination of soil cover of the right tributary of the stream Baitles, 44 soil sections making 9 section lines and 115 soil samples were taken along the genetic horizons (Figure 6, a). Of the above sections 15 sections (5 section lines) (Figure 6, b) characterize ecosystem of tunnel No. 176 stream.

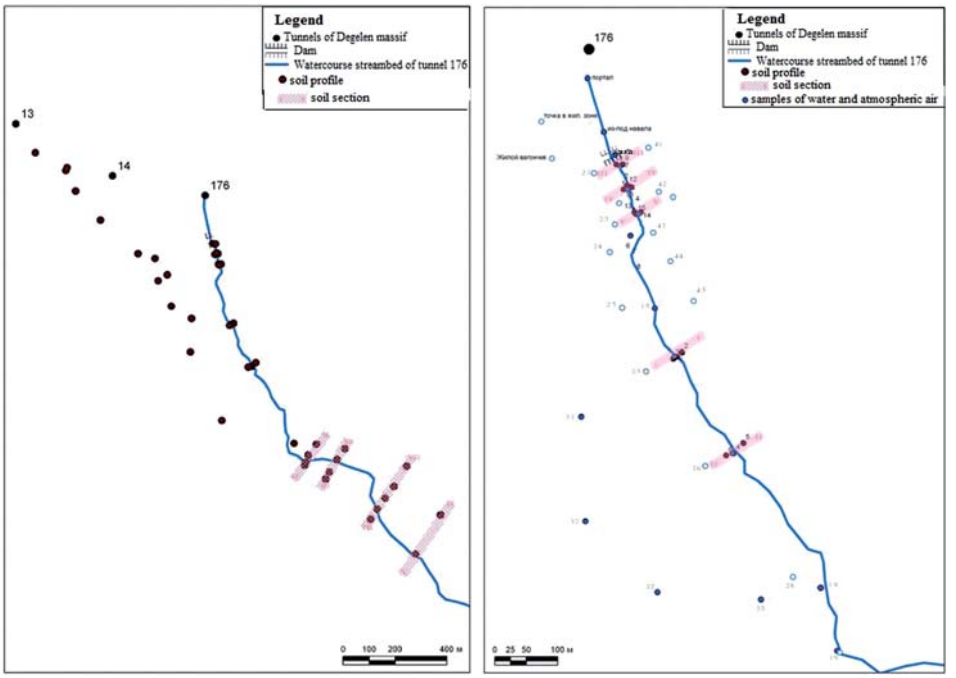


Figure 6. Scheme of location of soil sections in the ecosystem of tunnel No. 176 stream.

The section lines were made perpendicular to the water or stream flow and consist of three sections in the area of detailed investigations and 4-5 section lines (section lines VI-IX) in the lower part of the right tributary of the stream Baitles, which is the continuation of the stream.

For more detailed assessment of artificial radionuclide concentration in surface horizons of studied soils in 5 central sections of the tunnel No. 176 ecosystem, soil samples were taken in the intervals: 0-5, 5-10, and 10-15 cm.

To obtain characteristics of the soil cover of tunnel No. 177 ecosystem located in the right tributary of the stream Uzynbulak, 14 sections were made in the first year of investigations, 11 sections make three section lines perpendicular to the riverbed (Figure 7), 4 sections continue the row in the dry riverbed.

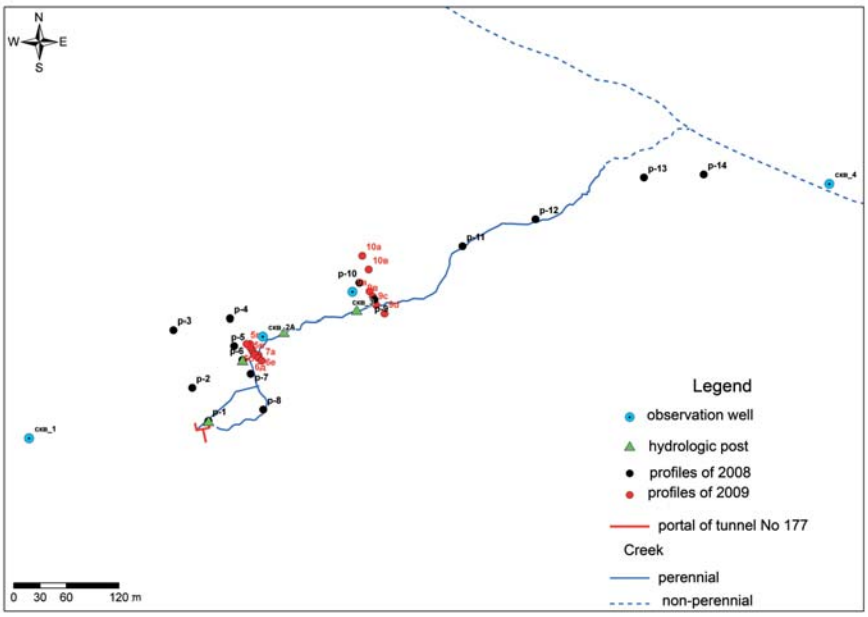


Figure 7. Scheme of soil sections in the ecosystem of tunnel No. 177

To obtain more detailed radiation characteristics of soils in the stream ecosystem, 1 additional section in the section line II-II and 6 sections in the section line III-III were made (Figure 7). Soil samples were taken with a 5-cm interval from the surface up to a depth of 25-50 cm. In addition, 2 sections to a depth of 120-150 cm were made. The interval between sections in section lines with account for additional sections is 1.5-2.0 m.

Soil sections were made to the depth of soil-forming rocks, and water samples were taken at the level of subsoil flow.

2.4.2. Soil cover characteristics

Based on the above data it should be noted that soils of the two examined areas are meadow-type soils. The vegetation cover consists of meadow motley grass. The soils range from undeveloped to insufficiently developed, soil thickness in the riverbed is not more than 40–80 cm and 20–40 cm in the riverbanks.

The total humus concentration in topsoil horizons presented by sod (0–7, 0–10 cm) ranges from 3.2 to 23.7 %, with average values within 8–15 %. Maximal humus quantities were detected on the riverbanks where soil has the highest moistening. In the second layer from the surface horizon the amounts of organic substance sharply decrease and rarely exceed 10 %. At the level of soil-forming rocks this value is about 1 %. Reduction in humus amount in soils as the distance from the water source increases is primarily caused by the weaker topsoil moistening.

In the conditions of washing regime soils do not have carbonate salts (CO_2 of carbonates) and the amount of easily dissolved salts is mainly not higher than 0.1 %. However, if the water mirror of subsoil waters is not far from the surface, which is registered on the banks of tunnel No. 177 stream (sections 2, 6, 7), salts can accumulate in the surface hori-

zon, which enables to refer them to low-brackish soils. pH of soil water suspension is rarely neutral, more often it is low-alkaline or alkaline. Cation Ca prevails in the soil-consuming complex. Its value in the upper horizons of loamy soils is 50 milligram-equivalent per 100 g of soil. In case of lighter mechanical composition of soils, which is observed in the vicinity of stream from tunnel No. 176, concentration of exchange cation Ca decreases to 25–28 milligram-equivalent per 100 g of soil. Mg concentration is about 10 % of that of Ca, but Ca concentration sharply decreases with depth whereas Mg concentration changes more slowly, which gives the other ratio of exchange cations at higher depths.

According to mechanical composition, the studied area of the stream ecosystem of tunnel No. 176 is mainly presented by light loamy and loamy sand soils, more rarely medium-loamy soils, whereas the main valley of the Baitles stream and the vicinity of tunnel No. 177 stream have prevailing medium- and heavy loams among loose sediments. It is caused by lower slopes and, as a result, weaker drainage of the area. These facts enable to conclude that concentration of silt particles in the meadow soils of the 176 tunnel ecosystem is within 3–8 %, whereas in the places less subjected to washing-out silt content may be as high as 26–32 %. All soil profiles of the studied meadow soils contain large amount of fragmental material: gravel, coarse sand and large-sized dust.

Therefore, the decrease in slope and, hence, stream flow causes changes in natural drainage of the area, which in turn causes changes in soil characteristics: the increase in the number of fine-dispersed particles and fine humus, and accumulation of easily dissolved salts in the surface horizons.

2.4.3. Formation of topsoil radionuclide contamination in the ecosystems of tunnel streams

2.4.3.1. Radionuclide distribution in riverbed soils of studied streams

Maximal radionuclide concentrations in the topsoil layer of the studied ecosystems are registered at some distance from tunnel portals. For tunnel No. 176 maximum is registered at a distance of about 170 m from the portal, for tunnel No. 177 this distance is about 100 m (Figure 8). In both cases maximal concentrations are observed in the places where the stream flows out from the heap of fragmental material, which shows its role as a barrier for streams.

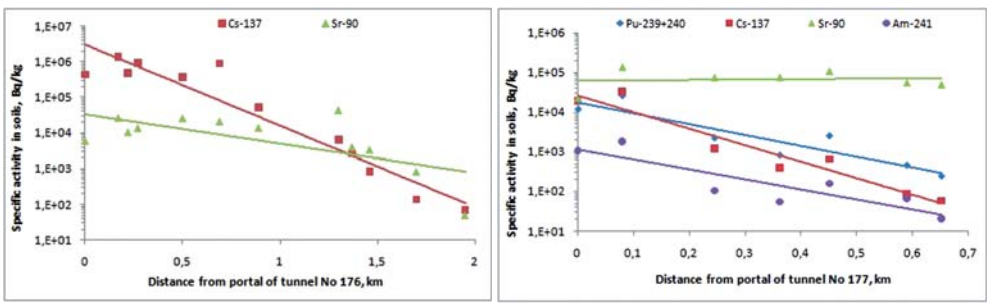


Figure 8. Radionuclide distribution in riverbed soils of studied streams

Accumulation of radionuclides in such zones may depend on the presence of various barriers created by microrelief (hollows, rock terraces, impoundments, etc.),

sorption (confluence of structures composed of rocks of different mechanical composition), biogeochemical (formed by different vegetation layers) and other barriers.

^{137}Cs concentration in the riverbed soils of streams from tunnels No. 176 and 177 is 3 times lower than the maximal concentration in the upstream at a distance of 1300 and 665 m from tunnel portals, respectively. $^{239+240}$Pu concentration in the riverbed soils of streams from tunnel No. 177 decreases 3 times at a distance of 300 m from the tunnel portal. A different picture is observed for ^{90}Sr distribution in the meadow soils of both ecosystems. In the soils of the 176 tunnel ecosystem, ^{90}Sr concentration decreases to almost background values at a distance of 2 km from the portal (^{137}Cs – 70 Bq/kg, ^{90}Sr – 50 Bq/kg). ^{90}Sr concentration in the riverbed of the 176 tunnel stream decreases slowly and at a distance of 650 m is an order of magnitude lower than its maximal value.

The curve of ^{241}Am distribution is practically the same as the curve of $^{239+240}$Pu distribution. Of interest is a slower change in the specific activity of $^{239+240}$Pu and ^{241}Am as compared to that of ^{137}Cs. $^{239+240}$Pu has lower specific activity in soil around the tunnel portals but higher migration ability, though it was believed that migration ability of $^{239+240}$Pu was lower than that of radionuclides ^{90}Sr and ^{137}Cs. In our case we get the following descending series: ^{90}Sr > $^{239+240}$Pu ≥ ^{241}Am > ^{137}Cs. It means that mobility of radionuclide $^{239+240}$Pu in soils of the tunnel 177 ecosystem is higher than that of ^{137}Cs, and it is transferred not only with surface waters. Recent investigations on the state of artificial radionuclides [23] showed that in tunnel waters of tunnel 177 up to 50–60 % of $^{239+240}$Pu are in ionic form, which explains their mobility. The subsoil flow brings a considerable amount of $^{239+240}$Pu and ^{90}Sr to the main riverbed of the Uzynbulak stream.

The figure (Figure 9) shows radionuclide distribution in the riverbed soils of streams from tunnels No. 176 and 177 as a function of distance from tunnel portals. The distributions of relative radionuclide concentrations are given as fractions of maximal values at various distances from the riverbed.

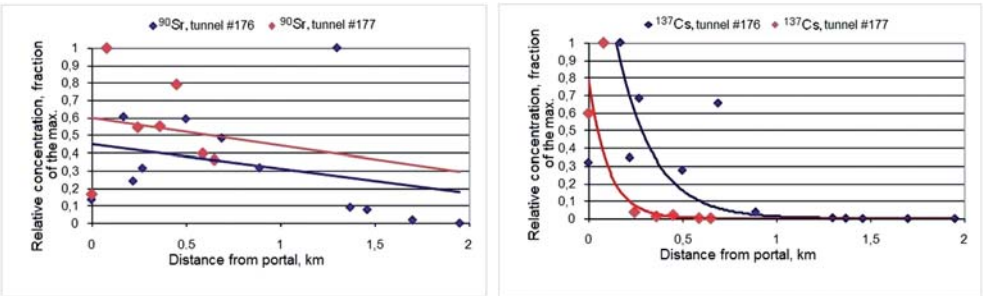


Figure 9. Radionuclide distributions in meadow soils of studied ecosystems as a function of distance from tunnel portals in fractions of maximal concentrations in meadow soils of studied ecosystems

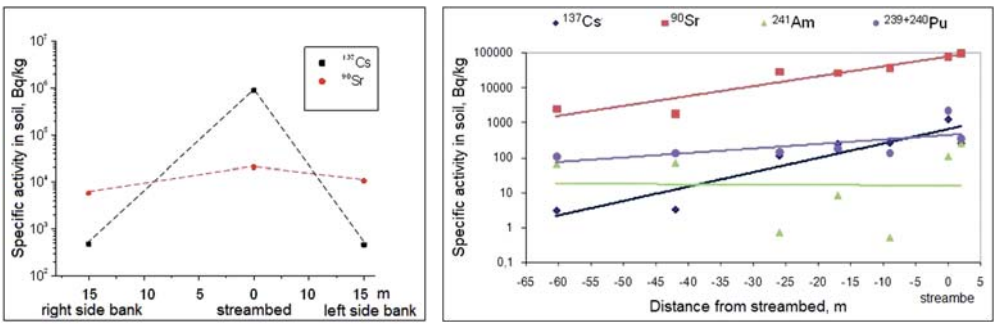
The results obtained show wide discrepancy in the values of ^{90}Sr relative concentrations, but even this does not prevent to see similarity in the character of ^{90}Sr distribution in the ecosystems of two tunnels (Figure 9). The lines representing ^{90}Sr distribution in the meadow soils of studied systems as a function of distance from portals are practically two parallel lines. The functions describing the process in both cases are also practically the same: y = -0.15x + 0.60 for tunnel No. 177 and y = -0.14x + 0.45 for tunnel No.176.

In both cases the ^{137}Cs distribution shows sharp decrease in radionuclide concentrations with increase in the distance from tunnel portals.

Such generalization of the results for streams from tunnels with water outcomes enables to predict behaviour of radionuclides in meadow soils of the other similar in genesis ecosystems of the Degelen mountain massif. Based on the lines showing radionuclide behaviour in horizontal directions it is possible to predict that at the present time ^{137}Cs will not spread farther than 1.5 km from the outcome to the daylight surface and ^{90}Sr will spread not farther than 2 km.

2.4.3.2. Radionuclide distribution in soils perpendicular to riverbeds

Radionuclide distribution in soils perpendicular to the riverbeds depends on the flow of surface and subsoil waters, which are sources of contamination. The largest spreading area in the water flow ecosystem due to its mobility and solubility is occupied by ^{90}Sr. The width of its distribution is formed both by surface and subsoil waters (Figure 10). Therefore its spreading depends on valley width and thickness of loose sediments in it.



a b

Figure 10. a) surface distribution of radionuclides (tunnel No. 176);
b) surface distribution of radionuclides in the section line IV-IV (tunnel No. 177)

The area of ^{137}Cs distribution is only formed by surface flooding and infiltration from streams at a distance not more than 10 m (Figure 10). $^{239+240}$Pu and ^{241}Am distributions are much wider as they are more mobile in the water-saturated environment.

The distributions of radionuclides $^{239+240}$Pu and ^{241}Am with distance from the tunnel portals and perpendicular to the riverbeds have some paradoxical features. The distributions of $^{239+240}$Pu and ^{241}Am specific activity in the central riverbed soils as a function of the distance from the portal are similar, the values lie close to each other, whereas radionuclide distributions perpendicular to riverbeds reveal certain changes in $^{239+240}$Pu to ^{241}Am ratios in different points at a distance from the riverbed center.

2.4.4. Formation of vertical radionuclide contamination in soil profile

As a whole, the vertical radionuclide distribution in meadow soils follows common tendencies established on the STS [24], i.e. reduction in concentration with depth. However, some places have specific features depending on their distance from water sources. The figure (Figure 11) shows summarized graphs of radionuclide concentrations in different parts of the ecosystem. Generalization was made for 6 sections located in the riverbed and 6 sections located at a distance not more than 2 m from the riverbed (riverbank zone).

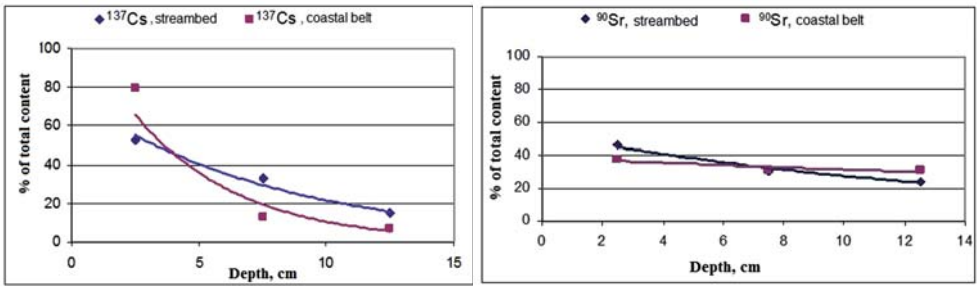


Figure 11. Vertical radionuclide distribution in the soil profile as a function of distance from the riverbed: central riverbed and riverbank zone (not farther than 2 m from the riverbed)

The figure (Figure 11, a) shows that ^{137}Cs concentration in riverbed soils of tunnel streams is distributed over the soil profile with slow decrease. In the riverbank zone, at a distance of 2–3 m from the central riverbed a much sharper decrease in the ^{137}Cs concentration with depth than in the riverbed is registered. This result may be explained by infiltration in the riverbed. Therefore vertical distribution of ^{137}Cs radionuclide in the soil profile depends on the degree of moistening, which in the studied conditions depends on the distance of observation points from the stream.

The amount of ^{90}Sr, both in the central riverbed and in the riverbank zone, decreases with depth of soil cover (Figure 11, b). The ^{90}Sr distributions in profile in the riverbed and in the riverbank zone show little difference but the riverbed has higher values in the upper horizon which is explained by the closeness of the source of contamination – a radioactively-contaminated stream.

Vertical distributions of $^{239+240}$Pu and ^{241}Am in the soil profile practically do not differ. Almost 95% of the radionuclides are in the upper soil layer (Figure 12), and to a depth of 25 cm their amount sharply decreases.

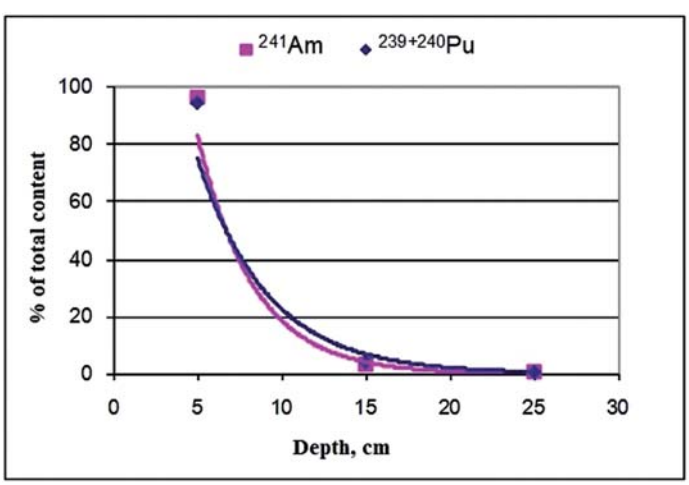


Figure 12. Distributions of $^{239+240}$Pu and ^{241}Am radionuclides in the vertical soil profile (based on 3 sections)

Therefore the deepest penetration in soil profile is registered for ^{90}Sr, the shorter is the penetration depth for ^{137}Cs and the least mobile in soil vertical are $^{239+240}$Pu and ^{241}Am.

Vertical distribution of the main artificial radionuclides in the water-saturated soil in the central part of the ecosystem of the tunnel 177 stream also confirms the deepest penetration of ^{90}Sr (Figure 13). High specific activity of $^{239+240}$Pu in water-saturated soil is observed to a depth of 90 cm (>1,000 Bq/kg). Such high mobility of $^{239+240}$Pu is explained in the same way as in the case of horizontal distribution i.e. 50–60 % of $^{239+240}$Pu in tunnel waters of tunnel No. 177 are in ionic form [23].

^{241}Am concentrations exceeding 100 Bq/kg are registered at depths from 100 to 120 cm (Figure 13, a).

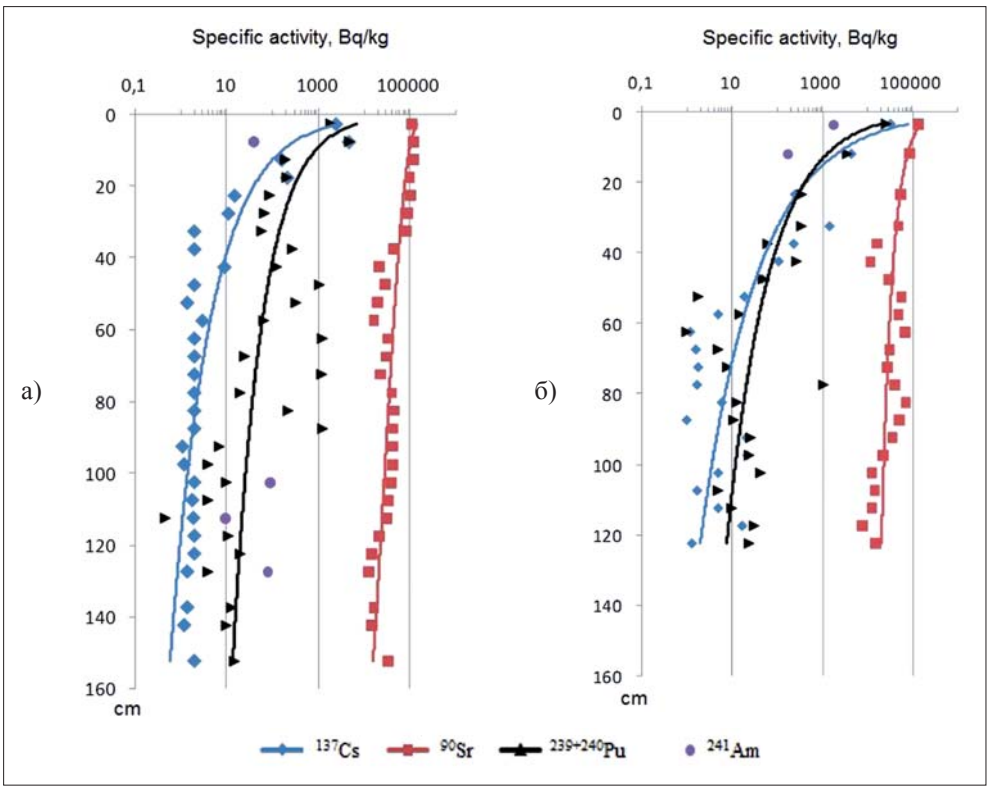


Figure 13. Vertical radionuclide distribution in the riverbed soils of stream from tunnel No. 177. a) section 6, b) section 6a.

2.5. Specific features of radioactive contamination of surface and subsoil waters in the ecosystems of tunnel waters of the Degelen mountain massif

2.5.1. Methodology of investigations

Investigation of surface waters in the ecosystems included studying of hydrological and hydrochemical regimes of streams and dynamics of radionuclide concentration in the stream

water from its outcome from the tunnel to its disappearance. Studying of hydrochemical regime included studying of concentration of artificial radionuclides in different parts of the areas.

2.5.1.1. Investigation of hydrological regime of studied streams

Observations of hydrological regime of stream from tunnel No. 176 included daily measurements of the surface water discharge. For this purpose a metallic gutter 2 m long, 15 cm wide and 20 m high was mounted at the outcome of tunnel No. 176. Water discharge was measured in the gutter. The gutter was used to measure geometrical sizes of the stream and its speed, and to calculate stream discharge.

To determine discharges of stream from tunnel No. 177, hydrological stations were made in different places along the stream according to the scheme (Figure 7), and to make precise measurements the riverbed geometry was flattened. Observations of the hydrological stream regime consisted in daily measurements of water discharge in hydrological stations. As water outcome to the daily surface in the vicinity of portal of tunnel No. 177 was very small (<6 l/min), water discharge on the hydrological station No. 1 was not measured.

2.5.1.2. Methodology of studying dynamics of radionuclide concentration in stream water

To study dynamics of concentrations of artificial radionuclides in water of stream from tunnel No. 176, every three days in the points of water outcome water samples for ^{137}Cs and ^{90}Sr concentration were taken and every 12 days samples – for $^{239+240}$Pu concentration. In the stream from tunnel No. 177, samples were taken weekly in the hydrological posts.

2.5.1.3. Methodology of studying radionuclide distribution along the stream from upper to lower reaches

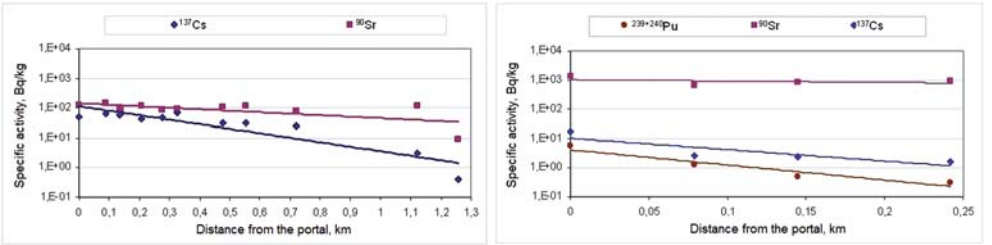
To study spatial radionuclide transfer with water from tunnel No.176 the profile – portal, heap, dam (1/1-1/9) (Figure 6, b) was made. In order to make γ-spectrometric and radiochemical analyses, two-run water samples were taken in each point of the profile with a 2-month interval. Spatial radionuclide transfer with water from tunnel No.177 was studied by water sampling at the hydrological posts.

2.5.2. Results

2.5.2.1. Specific features of radionuclide behaviour in surface waters of studied streams from tunnels

Studying of radionuclide behaviour in tunnel streams from "Degelen" test ground enables to discover characteristics of radionuclide redistribution with water flow.

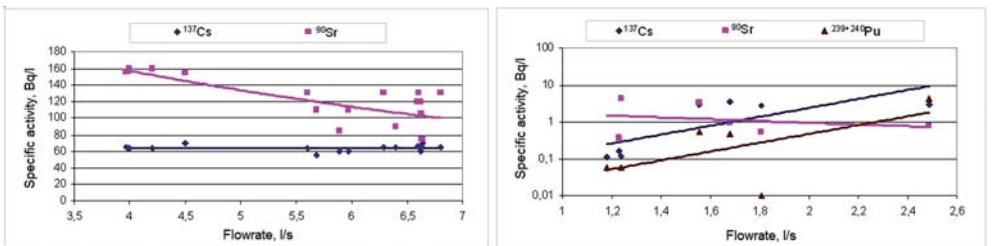
Concentrations of artificial radionuclides decrease as the distance from the tunnel portal increases. The lines describing decrease in concentrations of radionuclides $^{239+240}$Pu and^{137}Cs in the stream water as the distance from the portal increases show sharp reduction as compared with lines corresponding to ^{90}Sr (Figure 14). For example, at a distance as far as 250 m from the portal, concentrations of $^{239+240}$Pu and ^{137}Cs in the stream water from tunnel No. 177 are close to the lower limits of the measuring equipment, whereas ^{90}Sr concentration does not decrease considerably (Figure 14). The presence of ^{137}Cs (close to the lower limits of the measuring equipment) in the water from tunnel No. 176 is registered at a farther distance from the portal – 1250 m (Figure 14).



Tunnel # 176 Tunnel # 177

Figure 14. Horizontal distribution of radionuclides in surface waters of studied ecosystems as a function of distance from tunnel portals

Concentrations of radionuclides in streams in places where they appear on the daylight surface also depend on water discharge. It was registered that ^{90}Sr concentration in water of both streams decreases as discharge increases, though this dependence is stronger for tunnel No. 176. For example, in this tunnel correlation coefficient between water discharge and Sr concentration was r = -0.76, and for tunnel No. 177 it was r = -0.42. Each stream has different dependences of ^{137}Cs concentration on water discharge. Tunnel No. 176 does not show any noticeable dependence of concentration on water discharge (r = 0.04), whereas tunnel No. 177 gives a direct dependence i.e. increase in water discharge causes increase in ^{137}Cs concentration (r = 0.88), the same dependence is observed for $^{239+240}$Pu (r = 0.87) (Figure 15).



Tunnel # 176 Tunnel # 177

Figure 15. Radionuclide concentrations in water of studied streams as a function of water discharge

Such inconsistent dependencies of concentration on water discharge on the studied objects may be caused by different hydrogeological characteristics of the objects. The water from tunnel No. 176 has only one outcome near the portal, and it is an outlet pipe from the tunnel cavity. The water from tunnel No. 176 goes out from many points as all hill foot where the tunnel is located is drained. Hence, an increase in water discharge may cause additional inflow of ^{137}Cs and $^{239+240}$Pu from uncontrolled water outcomes.

2.6. Formation of radionuclide contamination of ground waters in the ecosystem of tunnel No. 177

2.6.1. Methodology of investigations

In order to determine radionuclide contamination of ground waters in the vicinity of tunnel No. 177 within the stream ecosystem, points for drilling observation boreholes

were chosen (Figure 7). 4 boreholes were drilled, 2 of them determine inflow of radioactive contamination from the closest tunnels in the valley (borehole 1) and inflow of stream from tunnel No. 177 to the main riverbed of the Uzynbulak stream (borehole 4). Boreholes 2 and 3 determine radionuclide contamination in the middle of the stream.

2.6.2. Results of investigations

Specific activity of radionuclides in ground waters of the ecosystem of tunnel No. 177 is given in the table (Table 1).

Table 1.

Radionuclide concentration in ground waters of ecosystem of tunnel No. 177

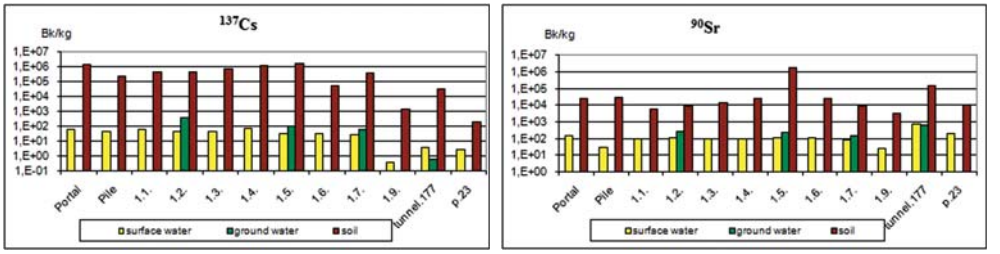
Sampling point	Specific activity of radionuclides (Bq/kg)				
	^{137}Cs	^{241}Am	^{90}Sr	$^{239+240}$Pu	^{3}H
well 2	0.6±0.2	<0.17	670±110	<0.01	(3.1±0.3)*10^{5}
well 3	<0.18	<0.17	6±2	<0.01	(3.0±0.3)*10^{5}
well 4	0.8±0.3	<0.17	3450±550	<0.46	(2.7±0.3)*10^{5}
Note: no water was revealed in the well 1 at the time of sampling.					

^{137}Cs concentration in ground waters in the studied area did not change considerably (from < 0.18 to 0. Bq/kg). ^{90}Sr concentration in subsoil waters changes dramatically (0.006–3.5)*10^{3} Bq/kg. In the valley of the main riverbed of the Uzynbulak stream (borehole 4) ^{90}Sr concentration was 5 times higher than the initial value. The most probable explanation of this result is that borehole No. 4 crosses the subsoil flow with higher concentration of ^{137}Cs and ^{90}Sr.

It is not uninteresting to consider ^{3}H distribution in different types of waters. For example, in the subsoil water samples from boreholes Nos. 2 and 3, specific activity of ^{3}H has the values comparable with those in surface waters, which indicates their close interrelation. Lower ^{3}H values in the borehole No. 4 are most probably caused by inflow of "clean" with respect to ^{3}H flux of ground waters from the upper reaches of the Uzynbulak stream to the place of its confluence with the stream from tunnel No. 177.

2.7. Redistribution of radionuclides in the system surface waters – ground waters – soil

Ratios of radionuclide concentrations in different media but in the same sampling point show that lowest radionuclide concentrations are present in surface waters (^{137}Cs from 0. 41 to 68 Bq/kg, ^{90}Sr – from 9 to 155 Bq/kg), a little higher concentrations are registered in subsoil waters (^{137}Cs – 57 to 480 Bq/kg, ^{90}Sr – 155 to 270 Bq/kg), maximal values are registered in topsoil horizons (^{137}Cs – 5.1*10^{4} to 1.4*10^{6} Bq/kg, ^{90}Sr – 9*10^{3} to 2.6*10^{4} Bq/kg) (Figure 16).



a b

Figure 16. Concentrations of radionuclides ^{137}Cs and ^{90}Sr in soil, surface and subsoil waters of the 176 tunnel stream ecosystem

The results of conjugate analysis give different values of radionuclide concentrations is surface, subsoil waters and soils, which, on the one hand, indicates their sorption by soil and, on the other hand, reveals nonequilibrium of their state in the system.

Specific activity in components of the ecosystem of tunnel streams is used to calculate ratios of radionuclide specific activities in the stream water to that in soil in the same point (Figure 17). An analysis of ratios of ^{137}Cs specific activities in different environments measured along the stream from tunnel No. 176 from the portal to the confluence with the right tributary of the Baitles stream shows that most values range within two orders of magnitude – 10^{-4} and 10^{-5}. The ratios of ^{90}Sr specific activities are within one order of magnitude – 10^{-3} (8 out of 11 cases) (Figure 17).

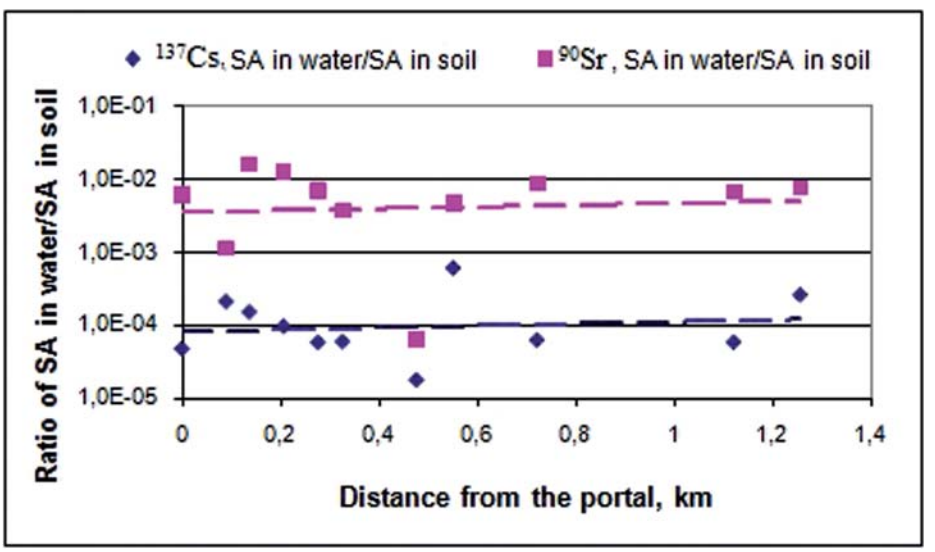


Figure 17. Ratio of specific activity of ^{137}Cs and ^{90}Sr in the stream water from tunnel No. 176 to specific activity of radionuclides in soil

Additional points from the stream from tunnel No. 177 and upper reaches of the Baitles stream do not change the picture (Table 2).

Table 2.

Ratio of radionuclide specific activity in water from tunnel No. 176 to radionuclide specific activity in soil

Tunnel #	Ratio of specific activity of radionuclides in water to specific activity of radionuclides in soil											
	^{90}Sr				^{137}Cs				$^{239+240}$Pu			
	min	max	average	N	min	max	average	N	min	max	average	N
176	$6.3*10^{-5}$	$1.6*10^{-2}$	$6.6*10^{-3}$	11	$1.8*10^{-5}$	$6*10^{-4}$	$1.5*10^{-4}$	11	-	-	-	-
177	$5.4*10^{-3}$	$1.8*10^{-2}$	$8.6*10^{-3}$	3	$1.1*10^{-4}$	$1.3*10^{-3}$	$7*10^{-4}$	2	$1*10^{-4}$	$8.1*10^{-3}$	$9.1*10^{-4}$	2

An analysis of ratios of maximal fixed values of radionuclide specific activity in water at the outcome from tunnel portals to maximal fixed values of radionuclide specific activity in soil of the near-portal tunnel areas showed that the ratios for radionuclide ^{137}Cs vary within 10^{-2}–10^{-5} with average value 10^{-3} (Table 3). The ratios for radionuclide $^{239+240}$Pu vary within 10^{-2}–10^{-6} with average value 10^{-3}. The ratios for ^{90}Sr vary within 10^{-1}–10^{-3} and the average value is 10^{-2}. The statistics for ^{241}Am is not sufficient with the average value equal to 10^{-4}.

Based on ratios of maximal radionuclide specific activities in soil of the ecosystem of tunnel streams and maximal radionuclide specific activities in water of tunnel streams, ratios of radionuclide specific activities in water to that in soil were obtained (Table 3).

Table 3.

Ratios of maximal radionuclide specific activities in water to radionuclide specific activities in soil of the near-portal grounds

Tunnel #	Maximal specific activities at near-portal areas for soil (Bq/kg), water (Bq/l) and ratios of specific activities water/soil											
	^{137}Cs			^{90}Sr			$^{239+240}$Pu			^{241}Am		
	soil	water	ratio	soil	water	ratio	soil	water	ratio	soil	water	ratio
802	$1*10^{3}$	22	$2.2*10^{-2}$	610	71	$1.2*10^{-1}$	832	0.258	$3.1*10^{-4}$	-	-	-
203	$1.3*10^{4}$	5	$3.8*10^{-4}$	330	11	$3.3*10^{-2}$	379	<0.02	$<5.3*10^{-5}$	-	-	-
177	$5.2*10^{4}$	65	$1.3*10^{-3}$	$1.3*10^{5}$	$2.1*10^{3}$	$1.6*10^{-2}$	$1.7*10^{4}$	2.49	$1.5*10^{-4}$	$2.4*10^{3}$	2.6	$1.1*10^{-3}$
163	646	2	$3.1*10^{-3}$	$1.8*10^{5}$	930	$5.2*10^{-3}$	134	-	-	-	-	-
179	64	-	-	-	185	-	200	0.05	$2.5*10^{-4}$	-	-	-
607	218	<0.6	$<2.8*10^{-3}$	42	-	-	$7.6*10^{3}$	0.043	$5.6*10^{-6}$	-	-	-
11	6*105	26	$4.3*10^{-5}$	$9.9*10^{4}$	-	-	116	-	-	-	-	-
165	$2*10^{6}$	495	$2.5*10^{-4}$	$1.3*10^{5}$	$1.5*10^{3}$	$1.2*10^{-2}$	79	0.88	$1.1*10^{-2}$	1200	<0.8	$<6.7*10^{-4}$
176	$1.7*10^{6}$	110	$6.1*10^{-5}$	$5.3*10^{4}$	490	$9.2*10^{-3}$	700	0.05	$7.1*10^{-5}$	-	<1.1	-
609	$6.5*10^{6}$	359	$5.5*10^{-5}$	$1.8*10^{4}$	730	$4.1*10^{-2}$	729	0.9	$1.2*10^{-3}$	-	<0.8	-
average	-	-	$3.3*10^{-3}$	-	-	$3.4*10^{-2}$	-	-	$1.6*10^{-3}$	-	-	$8.8*10^{-4}$

As a whole, the obtained ratios (Table 2, Table 3) enable to make estimation of maximal possible radionuclide concentration in soils of stream ecosystems based on radionuclide specific activities in water of the same streams.

Therefore, the above investigations enable to reveal specific features of radionuclide behaviour in components of the ecosystem of streams from tunnels Nos. 176 and 177. The

ratio of β-flux density to EDR enables to conclude that in the initial part of the stream from tunnel No. 176 the prevailing radionuclide is ^{137}Cs giving the main contribution to gamma-radiation, at the end of the stream the prevailing radionuclide is ^{90}Sr, the main beta-emitter. These preliminary conclusions were confirmed by the qualitative data characterizing radionuclide distribution in the riverbed soils.

It was established that radionuclide $^{239+240}$Pu has higher mobility than ^{137}Cs, both in transfer in horizontal direction along and perpendicular to the riverbed and to the depth of soil profile. Migration of ^{137}Cs in space is limited by a small distance and water flow. ^{90}Sr as the most soluble and mobile in water radionuclide distributes in space at rather large distances from the source of contamination. It has also been found out that radionuclide concentration in soil can be evaluated by specific activity of these radionuclides in water.

Different values of radionuclide concentration in the adjoint elements of the ecosystem (surface waters – subsoil waters – soil) indicate their non-equilibrium state in the system.

3. STUDIES OF RADIONUCLIDE CONTAMINATION OF MEADOW SOILS IN THE STREAM VALLEYS OF THE DEGELEN MOUNTAIN MASSIF

3.1. Investigation of radionuclide contamination of meadow soils in the stream valleys of the Degelen mountain massif

In studying migration of artificial radionuclide in ecosystem an important role is played by their motion in valleys from the upper reaches of streams to the final discharge zone. In case of water inflow from tunnels the process develops due to the flux strength in the stream. In this work the objects of investigations were valleys of the largest streams of the Degelen mountain massif – Uzynbulak, Baitles, Toktakushik and Karabulak, on the area of which underground nuclear tests of nuclear weapons in tunnels were made.

3.1.1. General methodology of investigations

In order to obtain initial data on radioecological situation in the stream valleys and to discover possible sources of radioactive contamination it was necessary to make a preliminary analysis of available radioecological data from archives of the IRSE, NNC RK. The archives contained data on the near-portal areas of tunnels including information on streams from tunnels in the studied valleys, water discharge in streams and radiation characteristics of the near-portal areas and water of tunnel streams. The archive data were taken into account in studying processes of radionuclide migration in valleys from the stream upper reaches to the final discharge zone. To study the process longitudinal profiles were made along the riverbeds of streams Uzynbulak, Baitles, Toktakushik and Karabulak from tunnel portals with and without water outcome. Transverse profiles were made perpendicularly to the valley of the Uzynbulak stream in tributaries from tunnels to the main riverbed (Figure 1).

A longitudinal profile consisting of 7 observation points, stretching from Uzynbulak upper reaches to its outcome from the Degelen mountain massif, was made in the Uzynbulak valley, in the riverbed, 30 m away from the portal of tunnel No. 034. The last observation point is located beyond the mountain massif, 200 m away from the protruding dyke – road. In the Baitles valley the profile consists of 6 observation points. It begins at a distance of 20 m from

the portal of tunnel 609, and the last point is 7–7.5 km away from the circular road beyond the Degelen mountain massif. In the Toktakushik valley the profile consists of 5 observation points, the first is at a distance of 50 m from the portal of tunnel 165, the last is 7 km away from the Degelen mountain massif, in the spreading zone of the stream discharge. In the Karabulak valley a longitudinal profile consisting of 4 observation points was made. One section is located at the beginning of riverbeds of the stream confluents, the third section is in the place of confluence; the last section is in the dry flat riverbed at the exit from the mountain massif.

The following transverse profiles in the Uzynbulak valley were made in the right bank confluent in the upper part of the stream: 20 m away from the portal of tunnel 705, it consists of three observation points; in the middle flow of the Uzynbulak, in the right-bank confluent, 135 m from the portal of tunnel 806, it has 4 observation points; in the lower valley part, in the left-bank confluent, 30 m from the portal of tunnel 126, it has 2 observation points.

It should be noted that not all profiles are made from tunnels with constant or temporary water outcome. For example, profiles in the Baitles and Toktakushik streams are made from tunnels No. 609 and No. 165, having constant water flow. The longitudinal profile in the Uzynbulak begins near the 034 tunnel portal without water flow. However, the confluents of the stream (tunnel No. 177) and the valley sides (tunnel No. 104) have tunnels with constant water flow whose runoff may reach stream riverbed. The transverse profiles in the Uzynbulak valley were made from tunnel portals without water outcome, located in the valley sides (A-A) and in the right-bank confluent of the middle part of the stream (B-B).

3.1.2. Results of investigations

3.1.2.1. Radionuclide contamination of meadow soils in the Uzynbulak stream valley

General characteristic of the Uzynbulak stream valley. The Uzynbulak is the largest water stream of the Degelen mountain massif. The Uzynbulak valley stretching for 20 km has the largest watershed area. About 50 tunnels are adjacent to the stream and its confluents. Atmospheric fallouts accumulate in tunnel cavities and reaching a certain volume form streams going out to the daylight surface. Streams from different tunnels inflow into the main stream and into confluents which flow to the main riverbed in the Uzynbulak valley. Five tunnels (Nos. 173, 181, 203, 802 and 204) have temporary seasonal water flows forming, as a rule, in May-June. Tunnels Nos. 177 and 104 have constant water flows.

Tunnels Nos. 168, 707,164, 163, 034, 162 are located in the Uzynbulak upper reaches (Table 4).

Table 4.

Characteristics of tunnels in the upper Uzynbulak river

Tunnel #	Test date	Explosion yield, kt	Maximal specific activity at near-portal area, Bq/kg (Bq/l in water)				Water outflow
			^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{241}Am	
168	17.07.87 06.02.88	20-150 0.001-20 <0.001 (x2)	$1.2*10^3$	<100	960	<1	no
707	29.03.77	0.001-20 кт (x2)	$4.9*10^5$	$1.8*10^4$	9	18	no

Tunnel #	Test date	Explosion yield, kt	Maximal specific activity at near-portal area, Bq/kg (Bq/l in water)				Water outflow
			^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{241}Am	
164	06.05.87 20.12.87	0.001-20 0.001-20	14	9	98	-	no
163	20.02.75 19.05.76	0.001-20 (x3) 0.001-20	646 (2)	$1.8*10^5$ (930)	134	-	occasionally
034	18.10.88	0.001-20	55	$5*10^4$	520	-	no
162	29.11.78	0.001-20	94	156	62	-	no

Downstream, on the left bank of the main riverbed inflows a confluent where tunnels Nos. 803, 803bis and 141 are located (Table 5).

Table 5.

Characteristics of tunnels at the first left Uzynbulak confluent

Tunnel #	Test date	Explosion yield, kt	Maximal specific activity at near-portal area, Bq/kg				Water outflow
			^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{241}Am	
803	26.12.77 20.12.78	0.001-20 0.001-20 (x2)	38	3	84	57	no
803bis	23.11.84	0.001-20 (x3)	$3.6*10^4$	$1.9*10^3$	-	-	no
141	31.05.79	0.001-20 (x3)	$3.9*10^3$	563	$2.8*10^3$	658	no

Further downstream, on the right bank of the main riverbed inflows a confluent where at the foot of hills the tunnels Nos. 172, 173, 706, 169/1, 177 are located (Table 6).

Table 6.

Characteristics of tunnels of the first right Uzynbulak confluent

Tunnel #	Test date	Explosion yield, kt	Maximal specific activity at near-portal area, Bq/kg (Bq/l in water)				Water outflow, Flowrate
			^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{241}Am	
172	25.12.82	0.001-20 (x2)	$5.4*10^4$	60	310	-	no
173	22.05.80	0.001-20 (x3)	200	$7.3*10^3$	130	-	no
706	25.04.71 30.12.76	90 0.001-20	640	29	390	-	no
169/1	23.11.88	0.001-20 (x2) <0.001	76	97	26	-	no
177	30.03.83	0.001-20	$5.2*10^4$ (65)	$1.3*10^5$ (2098)	$1.7*10^4$ (2.49)	$2.4*10^3$ (2.6)	continuous, *557 l/min
*Note: Flowrate of the waterflow here and below is presented based on studies in 2005.							

Of five the above tunnels, tunnel No. 177 has constant water flow. The distance from the 177 tunnel portal to the stream is 650 m. Radionuclide transfer by the stream from tunnel No. 177 was 880 Bq/min for ^{137}Cs and 1.4*10^{5} Bq/min for ^{90}Sr. Tunnel No. 178 is located downstream the inflow of the confluent in the main riverbed. It does not have water flows.

Tunnels Nos. 147, 901, 132 are located further downstream, in the left confluent (Table 7).

Table 7.

Characteristics of tunnels of the second left Uzynbulak confluent

Tunnel #	Test date	Explosion yield, kt	Maximal specific activity at near-portal area, Bq/kg				Water outflow
			^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{241}Am	
147			28	20	2*10^{3}	-	no
901	19.07.85. 28.12.88	0.001-20 0.001-20 (x2)	25	39	120	-	no
132	09.09.84 18.09.87	0.001-20 (x4) 0.001-20 0.001-20	300	250	760	408	no

No water flows were registered in the tunnels during current STS studies. In the small right-bank confluent there is tunnel No. 705 (Table 8). In the Uzynbulak valley side tunnel No. 181 is located. This tunnel has a temporary water flow inflowing directly into the Uzynbulak stream.

Table 8.

Characteristics of tunnels of the second right Uzynbulak confluent

Tunnel #	Test date	Explosion yield, kt	Maximal specific activity at near-portal area, Bq/kg				Water outflow
			^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{241}Am	
705	28.06.70	0.001-20 0.001-20	1.5*10^{3}	9.7*10^{3}	67	-	no
181	10.04.80	0.001-20 (x2)	37	1.2*10^{3}	4	-	occasional

In the middle flow of the Uzynbulak stream is the third right-bank confluent formed by flood and slope waters in the impact zone of four tunnels (Nos. 806, 174, 179, 708), not having water flows (Table 9).

Table 9.

Characteristics of tunnels of the third right Uzynbulak confluent

Tunnel #	Test date	Explosion yield, kt	Maximal specific activity at near-portal area, Bq/kg (Bq/l in water)				Water outflow
			^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{241}Am	
806	10.07.73.	0.001-20 (x2) <0.001	$4.7*10^3$	$8.5*10^4$	66	-	no
174			34	5.9	690	-	no
179	13.09.74.	0.001-20	64	22 (185)	200 (0.05)	-	no
708	16.08.72.	8	815	30	78	-	no

10 tunnels are located in the last left-bank confluent (Table 10). In the valley of the stream, on its left bank there is tunnel No. 802, which earlier had constant water flow, and tunnel No. 203 with temporary water flow.

Table 10.

Characteristics of tunnels of the fourth left Uzynbulak confluent

Tunnel #	Test date	Explosion yield, kt	Maximal specific activity at near-portal area, Bq/kg (Bq/l in water)				Water outflow, Flowrate
			^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{241}Am	
126	04.04.80.	0.001-20	55	930	11	-	no
802	28.01.70. 21.12.79.	0.001-20 (x3) 0.001-20	$1*10^3$ (22)	610 (71)	830 (0.258)	-	Previously - Continuous, 719 l/min
203	21.09.82. 02.11.83.	0.001-20 (x2) 0.001-20	$1.3*10^4$ (5)	330 (11)	379 (<0.02)	-	occasional
196	25.06.82.	0.001-20 0.001-20	$8.9*10^3$	$1*10^4$	38	32	no
195	10.07.74. 18.07.79.	0.001-20 0.001-20	$8.7*10^5$	$3.4*10^4$	710	231	no
607	01.10.69	0.001-20 (x2)	220 (<0.6)	42	$7.6*10^3$ (0.043)	-	no
194	31.10.78.	0.001-20	$2.1*10^5$	85	$6.8*10^3$	160	no
704	22.04.88.	0.001-20	$8.9*10^4$	26	$1.2*10^4$	99	no
129	26.12.83.	0.001-20	250	140	480	13	no
510	26.06.70.	20-150	76	8.1	660	-	no

Further downstream, tunnels Nos. 807, 187, 709, 190 are directed to the Uzynbulak riverbed from the right slope (Table 11).

Table 11.

Characteristics of tunnels of the right slope of the Uzynbulak stream

Tunnel #	Test date	Explosion yield, kt	Maximal specific activity at near-portal area, Bq/kg				Water outflow
			^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{241}Am	
807	22.03.71.	0.001-20	160	4.3	130	-	no
187	30.06.81.	0.001-20 (x2)	98	41	600	-	no
709	16.05.69. 16.12.74.	0.001-20 0.001-20	160	46	680	-	no
190	15.04.84.	20-150 0.001-20	180	67	$1.6*10^3$	-	no

At the level of the last, fourth left-side confluent from the right side, in a very small confluent there are tunnels Nos. 608, 104, 193, 101, 710. Farther, on the right bank there are tunnels Nos. 208, 201, 204, 205. Tunnel No. 104 has constant water flow, its portal is 1300 m from the riverbed. Annual carry-over of radionuclides by water flow was over 60 kBq/min for ^{137}Cs and over 118 kBq/min for ^{90}Sr. The average annual discharge was 742 l/min.

Radionuclide contamination of meadow soils in the Uzynbulak stream valley. The obtained data show that ^{137}Cs and ^{90}Sr concentrations in soils of transverse profiles in the Uzynbulak valley independent of location of their initial points decrease with relief lowering, i.e. in the direction of the present-day riverbed. For example, the values for the above radionuclides make a series: 12.2 kBq/kg and 6.2 kBq/kg, 65 Bq/kg and 1.8 kBq/kg, 15 Bq/kg and 20 Bq/kg (profile A-A). Hence, it is possible to conclude that radionuclides are transferred from tunnel portals, sources of radioactive contamination, by washing out by surface and subsoil waters.

An analysis of radionuclide concentration in soil samples taken in sections of profile observation points in the Uzynbulak valley gives a different picture of their distribution. ^{137}Cs concentration in surface horizons of meadow soils in profile (A-A) (observation points 2, 3, 4) is not high, not more than 66 Bq/kg (Figure 18). Only in the layer 0–20 cm of section 2, the ^{137}Cs concentration was $1.2*10^4$ Bq/kg. This result can be explained by the fact that the soil profile was made in the near-portal ground, and the sample was presented by radiation-contaminated soil. No further considerable ^{137}Cs transfer from the near-portal ground downstream with surface and subsoil waters, which contributes to radionuclide migration, was registered. ^{90}Sr concentration in the near-portal section of tunnel No. 705 in layer 0–20 cm was $6.2*10^3$ Bq/kg (section 2). However, downstream, in the surface horizon 0–4 cm of the soil section 3 the amount of ^{90}Sr was $1.8*10^3$ Bq/kg, it meant that ^{90}Sr migration was more intensive than that of ^{137}Cs. In section 4, located hypsometrically lower than sections 2 and 3, radionuclide specific activity does not exceed two tens of Bq/kg. $^{239+240}$Pu concentration remains within detection limits along the profile, and no considerable changes are registered.

In profile B-B (observation points 6, 7, 8, 9) ^{137}Cs concentration in the surface layer 0-5 cm is 380 Bq/kg and considerably decreases with distance from point 6 (the first in the profile) making a series: 380, 190, 75 and again 225 Bq/kg in the main Uzynbulak riverbed

(Figure 19). The increase in ^{137}Cs specific activity in soils of section 9 can be explained by carry-over of artificial radionuclides along the riverbed from the upper part of the valley.

Concentration of ^{90}Sr in soils of point 6 is as high as 8.5*10^4 Bq/kg. Farther, in the horizontal profile the amount of ^{90}Sr decreases but it is impossible to construct a continuously decreasing series. It is likely that mobility and solubility of this radionuclide depend on some other factors. It should be noted that ^{90}Sr specific activity even in the main Uzynbulak riverbed, i.e. at the end of the profile, is not low (section 9, horizon 0-6 cm, ^{90}Sr concentration – 660 Bq/kg). In the lower-lying horizons ^{90}Sr concentrations decrease, but to a depth of 40-50 cm remain higher than those for ^{137}Cs. For example, in section 6, located 130 m from the portal of tunnel No. 806 high ^{90}Sr concentration is registered in all horizons, and at a depth of 45 cm this value is as high as 3.3*10^4 Bq/kg. Sometimes maximal specific activity of this radionuclide is registered in the second from surface horizon, which is probably caused by its solubility and mobility (Figure 26). Concentration of $^{239+240}$Pu in all soil sections of the profile remains two tens of units, and does not show any noticeable tendency to changes.

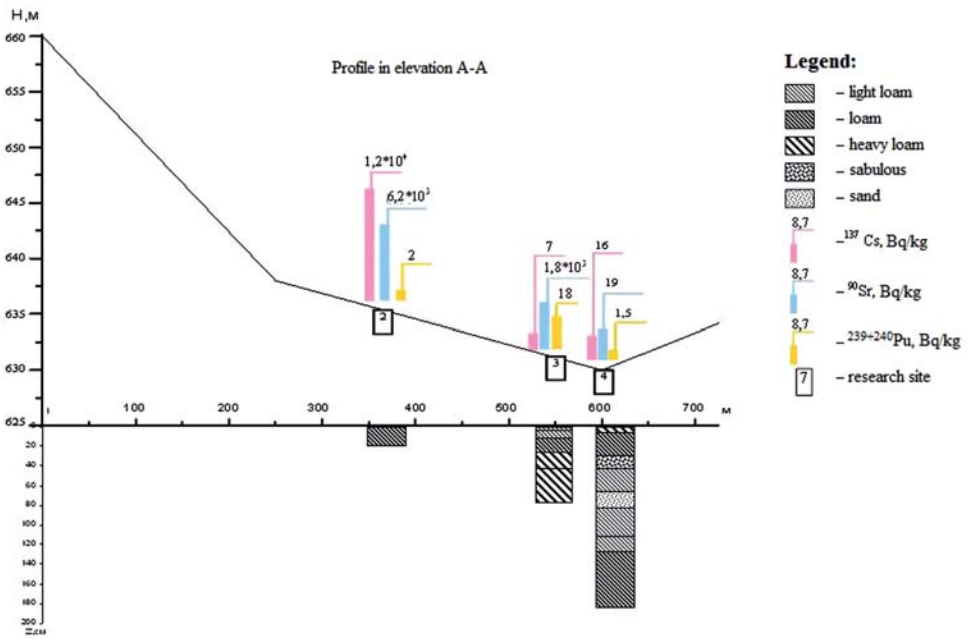


Figure 18. Distribution of artificial radionuclides in soils of A-A profile

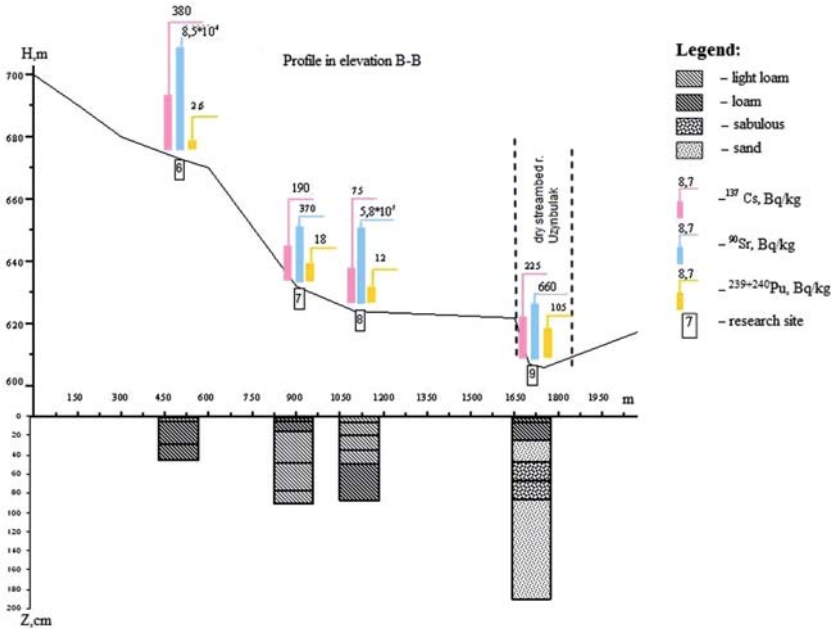


Figure 19. Distribution of artificial radionuclides in soils of B-B profile

Concentration of artificial radionuclide ^{137}Cs in soil samples of transverse profile B-B is rather small (Figure 20). Maximal concentration of 53 Bq/kg is registered in the surface layer 0–3 cm in soil section 11.

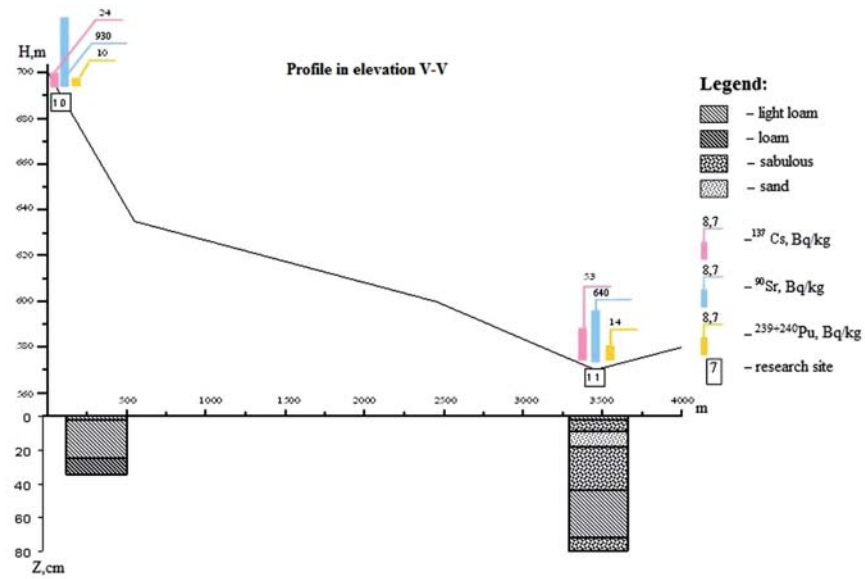


Figure 20. Distribution of artificial radionuclides in soils of C-C profile

^{90}Sr concentration has rather high values. The figure (Figure 20) shows that radionuclide concentration in surface horizon of meadow soils decreases along the confluent from 930 Bq/kg (section 10) to 640 Bq/kg.

An analysis of artificial radionuclide distribution in soil sections in the longitudinal profile stretching from the Uzynbulak upper reaches to the place of final discharge does not show any noticeable transfer of radionuclides downstream (Figure 21).

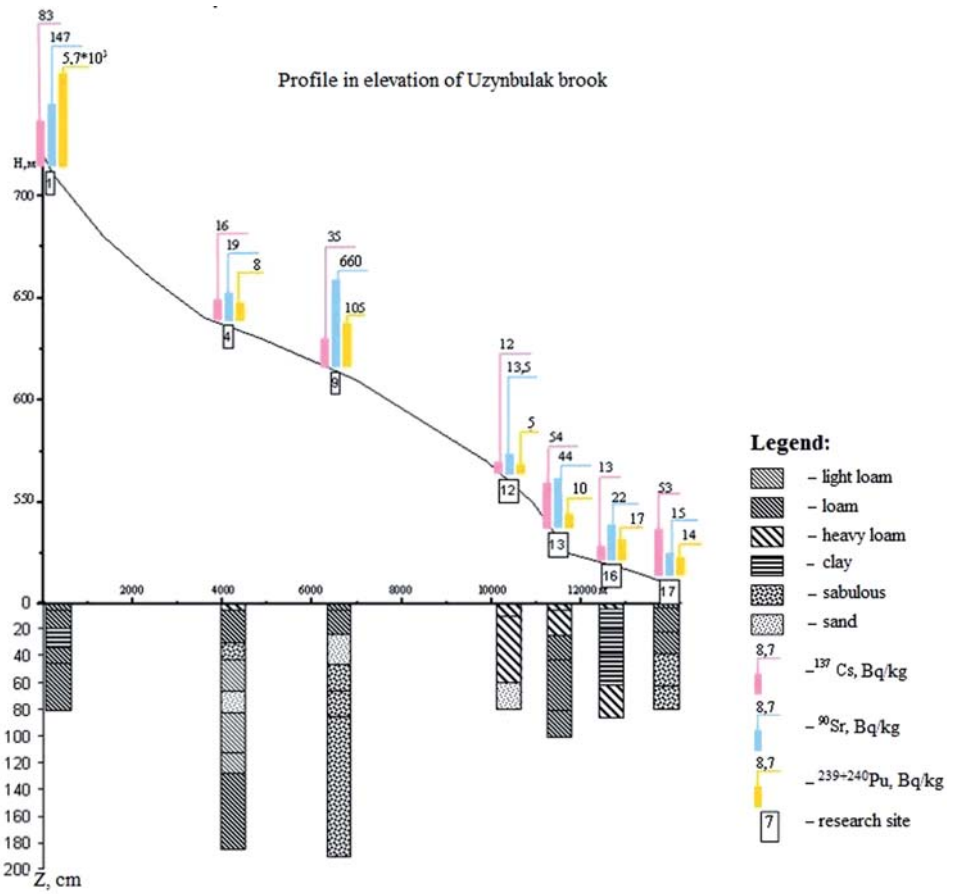


Figure 21. Distribution of artificial radionuclides in soil profiles along the Uzynbulak riverbed

^{137}Cs concentration in all soil horizons does not exceed several tens of Bq/kg with maximal values in the topsoil horizons (up to 83 Bq/kg in the layer 0-5 cm, section 1). ^{90}Sr concentration in soils is higher than specific activity of ^{137}Cs, especially in the upper part of the valley (5.7*10^3 Bq/kg in the layer 0-5 cm, section 1). Along the longitudinal profile the radionuclide concentration mainly remains within first two tens of Bq/kg. Maximal values are not always registered in the surface horizon. It is likely that high ^{90}Sr solubility and soil moistening cause radionuclide transfer both deeper, to the low-lying horizons, and along the runoff of surface waters.

3.1.2.2. Radionuclide contamination of meadow soils in the Toktakushik valley

General characteristic of the Toktakushik stream valley. The Toktakushik stream is situated in southern part of the Degelen mountain massif. Its watershed area is about 6 km^2, the length of the valley is 10-12 km. Maximal absolute elevation in the watershed area is 1070 m, absolute surface elevations range from 710 m in the upper part of the valley to 610 m at the edge of the mountain massif. The surface slopes in the upper part of the valley are 7-10º. The valley width ranges from 2 m to 70 m with no confluents. The stream has tunnels Nos. 610, 809, 8, 11, 169, 170, 165 and 169/2. Of all tunnels tunnel No. 165 has a constant water flow and tunnel No. 11 has a temporary water flow. The characteristics of tunnels are given in the table (Table 12).

Table 12.

Characteristics of tunnels in Toktakushik stream valley

Tunnel #	Test date	Explosion yield, kt	Maximal specific activity at near-portal area, Bq/kg (Bq/l in water)				Water outflow, Flowrate
			^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{241}Am	
8	06.09.70	0.001-20	3.7*10^3	5*10^3	400	-	no
11	20.03.66 28.05.67	100	6*10^5 (26)	9.9*10^4	120	-	occasional
165	08.06.75	0.001-20	2*10^6 (495)	1.3*10^5 (1.5*10^3)	79 (0.88)	1200 (<0.8)	continuous, 79 l/min
169			170	10	110	-	no
169/2-1	04.10.89	0.001-20	125	5	4	-	no
170			19	7.8	130	79	no
610	27.03.70	0.001-20	100	7	380	-	no
809	30.12.71	0.001-20	1.1*10^3	11	370|	-	no

Analysis of the table (Table 12) shows maximal contamination in the near-portal grounds of tunnels Nos. 165 and 11 with water outcome. During snow melting and rains the water flows from the tunnels are displaced and radionuclides are carried-over to the riverbed. The table (table 13) shows annual carry-over from tunnel No. 165.

Table 13.

Average annual concentration of radionuclides in water samples from tunnel No. 165 and annual carry-over from the tunnel

Annual average concentration and annual carry-over	2001			2002			2003			2004			2005		
	^{137}Cs	^{90}Sr	^{3}H	^{137}Cs	^{90}Sr	^{3}H	^{137}Cs	^{90}Sr	^{3}H	^{137}Cs	^{90}Sr	^{3}H	^{137}Cs	^{90}Sr	^{3}H
Specific activity of radionuclides, Bq/kg, for ^{3}H – kBq/kg	270	1200	290	270	1160	240	300	870	230	300	780	230	421	257	207
Annual carry-over, Ci/year	0.2	1	247	0.08	0.03	62	0.04	0.2	42	0.03	0.07	20	0.3	1.9	179

The presence of spots with increased radioactive background on the near-portal grounds of tunnels Nos. 8 and 809 enables to suppose radionuclide carry-over from tunnels by water flow with flood and rain waters to the stream riverbed and, therefore, produces indirect influence on contamination of the valley.

Radionuclide contamination of meadow soils in the Toktakushik stream valley. Radiation contamination on the near-portal grounds without water flows (Table 12) could appear not only during nuclear tests and rehabilitation activities on tunnel conservation but it can also be a result of radionuclide carry-over from tunnel cavities through rock fractures to the daylight surface in the periods when they are filled with water. An important role may be played by the slope stream during snow melting and showers when radioactive substances are washed away from all contaminated surfaces. It means that all tunnels in the valley exert direct and indirect influence on stream contamination.

To study radionuclide migration a longitudinal profile consisting of 5 observation points (sections) (18, 19, 20, 21, 22) was made in the stream valley along its dry bed. The choice of sections was based on the presence of thick layer of natural loose sediments, vegetation and necessity to characterize migration processes along the riverbed including the area of final discharge (Figure 22). The soils of the stream valley refer to the meadow-type soils.

The first observation point (section 18) is located 50 m away from the portal of tunnel No. 165, 1 m away from the stream. It is located in the hollow of 2-3 m in diameter. The riverbed width is 2 m, the valley width is up to 10 m; the valley surface is bumpy, covered with dense vegetation. The section was chosen as the closest section to the tunnel portal with fine soil (on the background of artificial soil for backfilling) and vegetation for sampling.

Section 19 is located 950 m away from section 18 (1000 m from tunnel No. 165), on a flattened part of the riverbed, 4-5 m-wide, the valley width in the section is 15-20 m. The water from the tunnel permeated into rock fractures and was not registered in the section. The choice of the section was determined by the presence of natural soil.

Section 20 is located 690 m away from section 19 (1690 m from tunnel No. 165), in dry flattened riverbed, 4-5 m wide, after inflow of a small narrow side stream (the valley width is 40-50 m). The choice was determined by additional inflow of radioactivity from the side tunnel to the side narrow stream.

Section 21 is located 520 m away from section 20 (2210 m from tunnel No. 165), on a flattened section below the place where the valley turns to the outcome from the mountain massif, which specified its choice. The valley width is 50-70 m.

Section 22 is located 7.5 km away from section 21 (9760 m from tunnel No. 165), in the final part of the runoff, which was the criterion for choice. Its surface is a flattered area with small-sized fragmental material brought by water.

^{137}Cs concentration in the longitudinal profile along the Toktakushik riverbed varies widely, from several hundreds of Bq/kg in the upper part to several units in the lower part of the profile (Figure 22). ^{90}Sr concentration in the surface horizon of section 18 located in the upper reaches of the stream is $2.5*10^4$ Bq/kg. Specific activity gradually decreases down-stream. $^{239+240}$Pu concentration in soils in the Toktakushik stream valley varies within several tens of Bq/kg.

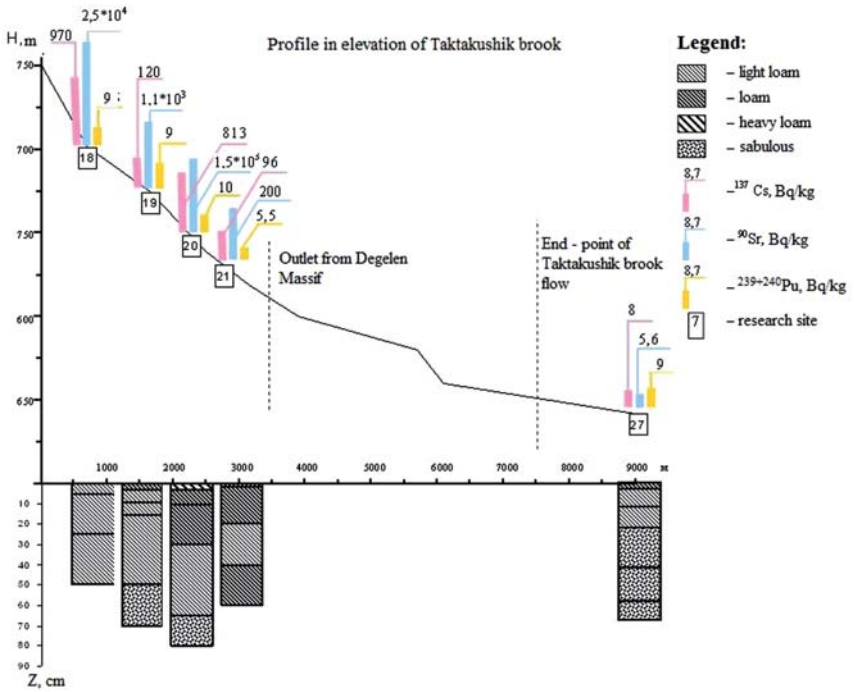


Figure 22. Distribution of artificial radionuclides in soil profiles along the Toktakushik riverbed

A comparison of radionuclide carry-over from tunnel No. 165 in different years (Table 13) shows that specific activity of radionuclides in the stream decreased but the ratio of carried-over ^{137}Cs to ^{90}Sr remains practically the same, i.e. the amount of carried-over ^{90}Sr is 3-6 times greater than that of ^{137}Cs. The specific activity of ^{90}Sr in soils is 100–130 times greater than that of ^{137}Cs, and as the distance from the profile beginning increases, it means that radionuclide specific activity decreases, the ratio sharply decreases.

In the outcome zone from the Degelen mountain massif (section 21) ^{137}Cs (100 Bq/kg) and ^{90}Sr (200 Bq/kg) specific activities in the surface horizon exceed the background of global precipitations. In the final discharge area specific activity of radionuclides is at the level of background of global precipitations (5-20 Bq/kg, section 27). Radionuclides may be transferred beyond the mountain massif but at small distances.

3.1.2.3. Radionuclide contamination of meadow soils in the Baitles stream valley

General characteristic of the Baitles stream valley. The Baitles valley is located in the southern part of "Degelen" test ground. Its runoff is formed by the two main confluents. The third, left-side confluent inflows into the main riverbed beyond the mountain massif. Weakly developed riverbed of the confluent indicates little runoff. The length of the stream valley is over 15 km, the stream width ranges from 2–3 m to 30 m, the valley width ranges from 10 to 500 m. The valley slopes go up to 5º.

The sources of radioactive contamination of the valley are near-portal grounds of 23 tunnels. Characteristics of the tunnels are presented in the table (Table 14).

Table 14.

Characteristics of the tunnels of the Baitles stream

Tunnel #	Test date	Explosion yield, kt	Maximal specific activity at near-portal area, Bq/kg (Bq/l in water)				Water outflow, Flowrate
			^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{241}Am	
13	19.10.66. 02.09.67.	20-150	$1.3*10^3$	120	835	63	no
14	03.12.66. 23.08.82.	0.001-20	$2.5*10^3$	40	52	<1	no
176	16.05.74. 24.06.83.	0.001-20	$1.7*10^6$ (110)	$5.3*10^4$ (490)	700 (0.05)	- (<1.1)	continuous, 890 l/min
175	30.07.1977	0.001-20	$3.6*10^3$	800	40	4	no
810	07.01.68.	0.001-20	$3.9*10^5$	74	$2.1*10^3$	-	no
207			$1.1*10^3$	190	23	-	no
609	30.12.71. 10.04.76.	20-150	$6.5*10^6$ (360)	$1.8*10^4$ (730)	730 (0.9)	- (<0.8)	continuous, 409 l/min
17	05.08.66.	0.001-20	280	210	300	-	no
180	29.11.83.	0.001-20	400	690	470	490	no
18	04.08.67. 27.11.77.	0.001-20	$3.3*10^4$	$6.8*10^4$	480	120	no
184	14.08.81.	0.001-20	$2.4*10^4$	9.2	580	42	continuous
19	25.03.67. 04.04.69.	0.001-20	$9*10^5$	$9*10^3$	410	19	no
210			$3.1*10^4$	36	820	18	no
185	23.07.76. 24.05.78. 02.06.78.	0.001-20	660	8.2	660	-	no
191	28.03.1972	0.001-20	280	7.1	650	10	no
509	05.09.1968	0.001-20	100	310	$1.7*10^3$	-	no
105	29.11.1971	0.001-20	520	28	360	-	no
21	26.02.1967	20-150	$3.2*10^4$	$6.3*10^3$	101	-	no
508	18.12.1968	0.001-20	46	4.6	$2.7*10^3$	-	no
430			53	4.1	4	-	continuous
K-85			14	9	150	-	no
216	29.11.1983	0.001-20	96	26	310	9	no
200MA, bis	18.10.1984	0.001-20	30	3	56	-	no

Four tunnels Nos. 176, 609, 184 and 430 had constant water flow (now only 2 tunnels have water flow), which directly influences contamination of valley meadow soils. The water flows from tunnels constantly bring radioactive substances directly to the valley, and the make considerable contribution to the total concentration of radionuclides in the valley. In some years the average annual specific activity of ^{137}Cs and ^{90}Sr in water is as high as 300 Bq/kg (Table 15), which is tens of times above the intervention level, which according to NRB-99-doc is 11 Bq/kg for ^{137}Cs and 5 Bq/kg for ^{90}Sr [25].

Table 15.

Average annual radionuclide concentration in water samples from tunnel No.609 and annual carry-over from the tunnel

Tunnel #		2001			2002			2003			2004		
		^{137}Cs	^{90}Sr	^{3}H	^{137}Cs	^{90}Sr	^{3}H	^{137}Cs	^{90}Sr	^{3}H	^{137}Cs	^{90}Sr	^{3}H
609	Specific activity of radionuclides, Bq/kg, for ^{3}H – kBq/kg	160	230	20	190	290	20	260	200	20	300	160	20
	Annual carry-over, Ci/year	0.7	0.8	44	0.3	0.3	20	0.2	0.2	17	0.3	0.1	59

The table (Table 15) shows that during the years of observations the annual carry-over of ^{137}Cs and ^{90}Sr by waters from tunnel No.609 varied but its values did not differ considerably (0.7-0.8; 0.3-0.3; 0.2-0.2; 0.3-0.1 Ci/year).

Tunnels Nos. 810, 18, 13, 14, 175, 210, 19, 207, not having water flows, can occasionally influence radionuclide inflow to the stream; it occurs when snow melting or showers wash out soil from slopes. An analysis of available data on radioactive contamination revealed maximal specific activity registered on the near-portal areas of each of the tunnels (Table 14). The highest values of ^{137}Cs specific activity are registered on the near-portal areas of tunnels Nos. 810 ($4*10^5$ Bq/kg), 609 ($3*10^5$), 18 ($5*10^4$ Bq/kg), 176 ($4*10^4$ Bq/kg), 210 ($3.2*10^4$ Bq/kg). By the degree of radioactive contamination the soils and man-made cover on tunnel portals in the upper parts of the stream can be classified as radioactive wastes as ^{137}Cs specific activity there is 250-300 times higher than the value of maximal permissible activity (SPORO-97).

Radionuclide contamination of meadow soils in the Baitles stream valley. To study accumulation of artificial radionuclides in the meadow soils in the Baitles valley, a longitudinal profile was made along the dry riverbed from tunnel No.609 to the final discharge area. The profile consists of observation points (sections) 22, 23, 24, 25, 26 and 28 (Figure 23).

The observation points were made at a distance of not less than 1km from each other, in places with natural ground and vegetation.

Section 22 is located at a distance of 20 m from the portal of tunnel No.609. Its surface is a shallow hollow of 2-3 m in diameter. The riverbed width is 2-4 m. The section was chosen as the area closest to the portal with a water flow covered with soil.

Section 23 is located at a distance of 1.2 km from section 22 in the dry riverbed, 4-5 m wide. The surface is flat. The section was chosen because of presence of soil and vegetation for sampling. At a distance of 3 m from the soil section is a water flow from the tunnel, where water for measuring radionuclide concentration was taken. A water sample was also taken from subsoil waters in the soil section.

Section 24 is located at a distance of 1.1 km from section 23 in the dry riverbed, 5–7 m wide. The surface is flat. The section was chosen downstream the inflow of temporary stream into the riverbed, where additional inflow of radioactive substances from tunnels Nos. 175, 210, 19, 184 may occur. There was no stream from tunnel No. 609 at the moment of measurements.

Section 25 is in the middle flow of the stream, at a distance of 1.3 km from section 24 in the place of confluence of two streams, which was the main criterion for choosing the

place for the section. The surface is flattened, the riverbed width is 5–7 m. No water flow was registered at the moment of measurements.

Section 26 is at a distance of 1 km from section 25 in the place where the stream flows out from the mountain massif, in the dry riverbed, 10–15 m wide. The surface is bumpy. The main criteria for choosing the section were natural soils, vegetation and place where the stream goes out from the massif.

Section 28 is at a distance of 7.3 km from section 26 in the area of final discharge of the Baitles stream, at a distance of 6–7 km from the mountain massif. The area is a sloped plain with dry gullies stretching to south-west. The soil section is made in the dry, 5 m-wide gully.

Concentrations of ^{137}Cs and ^{90}Sr in soil horizons of longitudinal profile in the Baitles valley are high in the upper reaches. For example, in two surface layers 0–5 cm and 5–30 cm (section 22) ^{137}Cs and ^{90}Sr concentrations are over 3000 kBq/kg and 46 kBq/kg, respectively. ^{137}Cs concentration sharply decreases downstream reaching several units of Bq/kg in the last section (Figure 23). Hence, the analysis of radionuclide behaviour along the profile in the Baitles riverbed shows sharp decrease in their concentration.

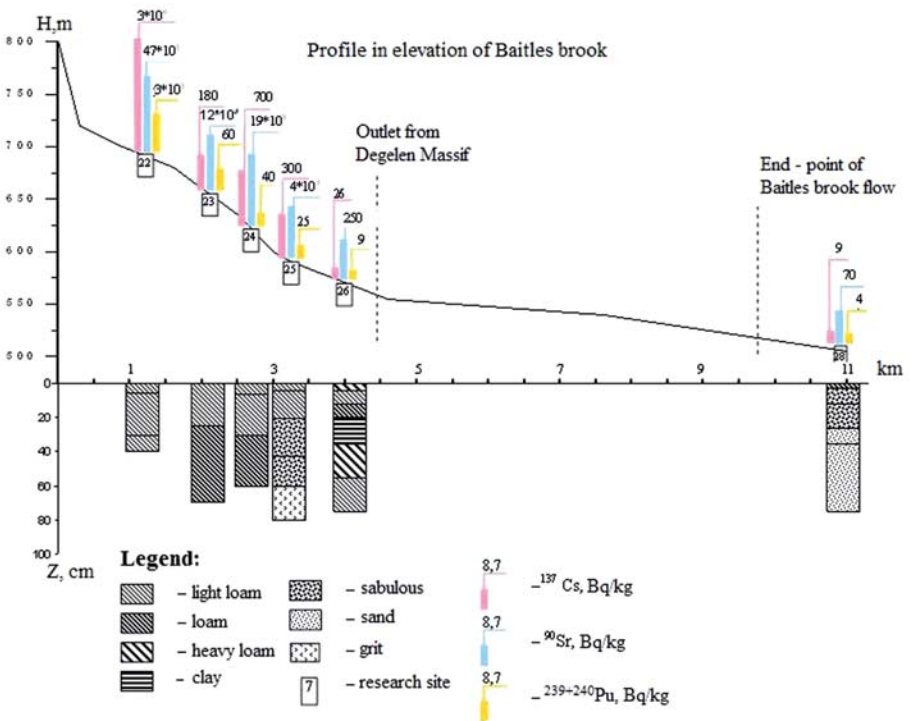


Figure 23. Distribution of artificial radionuclides in soils of longitudinal profile in the Baitles riverbed

A decrease in radionuclide specific activity along the profile is caused by their consumption in soils. As maximal concentrations of ^{137}Cs and ^{90}Sr are registered in the upper horizons (turf and humus), one can suppose that the main role in their absorption belongs to humus. Concentration of organic substance in surface horizons along the profile from upper

to lower part varies from 18.1 to 8.7 %. Its amount also changes with depth from 15.3 in the layer 0–5 cm to 1.4 in the soil-forming structure (horizon "C"); moreover, the soil exchange capacitance decreases from 54.4 to 10.4 mg-equivalent per 100g of soil. As these soil parameters decrease, radionuclide absorption also decreases [26].

Soils of all studied sections along the profile are light loams where sum of particles <0.01 (alphitite) is within 30% of sum of all particles. It indicates possibility of sorption of ^{137}Cs and ^{90}Sr radionuclides by fine-dyspersated soil particles. It is known that soils with high content of alphitite have higher sorption capacity than light soils [27]. Therefore, if in the upper stream the main role in the sorption processes is played by organic soil substance, in the lower stream as humus content decreases the main role in adsorption processes is played by fine-dyspersated particles whose amount is as high as 40–50 %.

The role of radionuclide absorption in soils can be indirectly estimated comparing the data for soils and subsoil waters from the same soil section 23. In soils (layers 0–25 and 25–75 cm) concentration of ^{137}Cs is 20 and $1.9*10^2$ Bq/kg, concentration of ^{90}Sr is $1.2*10^4$ and $1.1*10^4$ Bq/kg, respectively, whereas in subsoil water it is 9.5 Bq/kg for ^{137}Cs and 40 Bq/ kg for ^{90}Sr. It means that soils which were in the riverbed under the flow adsorbed part of radionuclides, which caused decrease in their concentration in water. In summer period, in the absence of high-water surface flow, the flow of subsoil and ground waters, which carry radioactive substances, remains. Therefore it is impossible to estimate radionuclide sorption in soil in natural conditions, where many processes proceed simultaneously. The aspects of absorption of artificial nuclides by soils are based on experiments carried out in the conditions of humid climate – data on East-Urals Trace, Chernobyl. To confirm the conclusions it is necessary to carry out field and laboratory investigations in the STS.

An analysis of radionuclide distribution along the profile (Figure 23) shows that ^{137}Cs concentration has more noticeable peaks of changes than that of ^{90}Sr. Though ^{137}Cs concentration at the beginning of the profile is rather high ($3*10^6$ Bq/kg), at the end it is lower (15 Bq/kg) than that of ^{90}Sr ($2*10^2$ Bq/kg). A sharp decrease in ^{137}Cs concentration is also registered after the first profile point (section 22), at a distance of 1 km from it, in section 23, ^{137}Cs specific activity is 12,000 times lower than its initial value, whereas ^{90}Sr concentration decreased only 4 times. The data show considerable increase in radionuclide activity in soils of section 24, which may be caused by additional inflow of materials from tunnels located on both sides of the stream valley.

The above data show that at the beginning of the profile (section 22) ^{137}Cs concentration in soils is 50 times higher than that of ^{90}Sr. Further, along the valley (starting from section 23) ^{90}Sr activity is many times higher than that of ^{137}Cs. Such distribution of radionuclides in valley soils can be explained by peculiarities of their behaviour. Thus, ^{137}Cs remains near the portals of tunnels located in the upper reaches of the stream, whereas ^{90}Sr due to its mobility and solubility is distributed throughout the valley. The results of investigations show that in water of the stream located 3 m away from the soil section 23, i.e. 1 km from the tunnel portal, specific activity of ^{137}Cs and ^{90}Sr was 2.5 and 205 Bq/kg, respectively. It confirms conclusions on different distributions of radionuclides in the valley in spite of equal values at the flow outcome from tunnel No. 609 (Table 15).

3.1.2.4. Radionuclide contamination of meadow soils in the Karabulak stream valley

General characteristic of Karabulak stream valley. The Karabulak stream consists of two confluents. Most tunnels Nos. 150, 151, 603, 703, 149 are located in the upper reaches of

the right confluent in the Karabulak basin. Tunnel No. 150 has constant water flow. Tunnel No.148/5 is located downstream, in the right confluent. Tunnel No.152 is in the left confluent.

Table 16.

Characteristic of the Karabulak stream tunnels

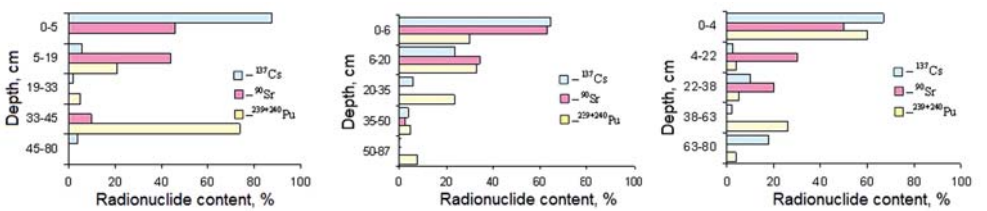
Tunnel #	Test date	Explosion yield, kt	Maximal specific activity at near-portal area, Bq/kg				Water outflow
			^{137}Cs	^{90}Sr	$^{239+240}$Pu	^{241}Am	
150	19.02.1982	0.001-20	$8.4*10^3$	-	-	-	continuous
151			23	12	$2.5*10^3$	-	no
603	30.01.1974	0.001-20	78	28	-	205	no
703	29.06.1967	0.001-20	44	22	$2.7*10^3$		no
149			82	-	-	70	no
148/5	16.12.1974	3.8	$1.2*10^5$	250	850	910	no
152	25.07.1985	0.001-20 (x2) 0.001-20 (x2)	710	860	360	-	no

Radionuclide contamination of meadow soils in the Karabulak stream valley

Meadow soils of the Karabulak valley show maximal concentration of ^{137}Cs and ^{90}Sr in the upper reaches of the stream as well as in other valleys of the Degelen mountain massif. Maximal concentration of radionuclide ^{90}Sr, 64.5 Bq/kg, was registered near tunnel No. 150. Concentrations of artificial radionuclides decrease in soils along the longitudinal profile and in vertical direction.

3.1.2.5. Radionuclide contamination of vertical soil profile of stream valleys of the Degelen mountain massif

Vertical radionuclide distributions in soil sections in the Uzynbulak valley located in different parts have some differences, therefore for comparison we use the data on concentration of radionuclides ^{137}Cs, ^{90}Sr and $^{239+240}$Pu in the upper reaches – section 1, middle part – section 8, and in the lower reaches – section 17 (Figure 24).



a b c

Figure 24. Distribution of artificial radionuclides in soil horizons of longitudinal profile in the Uzynbulak riverbed

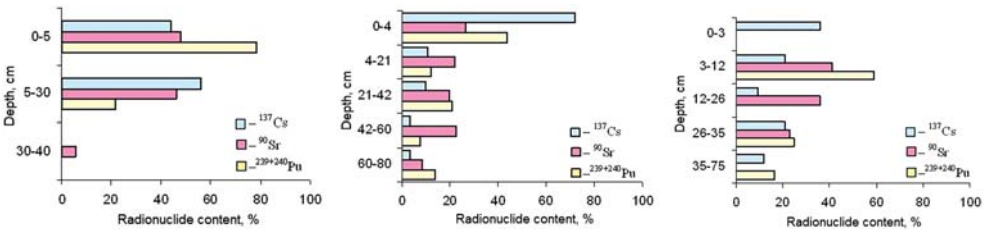
a) section 1 – upper reaches, b) section 8 – middle part, c) section 17 – lower reaches

The figure shows the highest ^{137}Cs and ^{90}Sr concentrations in the surface horizon, with long vertical tail of ^{90}Sr distribution. This result may be explained by large amount of frag-

mentation material in the topsoil horizons, which increases water permeability and allows a considerable part of ^{90}Sr to move to the lower-lying layers. This process can play an important role in the upper reaches (section 1). However, the main factor responsible for vertical radionuclide penetration is its solubility and mobility. $^{239+240}Pu$ concentration in soil horizons, in soil sections, varies within two tens of Bq/kg.

In the Baitles riverbed no ^{137}Cs transfer in the horizontal plane with water flows is observed. ^{137}Cs transfer into lower-lying layers is also insignificant. Having high concentrations in the upper soil layers, the radionuclide demonstrates sharp, by several orders of magnitude, decrease in specific activity with depth, for example, in section 22, in horizon 30-40 cm, ^{137}Cs concentration does not exceed ten Bq/kg (Figure 25).

^{90}Sr concentration decreases along the longitudinal profile and in vertical direction in soil horizons, but its concentration still remains rather high. For example, at a depth of 60-80 cm its concentration is $1.6*10^3$ Bq/kg with specific activity on surface of about $4*10^3$ Bq/kg (section 25). Concentration of $^{239+240}Pu$ in soil horizons of profile sections varies widely. Its maximal values like maximal values of other radionuclides in the valley meadow soils were registered in the upper reaches. Downstream, the values fall to several units of Bq/kg.



a b c

Figure 25. Distribution of artificial radionuclides in soil profiles in the Baitles stream: a) – section 22, b) – section 25 and c) – section 28

In different parts of the basin soils were formed under different conditions, and, hence, have different characteristics, which causes different behaviour of radionuclides. It has been determined that meadow soils in stream valleys of the Degelen mountain massif formed under moistening regime have dark-grey profile colour, thick humus horizon up to 50-60 cm with high concentration of organic substance, up to 19-20 % in the upper reaches, which is in agreement with literature data [28]. In the lower reaches, in the valleys of each stream many soil characteristics change: humus content reduces to 4-4.5 %, concentration of CO_2 carbonates in soil increases up to 14 % (no carbonates are detected in the upper streams), alkalinity increases to 8.5 % from 6.5 % in the upper part of confluents or the main streams. At the same time, the sum of absorbed bases decreases from 30 to 15 %, the amount of fragmentation material falls to 5 % whereas in the upper reaches its amount is as high as 70 %. The above changes cause increase in soil salinity, and at the foot of the mountains the sum of easily soluble salts goes up to 0.7 % and even higher values in the surface horizon [24].

The characteristics of soils show that an important role in sorption of artificial radionuclides in the upper reaches can be played by humus, which has a rather high percentage in soil composition. A certain role may be played by the soil-consuming complex, less important is holding capacity of soil units. However, as the distance from the upper reaches increases, absorption of radionuclides can mainly occur in soil particles among which the percentage of fine-dust and silt fractions increases by a factor of 2. An analysis of soil mechanical composition shows that the sum of particles less than 0.01 mm in diameter (alphitite) is about 30 % in the upper reaches and increases to 40-50 % in the lower reaches.

Here we present histograms of distributions of artificial radionuclides in soil sections in different landscape conditions in stream valleys. The figure (Figure 26) shows histograms of distribution of artificial radionuclides in the upper reaches of the main riverbed and confluents of the Uzynbulak stream.

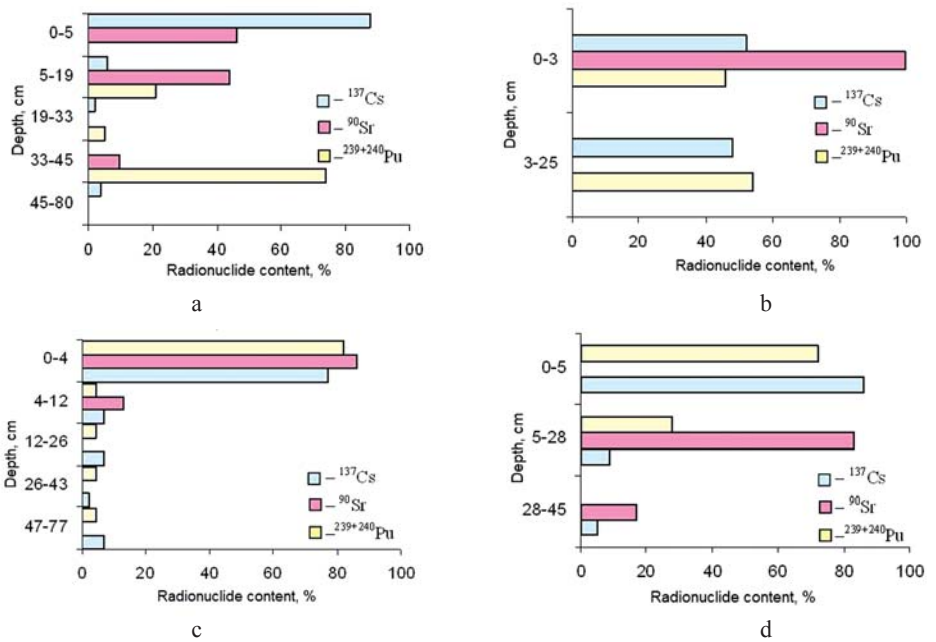


Figure 26. Distribution of artificial radionuclides in the Uzynbulak meadow soils, in the upper reaches of confluents:
a) – section 1, b) – section 10, c) – section 3, d) – section 6

Histograms of radionuclide distributions in meadow soils in the vicinity of the near-portal grounds show that maximal radionuclide concentrations are mainly registered in the upper horizons. No considerable amounts of ^{90}Sr transfer into deep layers were registered, which can be explained by higher hypsometric location of the studied ground relative to the surrounding area and intensive washing out of underlying layers down the slope.

The figure (Figure 27) shows histograms of distributions of artificial radionuclides in soil sections located near outcomes of the main streams from the mountain massif.

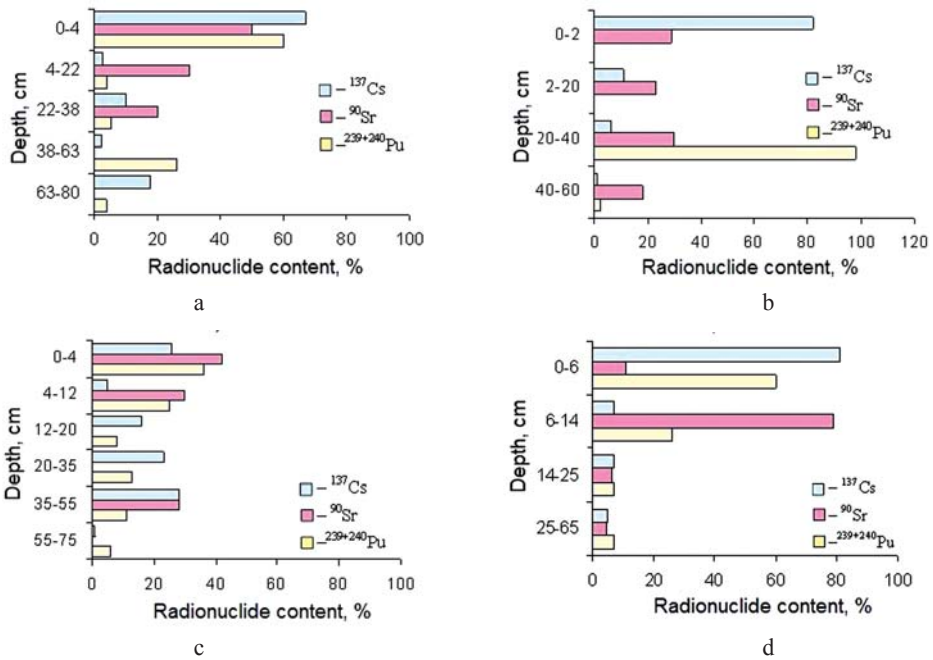


Figure 27. Distribution of artificial radionuclides in meadow soils in the vicinity of the outcome of the main streams from the Degelen mountain massif: a) section 17 (Uzynbulak stream), b) section 21 (Toktakushik stream), c) section 26 (Baitles stream), d) section 29 (Karabulak stream)

It should be noted that in the oldest valley such as the Uzynbulak valley, at the outflow from the massif the radionuclide concentration in soil horizon has a more ordered character than in young valleys. This valley has clearly formed maximum of radionuclide concentrations in surface layers, which may indicate an initial phase of redistribution of newly coming radionuclides.

Radionuclide distribution in soil sections located in the final discharge zone of streams is given in the figure (Figure 28).

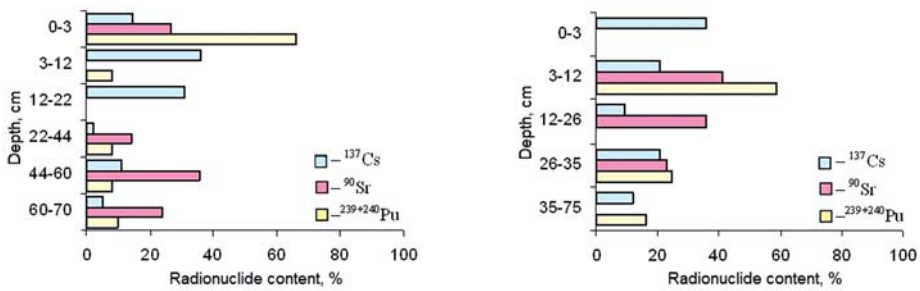


Figure 28. Distribution of artificial radionuclides in meadow soils in the final discharge zone: a) – section 27 (Toktakushik stream), b) section 28 (Baitles stream)

Zones of final discharge of the Toktakushik and Baitles streams are at a distance of 7-7.5 km from the Degelen mountain massif. The soils have light mechanical composition with prevailing fraction presented by sands. Sand is brought by spring floods as there is no runoff in the other seasons. The soils washed from easily soluble salts contain small amounts of carbonates; water pH varies from alkalescent to alkaline reaction. An analysis of histograms shows that radionuclide distribution in sections 27 and 28 is rather uniform along the soil profile with a little decrease with depth.

CONCLUSION

Specific features of radioactive contamination of natural ecosystems of stream valleys subjected to the most intensive contamination by artificial radionuclides are studied based on the long-term complex radioecological investigations in the territory of "Degelen" test ground.

Distributions of radionuclides ^{137}Cs, ^{90}Sr, ^{241}Am and $^{239+240}$Pu in the ecosystems surface waters – soils – subsoil waters are determined by the direction of the main flow from upper to lower reaches but most clearly these processes are seen in the valleys in case of continuous water outcome in tunnels located in the upper part. The main amount of ^{137}Cs carried-over from tunnel cavities remains near portals, most often in the gravel-rubbly filter. ^{241}Am and $^{239+240}$Pu are carried away by surface and subsoil flows at a distance of several hundreds of meters from the portal, which is especially well seen in case of thick water-saturated layer of soils with underlying weathered dense rocks. An unexpected result is rather high migration abilities of ^{241}Am and $^{239+240}$Pu confirmed by observations of their distribution in perpendicular and vertical directions to the riverbed. ^{90}Sr as the most mobile and soluble element can travel with surface and subsoil waters within loose sediments of the valley. In spite of continuous radionuclide carry-over with water to the daylight surface, no extremely high concentrations in the ecosystem components are registered. At the present moment the ratios of maximal specific radionuclide activities in soil of the near-portal tunnel areas do not exceed 10^{-3} for ^{90}Sr, 10^{-5} for ^{137}Cs, 10^{-4} for ^{241}Am and 10^{-6} for $^{239+240}$Pu.

The direction of radionuclide migration in valley soils is determined by geomorphological conditions but the distance passed by radionuclides depends on their initial concentration in tunnel water, slopes and flow discharges. An important role in radionuclide distribution is played by their chemical properties, i.e. forms of their presence in tunnel water and processes of their interaction with ground and soil. It should be noted that the ecosystem surface waters – soils – subsoil waters is itself very dynamical, and artificial radionuclides permeating into the system enhance its nonequilibrium by sorption and desorption processes.

The lengths of tunnel streams are, as a rule, tens or hundreds of meters, however, radionuclides remain within the mountain massif. The main reason for their sharp disappearance in the valleys is ruptures, small and large fractures through which water goes down to the underground basin, however, reduction in radionuclide contamination of soils in stream valleys from portals of tunnels, especially with water outcomes, downstream is explained not only by geomorphological conditions and chemical properties of elements, but also by physical-chemical properties of soils which absorb and retain radionuclides.

Therefore, in spite of complex processes in the ecosystem artificial radionuclides remain within "Degelen" test ground, and do not move beyond its boundaries with surface and subsoil waters.

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ДЕГЕЛЕҢ ТАУЛЫ МАССИВІ ШТОЛЬНЯ СУЛАРЫНЫҢ ЭКОЖҮЙЕЛЕРІ ТАБИҒИ ОРТАСЫНЫҢ РАДИОАКТИВТІ ЛАСТАНУЫНЫҢ ӨЗІНДІК ЕРЕКШЕЛІКТЕРІ

А.В. Паницкий, Р.Ю. Магашева, С.Н. Лукашенко

Жұмыста радинуклидтердің судағы жылыстау әсерінен экожүйе құрауыштарының ластануы туралы деректер ұсынылған. Зерттеулер нәтижесінде Дегелең таулы массиві штольнялары сулары экожүйесінің топырақ жамылғысы, жерүсті сулары, және арнаасты ағын суларының радиоактивті ластану сипаты туралы деректер алынды. Радионуклидтердің табиғат ортасының аталған құрауыштарындағы кеңістіктік таралу сипаты көрсетілген. Сондай-ақ осы радионуклидтердің судағы тиесілі белсенділігі бойынша топырақтағы радионуклидтердің құрамын бағалау болжамы мүмкін екендігі көрсетілген. Экожүйенің түйіндес құрауыштарындағы (жерүсті сулары-топырақ сулары-топырақ) радионуклидтер құрамының түрлі мөлшерлері олардың жағдайының аталған жүйедегі салмақ тепе-теңсіздігін көрсетті. Аталған жұмыста зерттеу нысаны ретінде таңдап алынған су шыққан штольня суларын зерттеу нәтижелерінің қорытындысы радионуклидтердің "Дегелең" алаңы штольня суларының басқа экожүйесінің жайылым топырағындағы қалпын болжауға мүмкіндік береді. Аталған уақыт кезеңінде ^{137}Cs и $^{239+240}$Pu күндізгі жер бетіне шығудан 1,5 км, ^{90}Sr – 2 км алыс тарамайтын радионуклидтердің горизонталды бағыттағы қалпын суреттейтін қисық талдау негізінде болжауға болады.

Түйін сөздер: ядролық жарылыстар, радиоактивті ластану, радионуклидтер, штольнялар, жайылым топырағы, радионуклидтердің таралуы.

УДК 577.391:577.4:504.4.054:539.16:622.258.4

ХАРАКТЕРНЫЕ ОСОБЕННОСТИ РАДИОАКТИВНОГО ЗАГРЯЗНЕНИЯ КОМПОНЕНТОВ ПРИРОДНОЙ СРЕДЫ ЭКОСИСТЕМ ВОДОТОКОВ ШТОЛЕН ГОРНОГО МАССИВА ДЕГЕЛЕН

А.В. Паницкий, Р.Ю. Магашева, С.Н. Лукашенко

В работе представлены данные о загрязнении компонентов экосистем, обусловленном водной миграцией радионуклидов. В результате исследований получены данные о характере радиоактивного загрязнения почвенного покрова, поверхностных вод и вод подруслового потока экосистем водотоков из штолен горного массива Дегелен. Показан характер пространственного распределения радионуклидов в данных компонентах природной среды. Установлены ряды подвижности как вертикального, так и горизонтального перемещения радионуклидов в почвах вдоль и поперек русла водотока. Также показана возможность оценочного прогноза содержания радионуклидов в почве по удельной активности этих радионуклидов в воде. Различные величины содержания радионуклидов в сопряженных компонентах экосистемы (поверхностные воды – грунтовые воды – почвы) показали неравновесность их состояния в данной системе. Обобщение результатов исследований водотоков штолен с водопроявлениями, выбранных в качестве объектов исследований в данной работе, позволяет прогнозировать поведение радионуклидов в луговых почвах других экосистем водотоков из штолен площадки "Дегелен". На основании анализа кривых, описывающих поведение

радионуклидов в горизонтальном направлении, можно прогнозировать, что на данном временном этапе ^{137}Cs и $^{239+240}Pu$ не будет распространяться дальше 1,5 км от выхода на дневную поверхность, ^{90}Sr – не далее 2 км.

Ключевые слова: ядерные испытания, радиоактивное загрязнение, радионуклиды, штольни, луговые почвы, распределение радионуклидов.

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UNDERGROUND MIGRATION OF ARTIFICIAL RADIONUCLIDES BEYOND DEGELEN MOUNTAIN MASSIF

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The article reports on the investigation of the nature of carry-over of artificial radionuclides with groundwater from the nuclear test grounds at "Degelen" site. At the main water flows radiological research has been done and observation wells are equipped for long-term monitoring of groundwater. New findings on the levels of radioactive contamination of groundwater, both within Degelen massif and beyond its borders were obtained. Based on these data conclusions have been made on the nature of artificial radionuclides migration with the groundwater flow, prognostic assessment was carried out for the possible development of the radiological situation at Degelen mountains.

Key words: groundwater, nuclear test, STS, Uzynbulak, Baitles, Karabulak, Altybay, migration of radionuclides, geological structure and hydrogeological conditions of Degelen massif.

INTRODUCTION

The underground nuclear explosions (UNE) entailed radioactively- contaminated areas, which with time undergo considerable changes, mainly, as a result of radionuclide migration. Of special importance is studying of the character of artificial radionuclide migration with ground waters as this process may cause radioactive contamination of drinking water sources and waste of geological environment with all its resources. In the territory of the Semipalatinsk Test Site (STS) the problem of migration of UNE radioactive products beyond the test grounds is most important and pressing for the "Degelen" test ground . It is primarily explained by the geological structure and hydrogeological conditions of the Degelen mountain massif which define mechanisms and character of radioactive contamination of ground waters and conditions of further migration of artificial radionuclides with water flows. It should be noted that ground waters of the "Degelen" test ground have high concentrations of artificial radionuclides many times higher than maximal permissible concentrations for drinking water (up to 700 Bq/kg for ^{137}Cs, up to 2000 Bq/kg for ^{90}Sr, 1300 Bq/kg for ^{3}H and up to 110 Bq/kg for $^{239+240}$Pu) [1]. The prognosis estimations made on the base of current data and techniques show that ground waters of the Degelen mountain massif flowing beyond the UNE central areas will not experience any sharp changes but will show slow reduction in ^{90}Sr and ^{137}Cs concentrations and weak increase in $^{239+240}$Pu concentration. Therefore, it is not impossible that in the foreseeable future considerable changes in the radioecological situation may be observed beyond the "Degelen" test ground [1, 2, 3].

This problem has become especially pressing nowadays, when complex investigations aimed at transfer of some STS part into economic usage are carried out. To make a complete assessment of safety of transferred lands it is necessary to have not only concentrations of artificial radionuclides in ground waters spread on the STS territory but also prognosis estimations of possibility of carry-over of contaminated ground waters from the UNE places to the ground waters of other territories. This review presents the results of investigations of the problem in question carried out by the NNC institutions in different time intervals.

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1. GENERAL INFORMATION ON THE "DEGELEN" TEST GROUND

1.1. General information on geological structure and hydrogeological properties of the Degelen mountain massif

The Degelen mountain massif is situated in the STS southern part, has an isometric shape and towers over the surrounding area up to 500 m (Figure 1). The absolute peak altitudes are a little higher than 1000 m.

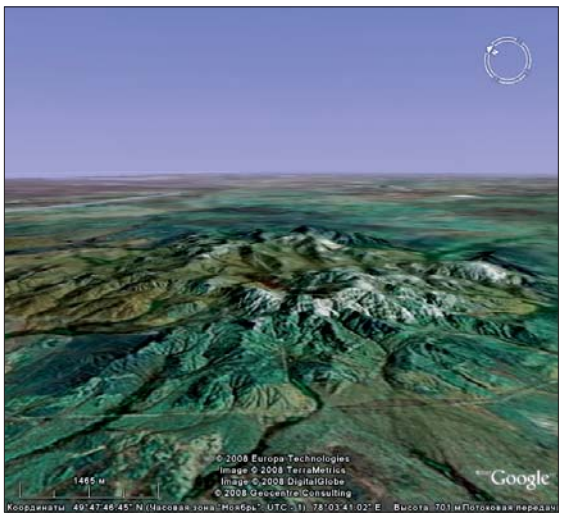


Figure 1. Degelen mountain massif

Degelen mountain massif is a complex volcanic-abyssal structure located in the eastern side of the Chingiz-Tarbagatay fold system. The structure was formed as a result of tectonic-magmatic activation in the Upper Paleozoic. The beginning of magmatic activity refers to the Middle Carbon when lavas of the main and mean composition flowed over the Devon coal mold. The Upper Carbon sediments outcrop to the south and southeast from Degelen mountains and are presented by andesites, andesite-basalts, their tuffs, tufas, sandstones and siltstones.

In the Upper Carbon – Lower Permian period large central-type volcanic structures were formed. Volcanic structures mainly consist of andesites and basalts in the lower part of the thickness and acid lavas in the upper part. The rocks make a slope of 15–35° to the center of the structure. The central and western parts of the volcanic structures are broken through by the Upper Permian oval-shaped granite range. The range stretches in the north-northwest directions, its length is 18 km and transverse size is 10 km. Granite penetrations into volcanic rocks form intrusions and cornification zones of thickness up to several tens of meters. The granite rock was formed in two stages. The first stage is presented by large- and middle-grain granites, whereas the second stage is presented by small-grain biotite and leucocratic granites. The tunnels made in the second-phase granites are more stable.

At the final stage of granite range formation an important role played a high-temperature hydrothermal process. In the apical part of the granite range greisens with

tungsten mineralization (Degelen deposit) were formed. Hydro-thermally changed rocks are enriched in pyrite, which increases sulphate concentration in fracture waters.

Fractures cross Degelen mountain massif in the northwest, northeast and latitudinal directions. The largest fracture passes along the central part of the Uzynbulak valley. Its length is over 10 km. In the southern part of the mountain massif there are latitudinal fractions stretching as far as 7 km. The fracture zones are often water-bearing zones and form the main water outcomes in tunnels crossing zones.

During the tests of nuclear devices in the Degelen mountain massif, 181 tunnels were made, of which 124 were made in granite, 35 – in liparite lavas and tuff-lavas, 17 – in basalts and andesite-basalts and 5 – in subvolcanic syenite porphyries and syenites. The tunnels with water outcomes are usually located in granites; more rarely water outcomes are registered in tunnels made in liparite lavas, syenites and other rocks.

The mountain massif is crossed by narrow valleys with runoff in different directions. The largest watershed area has the Uzynbulak valley of southeastern direction. Degelen mountains is the place where surface water systems, mainly of northern and eastern directions, are formed. Therefore the state of surface, subsoil and fracture waters mainly depends on ecological conditions in the Degelen mountain massif. The area does not have any transit valleys.

Large areas of the mountain massif are occupied by rocky structures and serve as drainage areas for atmospheric fallouts. In the periods of intensive precipitations and spring snow melting some part of water is transported by surface runoff beyond the mountain massif, and the other part permeates along open fractures. The ground water level in the inter-mountain valleys is at depths varying from 0 to 2-6 m. Beyond the bounds of the mountain massif the groundwater level decreases to 10-12 m. In the watershed area this level goes down to 100-200 m. This thick zone is a zone of filtration of atmospheric fallouts to the level of regional basin of fracture waters. Therefore in the mountain area the fracture waters are divided into fracture waters of filtration zone and fracture waters of regional basin (Figure 2). Almost all tunnels are located in the filtration zone, which explains water inflow to tunnels from the roof.

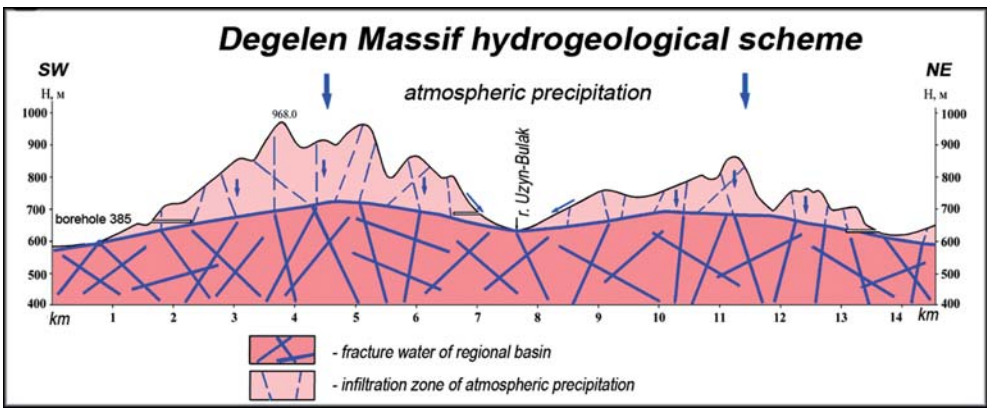


Figure 2. Scheme of groundwater recharge

Therefore Degelen mountains have the following types of waters:

- Porous waters of diluvial-proluvial sediments with limited propagation;
- Porous waters of diluvial-proluvial sediments including underflows;
- Fracture waters of Paleozoic structures.

A special type of fracture waters is presented by waters of water-bearing fracture zones. Some valleys have sediments of waterproof Neocene clays. They divide the areas of porous and fracture waters. In other cases porous and fracture waters have direct connection with each other.

The map of the roof of fracture waters horizon in the Degelen mountain massif (Figure 3) is constructed based on the archive data on hydrogeological wells, springs, water-bearing tunnels and materials obtained during project investigations.

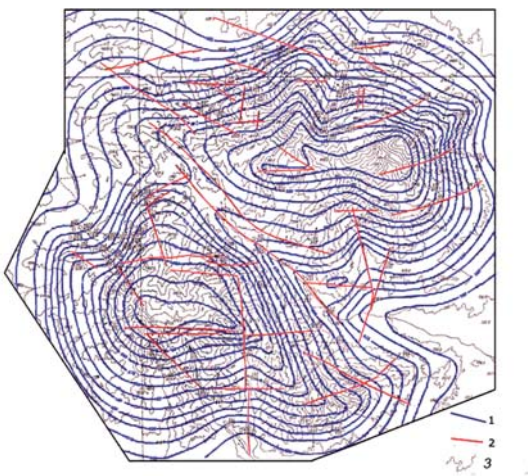
The hydroisohypses show the most probable directions of groundwater motion. The most probable direction is southeastern; it coincides with the direction of the Uzynbulak valley. The map shows that regional fracture waters in Degelen mountains form two dome-like elevations. The groundwater plane under southern watershed is located at a depth of 350m below the daylight surface. The groundwater plane under northern watershed is located at a depth of over 390 m.

Hydrodynamic parameters of the massif directly depend on the hydrometeorological conditions of the region. The investigations enabled to detect about 20 continuously acting springs. The spring outputs vary from 0.5–0.6 to 1000 dm^3/min. The springs with the highest output are located at a depth of 545–650 m. They are connected with the main water-bearing fractures and are discharge points for fracture-vein waters. They have an ascending character. Some springs are connected with subsoil waters and have a descending character.

In addition to the above-mentioned there are many (~30) seasonal springs. Seasonal nature of springs is indicated by hygrophilous plants, soil moistening and erosion incisions.

During operation of the "Degelen" test ground water outcomes were registered in more than 50 tunnels.

According to chemical composition the ground waters of Degelen mountain massif refer to sulphate and hydrocarbonate-sulphate waters. The prevailing cation is Ca^{2+} registered in the waters of practically all tested observation points. Cations Na^{+}+K and Mg^{2+} occupy secondary position. The waters are very soft and fresh with mineralization less than 1 g/dm^3. The ground waters have stable composition which does not depend on season [4, 5].



1- Hydroisohypses; 2 – regional faults; 3 - Isohypses

Figure 3. Map of the roof of fracture waters horizon of Degelen mountain massif

1.2. Nuclear tests on the "Degelen" test ground

"Degelen" test ground is one of the main STS test sites where underground nuclear explosions (UNE) of power up to 150 kilotons were made in horizontal excavations – tunnels. In the period from 1961 to 1989, 181 tunnels were made and 295 nuclear devices were tested (table 1) [6]. Group nuclear explosions were made in one excavation (tunnel) and simultaneously in several excavations. The first group explosion was made on 03.12.1966 in tunnel No.14, it was simultaneous blasting of two nuclear devices. 2 nuclear charges were tested simultaneously 43 times, 3 charges were tested 16 times, 4 charges – 5 times and the maximal number of charges – 5 were tested 2 times.

Table 1.

Distribution of nuclear tests on the "Degelen" test ground as a function of nuclear power release

Yield, kt	Number of explosions	Yield, kt	Number of explosions
<0,001	21	от 20 до 150	25
0,001-20	249	total	295

The explosions were accompanied by various radiation effects whose quantitative distribution is presented in table 2.

Table 2.

Quantitative analysis of radiation effects accompanying nuclear tests

Radiation effect	Number of explosions
No noble gas released to atmosphere at explosion	111
Minor amounts of noble gases released to atmosphere at explosion	49
Gas and steam outburst at explosion	2
Radioactive gas and steam outburst followed by combustion of the gas	1

The main UNE destructive factor is seismic waves generated in the test hypocenter. As the wave passed through the boundary of two media (rock – atmosphere), the upper part of the rock was broken off. The breakoff was accompanied by rock destruction into blocks along fractures with formation of a great amount of large-sized blocks on massifs and large-sized colluvium on slopes. The natural colluvium which covered the slopes before explosion, after shaking moved down from its initial place and formed middle-small-sized slide-rocks contouring large-block fraction. In the explosion epicenter the destructions developed according to two scenarios depending on charge power, tectonic characteristics and fracturing:

- the first scenario – formation of heaps and slide-rocks;
- the second scenario – formation of subsidence crater.

In the second scenario seismodislocations were formed in 84 cases. The craters had different sizes, depths, and in most cases, are of oval shape. The contour of the inner crater had seismovibration cones and bumps typical of epicenter zones of strong and catastrophic earthquakes. During explosions most likely gas broke through to the surface causing formations of open faults probably reaching the explosion chamber.

The UNE caused considerable deformation of the mountain massif, formation of numerous crushing zones, subsidence craters and open faults. As a result, rock permeability increased which caused increase in descending filtration and partial transformation of surface flow into ground flow. Therefore as a result of nuclear tests a new type of ground waters was formed in Degelen mountain massif. It unites the flows of fracture-vein and fracture waters of the zone of filtration of atmospheric fallouts and the inflow of tunnel waters (Figure 4).

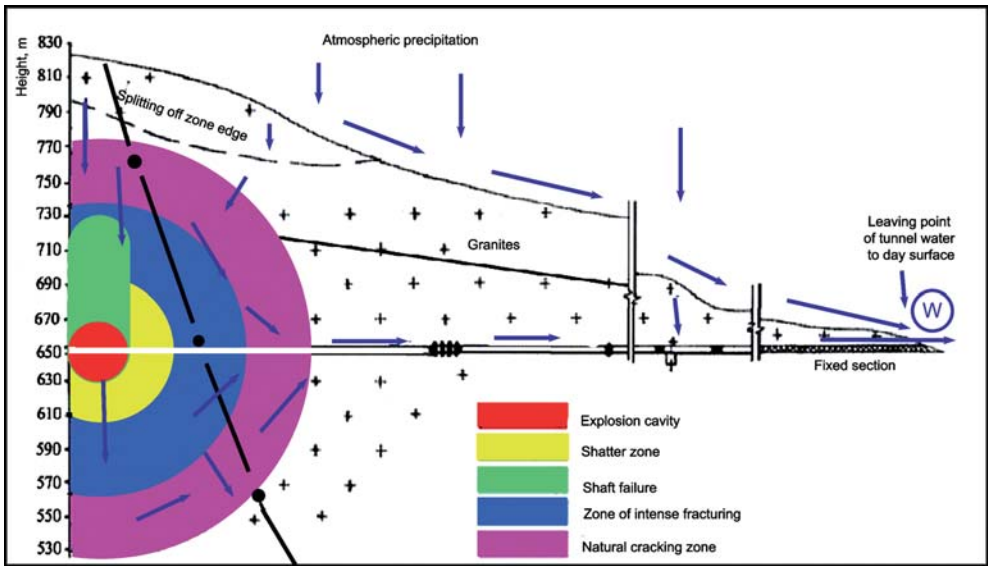


Figure 4. "Degelen" test ground. Migration of ground waters in the tunnel after the UNE

The inflow of atmospheric fallouts and fracture-vein waters in zones of irreversible deformations and directly into the crater causes formation of radionuclide contamination of tunnel waters. Moving along the system of fractures and tunnel cavities radioactively contaminated waters penetrate into the basin of ground waters or outcome to the daylight surface in the vicinity of tunnel portals.

1.3. Works on tunnels conservation

1.3.1. Assessment of influence of tunnels conservation on water regime

In the period from 1996 to 1998, 178 tunnels in Degelen mountain massif were closed. In 1996, 59 tunnels were conserved, in 1997 – 65 tunnels, in 1998 – 54 tunnels (Figure 5).

The methods used to conserve a tunnel were determined by its technical state, its specific features and mainly by maximal safety of personnel fulfilling operations on tunnel conservation. The most ecologically sound method is placing a concrete plug excluding additional artificial impact such as failure of overlying rocks in case of detonation of explosives. The description of methods of tunnel conservation by the years of carried out works is presented in table 3.

Table 3.

Methods of tunnel conservation by years

No.	Method	1996	1997	1998	Total
1	Overlaid charges	1	7	12	20
2	Vertical blastholes from the surface	21	24	15	60
3	Blastholes in the tunnel	22	18	8	48
4	Concrete placing	15	16	14	45
5	Horizontal boreholes	-	-	11	11
6	Restoration to the natural relief	59	65	54	178

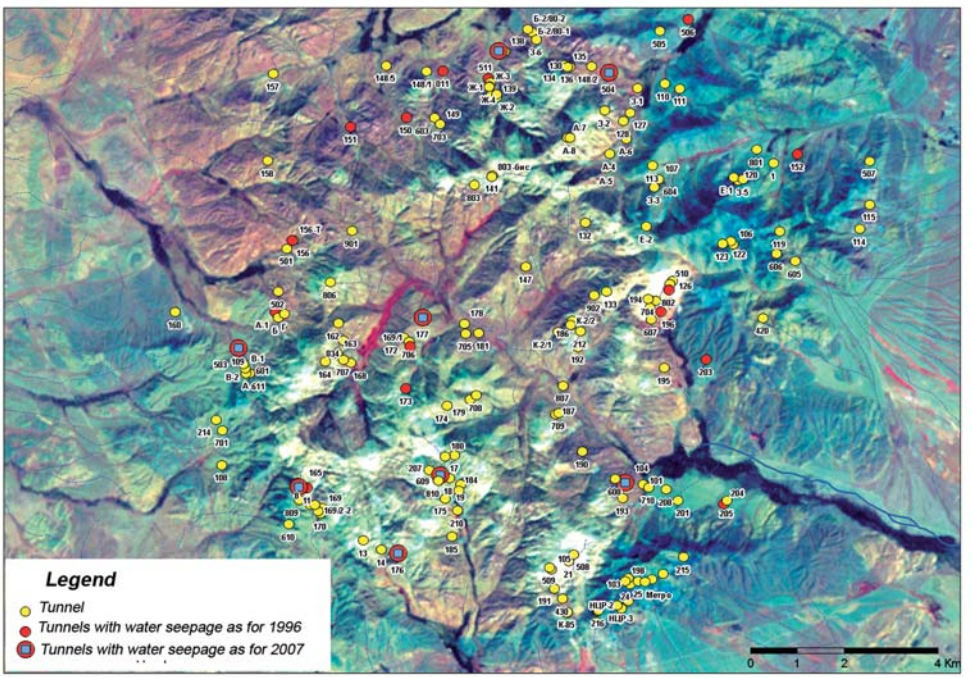


Figure 5. Degelen mountain massif. Layout of tunnels.

After conservation of each tunnel using one of the above-described methods or their combinations, the tunnel portal and cut-in trench were filled with earth up to the level of natural relief of the mountain.

Almost all tunnels are located above the level of the regional basin of fracture waters, in the zone of atmospheric moisture filtration. The underground nuclear tests caused irreversible changes in the structure of enclosing rock, which changed water regime. Large camouflet cavities, zones of artificial fracturing and tunnel spaces appeared in the filtration zone. Moreover, water permeability of natural structures changed and open cavities appeared, especially near the surface. All structures newly formed in the enclosing rock may play role of drainage systems, temporary moisture accumulators and new ways of migration. As a result,

such phenomena as unexpected stoppage of water inflow into tunnel, breakthrough of large amounts of water, inflow of water accumulated in the tunnel from behind concrete walls were observed. These phenomena were registered during the double-run tunnel investigations in 1996. Thus, in the winter-spring period an intensive water inflow was registered in tunnels Nos. 177 (up to 1250 l/min) and No. 802 (600 l/min). In the summer-autumn period the water flow from tunnel No. 177 decreased to 302 l/min, and the water flow from tunnel No. 802 stopped. At the same time the water flow from tunnels Nos. 104 and 503 increased up to 1,400 l/min.

In 1996 water was registered in 34 tunnels. Only in 23 tunnels water appeared in amounts sufficient for its appearance in portal.

A preliminary examination showed that total water runoff from tunnels was 4,200–4,400 l/min. Taking into account that flows of total intensity of about 3600 l/min came from radioactively contaminated tunnels, it becomes clear why it is necessary to take all possible measures to reduce flows intensity or to isolate them on the surface.

In 1996 five water-bearing tunnels (Nos. 169/1, 173, 196, 203 and 204) were conserved. Only on the portal of tunnel No. 173 a small water flow along the drainage pipe was registered. The flows in four tunnels (Nos. 11, 143, 150 and 802) either fully stopped or water retarded to the tunnel cavity and did not appear in the portal. That is why regular observations of water inflow in 1997 were made in 14 tunnels out of 23. It should be noted that in 1996 gradual decrease in water intensity was observed. Therefore tunnel conservation caused reduction in the total water flow and natural disappearance of some flows of discharge of 1440 l/min.

As a whole, the year 1997 was characterized by decrease in flow intensity from tunnels. The phenomenon could be explained by on-going process of portal sealing and unusual climatic conditions. By October, the total flow volume decreased to 716 l/min. In 1997, seven portals of 15 observed were sealed (tunnels Nos. 011, 151, 152, 156, 156-T, 504 and 506). At the moment of conservation the total flow in tunnels was 239 l/min. It is the amount by which the observed water flow to tunnels portals decreased. After portal sealing and slope flattening in tunnel No. 504 the water continued to infiltrate running down as a thin stream (up to 6 l/min). Water seepage through the sealed portal was also observed in tunnel No. 156-T. The water amount infiltrated into soil and discharged on the surface did not exceed 5-6 l/min. In October, as soil was saturated with water, the wet area occupied over 100 m^2, and water came to the surface with discharge over 10 l/min.

Therefore, in 1997 it became clear that water could permeate through sealing structures and outcome to the surface. The water inflow to the portals of two sealed tunnels Nos. 156-T and 504 did not stop. The water from tunnel No. 511 conserved on 3 December, 1997 also went on flowing. The above flows continued to bring radionuclides to the surface. The situation made it necessary to take a special decision on placing special filters consisting of a water collector and a water dam from large fragmental material shutoff near sealed portals.

In October 1998 the portal of the last water-bearing tunnel No. 609 was sealed, and the water flow stopped soon after sealing. Since that time observations of water flows have been made near portals of sealed tunnels. Filters were made in front of 10 tunnels with constant water inflow. The examination of tunnels showed that in 5 portals filters provided complete water absorption in the waste heap. The flow intensity in portals was measured directly near filters which were not fully dammed by waste material. More massive dams enabled to isolate flows

from tunnels Nos. A-1, 156-T and 165. In tunnel portals where flow intensity exceeded 200 l/min the water started to make riverbeds and flow down to the nearest reach. High intensity flows quickly saturated the heap; as a result swampy areas were formed around it.

Therefore after conservation of all water-bearing tunnels water continued to inflow from sealing structures in 8 portals: Nos. 104, 165, 176, 177, 503, 504, 511 and 802. Figure 6 shows the results of long-term observations of tunnel flow discharges after tunnel conservation.

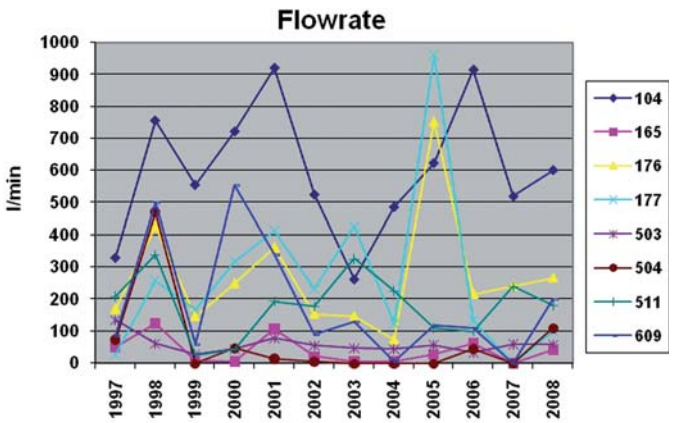


Figure 6. Discharge of tunnel streams

The diagrams show the average values obtained by the results of quaternary measurements. The available data show that most tunnel streams have unbalanced (pulsing) flow.

1.3.2. Assessment of influence of tunnel conservation on radionuclide carry-over with water flows

Exploration of tunnels made in 1996 before portals sealing showed that most tunnels with water ingress had radioactive contamination. Therefore water samples were analyzed on concentration of ^{137}Cs and ^{3}H, and in 1999 ^{90}Sr analysis was added.

After tunnels conservation the information on radioactive contamination of flows was obtained from portals of 29 tunnels. In the portals of 19 tunnels water appeared occasionally, that is why single samples were taken there. In all flows except for the flow from tunnel No. 173, concentration of ^{137}Cs was lower than the permissible concentration for drinking water. It can be supposed that waters permeating to the tunnels through fracture systems were not contaminated during nuclear tests.

The most widely spread water contamination is ^{3}H contamination. For example, of all analyzed flows high ^{3}H concentrations were registered in 21 tunnels, whereas high ^{137}Cs concentrations were detected only in 8 tunnels. Figure 8 shows that ^{137}Cs amount carried-over to the surface gradually decreased. It is caused not only by the decrease in flow intensity (Figure 7) but also by the decrease in ^{137}Cs concentration in flows due to sealing of tunnels portals. At least, ^{137}Cs specific activity in the same flow in most cases decreased after tunnel sealing. Calculation of average ^{137}Cs values before and after portal sealing is given in the figure (Figure 9).

^{137}Cs concentration in water flows probably decreases due to absorption processes. Indeed, before flow appearance on the surface its water is filtered through sealing structures including small-grained material with clay and silt particles. This material has better absorption properties for ^{137}Cs [7, 8].

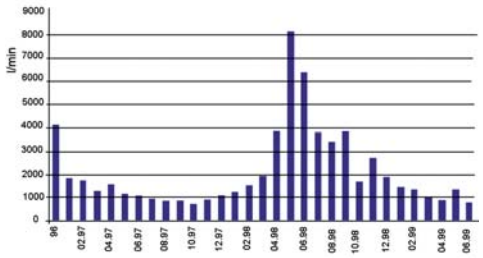


Figure 7. Total water flow from tunnels

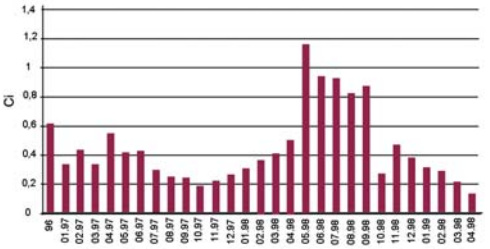


Figure 8. Total amount of ^{137}Cs monthly carried-over with water flows from tunnels

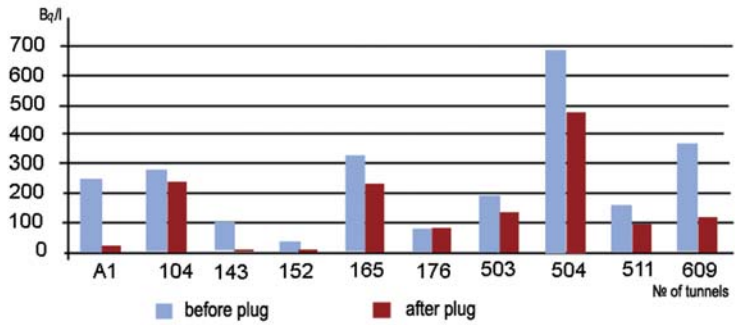


Figure 9. Average ^{137}Cs specific activity in water before and after tunnel conservation

1.4. Radioactivity on the "Degelen" test ground

1.4.1. Information on accumulated activity and its distribution in mountain rocks

Contamination of the central UNE zones by artificial radionuclides is the result of action of the following factors:

- remainders of fissionable material;
- fission fragments;
- radionuclides with induced activity.

To estimate possible contribution of blocks of mountain rocks into radioactive contamination caused by UNE, let us assume that all nuclear charges were made on the base of ^{239}Pu.

Remainders of fissionable material. Absolute activities were calculated according to the methodology described in [9]. The amount of fissionable material remaining to the moment when nuclear charge construction is destructed is determined by efficiency η of nuclear explosion which depending on the type and construction of nuclear device may vary from 1

to 30%. Due to the absence of official data on each explosion, in further calculations we take η equal to 20%.

The total number of underground tests on the "Degelen" test ground was 209. All power range of explosions in trinitrotoluol equivalent divides into two parts: 0.001 – 20 kilotons and 20 – 150 kilotons. Absolute activities were calculated for upper and lower limits, then the data for two groups were summed up giving minimal and maximal values of total activity for the given moment. The results obtained are presented in table 4.

Table 4.

Maximal and minimal possible total activity of plutonium and daughter isotope ^{241}Am on the "Degelen" test ground

Isotope	Half-life, years	Lower activity limit, Bq	Upper activity limit, Bq
^{238}Pu	87.7	6.9×10^{13}	1.2×10^{15}
^{239}Pu	2.4×10^{4}	1.9×10^{14}	3.4×10^{15}
^{240}Pu	6.5×10^{3}	6.0×10^{13}	1.1×10^{15}
^{241}Pu	14.4	6.9×10^{14}	1.4×10^{16}
^{242}Pu	3.7×10^{5}	7.3×10^{10}	1.3×10^{12}
^{241}Am	14.4	1.3×10^{14}	2.2×10^{15}

The real activity of ^{241}Pu may be a little higher due to ^{240}Pu activation by fission neutrons but the reaction cross-section is too small to make a considerable contribution. It is obvious that in case of uranium charge mountain rocks will contain uranium with increased concentration of ^{235}U.

Fission fragments. As in case of calculation of remainders of fissionable material, calculation of fission fragments was made for the two intervals of explosion capacity 0.001 – 20 kilotons and 20 – 150 kilotons with estimation of the upper and lower values of current total activity. This calculation as well as the previous one used the methodology described in [9]. The results of calculations are presented in Table 5.

Table 5.

Theoretical estimation of current minimal and maximal possible absolute activity of the most important radionuclides – fission products

Radionuclide	Half-life, years	Elaborated activity, Bq		Approximate ratio of the isotope activity to activity of ^{137}Cs
		Max	Min	
^{79}Se	65000	6.8×10^{9}	1.2×10^{11}	< 0.00001
^{90}Sr (^{90}Y)	28.5	8.1×10^{14}	1.5×10^{16}	0.74
^{99}Tc	213 000	1.7×10^{13}	3.0×10^{14}	0.015
^{121}Sn-m	50	4.2×10^{11}	7.5×10^{12}	0.0004
^{125}Sb	2.77	5.9×10^{10}	3.0×10^{12}	< 0.00006
^{137}Cs (^{137}Ba-m)	30.17	1.1×10^{15}	1.9×10^{16}	1
^{151}Sm	93	4.2×10^{11}	7.5×10^{12}	0.0004

Radionuclides with induced activity. Approximately 50% of fission neutrons formed in the explosion are absorbed in the nucleus charge, whereas the others spread in the environ-

ment. Passing through substance fission neutrons take part in elastic and inelastic scattering on nuclei losing their kinetic energy and in nuclear reactions (including radiation capture) whose probability rapidly increases as neutron energy decreases. It should be noted that the above processes undergo in plasma heated to several million degrees into which the substance surrounding nuclear charge turns when the chain reaction stops. The fission neutrons almost instantaneously thermalize, and their energy and, hence, absorption cross-sections are fully determined by rapidly changing plasma temperature. These neutrons are finally fully absorbed by the environmental nuclei forming new stable and radioactive isotopes. Therefore, in order to obtain a reliable estimation of the amount of an isotope formed in the environment, it is not sufficient to know chemical and isotope composition of the environment it is necessary to describe dynamics of processes in the plasma cloud.

The most important radionuclides, products of activation, are radioactive isotopes ^{152}Eu, ^{60}Co, ^{36}Cl and ^{14}C. These radionuclides are formed in the following nuclear reactions:

^{151}Eu (n, γ) ^{152}Eu; ^{59}Co (n, γ) ^{60}Co; ^{35}Cl (n, γ) ^{36}Cl; ^{14}N (n, p) ^{14}C

A special attention should be paid to such radioactive isotope as ^{3}H. In nuclear explosions the fusion reaction gives ~$7*10^{14}$ Bq/kt and fission reaction gives ~$4*10^{10}$ Bq/kt. The underground explosions make the above numbers higher due to nuclear reactions with boron and lithium in the rock structure [8,9]:

$$^{10}B_5 + n \rightarrow 2\ ^4He_2 + T; \qquad ^6Li_3 + n \rightarrow\ ^4He_2 + T$$

It should be expected that some part of tritium formed in the above reactions will remain in the crystalline rock and slowly diffuse to the surface of mineral structures with subsequent transition into water and air environments.

As a result of underground nuclear explosions the two main sources of radionuclides are formed in the central zone. The first one is a radioactive aerosol-dust component localized in crushed rock and massif fractures. The second source is a radioactive vitrified slaggy mass containing the main fraction of ^{90}Sr, ^{137}Cs and practically all $^{239+240}$Pu. Most radioactive isotopes (fission products) have form of inert gases or volatile elements during the period of time comparable with the time of cavity breakdown. The isotopes having form of inert gases do not condense until they decay into other elements, whereas more volatile substances do not condense until temperature decreases or they decay into less volatile elements. As the process of radioactive decay after explosion goes on, the distribution of gaseous and nonvolatile isotopes changes. The most part of ^{90}Sr is formed as an inert ^{90}Kr (about 80%) except for small amounts of directly formed ^{90}Sr or ^{90}Rb. Therefore if the cavity breaks down during the time comparable with ^{90}Kr half-life (33 s), the most part of final ^{90}Sr is not captured by molten material and is released from the cavity with other gases. It was found that even in case of full camouflet explosion, and formation of insoluble vitreous mass containing most part of decay products, the main fraction of ^{90}Sr and ^{137}Cs, existing in the form of inert gases (^{90}Kr and ^{137}Xe) during cavity breakdown, is not absorbed by the vitreous mass but moves with other gases [10, 11, 12]. Therefore, large amounts of ^{90}Sr and other isotopes, whose predecessors were gaseous or volatile elements, precipitate at rather large distances from the zone of high radioactivity and spread in the destroyed space. At the temperature in the explosion cavity as high as 1000–1500 °C, some radionuclides and fission products become volatile and escape from the vitreous material, which is typical, for example, for cesium.

Therefore, waters washing a block of contaminated mountain rocks must contain the following radionuclides: radioisotopes – residuals of fissionable substance (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am or ^{235}U), radioisotopes – fission fragments (^{90}Sr (^{90}Y), ^{99}Tc, ^{137}Cs), and radioisotopes – activation products (^{152}Eu, ^{60}Co, ^{36}Cl, ^{14}C, ^{3}H). Special attention must be paid to such radionuclides as ^{99}Tc, ^{36}Cl, ^{14}C, which have little absolute concentrations but high migration properties.

1.4. Radionuclide contamination of surface waters

As it was mentioned above tunnel waters are the main radioactive pollutant of surface and ground waters. The tunnel waters have rather low concentrations of artificial radionuclides ($^{239+240}$Pu, ^{90}Sr and ^{137}Cs), $n\times10^{-1}$–$n\times10^{3}$ Bq/l, but their high volumes gradually give quite noticeable values of environment contamination. The main sources of contamination are ^{90}Sr, ^{137}Cs and ^{3}H.

The figure (Figure 10) shows the results of long-term radionuclide monitoring of tunnel waters.

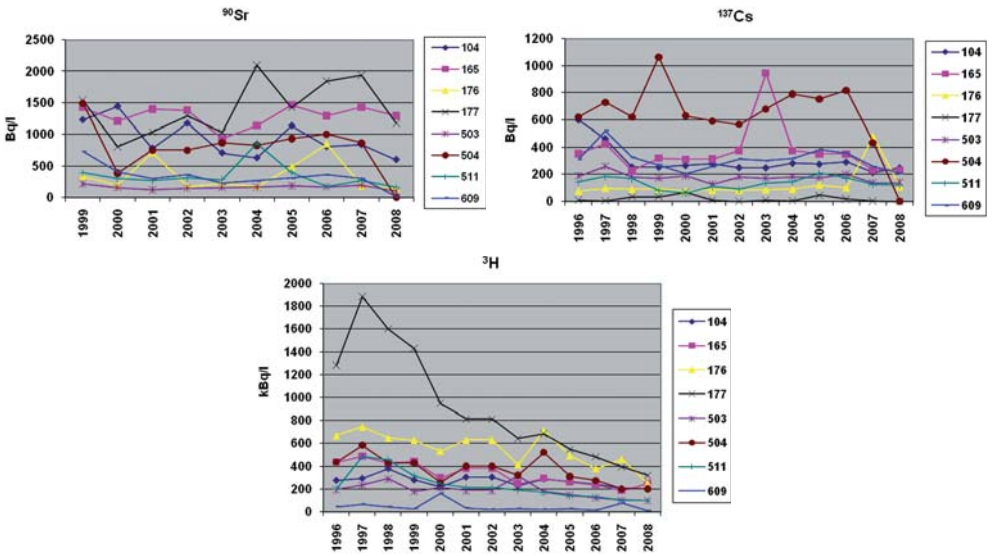


Figure 10. "Degelen" test ground. Monitoring of tunnel waters.

The curves of radionuclide monitoring in tunnel waters show that ^{3}H had maximal specific activity during all observation period in tunnel No. 177 with maximal concentration, 1900 kBq/kg, registered in 1997. A similar picture was observed for ^{137}Cs, with maximal concentration, 1100 Bq/kg, observed in 1999. Maximal ^{90}Sr concentration (2100 Bq/l) was registered in water of tunnel No. 177 in 2004.

The data of tunnel waters monitoring show that the radioactive contamination of waters of Degelen mountain massif still goes on and can be described as relatively stable [7, 13].

The concentrations of artificial radionuclides ($^{239+240}$Pu, ^{90}Sr, ^{137}Cs) in water flowing out from tunnels of Degelen mountain massif were studied in order to solve several problems, the most important of which was determination of the character of seasonal variations. The other task was estimation of efficiency of remediation measures, i.e. estimation of contamination

levels after conservation of some tunnels. An important component of investigations was more or less regular monitoring and observation of radioecological characteristics of this complex object.

Taking into account high radionuclide concentrations in waters flowing out from tunnels of Degelen mountain massif, in 13 tunnels more detailed observations of possible seasonal variations were made. General variations in the dynamics of radionuclide concentration in tunnel waters are shown in the figure (Figure 11).

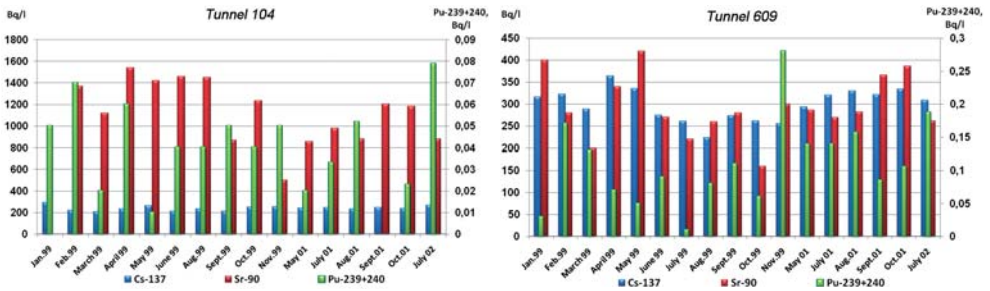


Figure 11. Results of tunnel monitoring in Degelen mountain massif

Table 6 presents the results of monitoring of tunnels in Degelen mountain massif for the period from 1999 to 2002. The radionuclide composition and the levels of contamination in all observed tunnels differ widely, but almost in all tunnels these characteristics practically do not change with time. However, ^{137}Cs concentrations vary significantly in tunnels Nos. 11 and 165, ^{90}Sr concentrations show variations in tunnels Nos. 104 and 165, and $^{239+240}$Pu concentrations vary in tunnel No. 504. A noticeable difference between maximal and minimal values for all analyzed radionuclides is registered in tunnel No. 177.

Table 6.

Results of tunnel monitoring in Degelen mountain massif (min-max / average)

Tunnel #	^{137}Cs, Bq/l	^{90}Sr, Bq/l	$^{239+240}$Pu, Bq/l	^{3}H, kBq/l
104	(210 – 290) / 240	(500 – 1540) / 1120	(0.01 - 0.08) / 0.04	(0.6 – 305) / 210
11	(27 – 400) / 110	(220 – 350) / 290	(0.03 - 0.15) / 0.1	-
143	(0.3 – 4) / 1.4	(150 – 310) / 220	(0.008 - 0.03) / 0.01	-
156	(0.2 – 8) / 3	(130 – 175) / 150	(0.006 - 0.13) / 0.05	(6 – 140) / 23
165	(1.2 – 360) / 260	(120 – 1700) / 1080	(0.004 - 0.09) / 0.02	(50 – 440) / 230
176	(80 – 100) / 90	(187 – 260) / 210	(0.004 - 0.03) / 0.01	(30 – 710) / 370
177	(0.8 – 610) / 50	(620 – 1650) / 970	(0.11 – 12) / 3.7	(160 – 1430) / 430
503	(170 – 180) / 175	(147 – 200) / 160	(2 - 5.6) / 4	(75 – 310) / 140
504	(590 – 755) / 690	(800 – 1160) / 920	(0.01 - 0.4) / 0.13	(20 – 520) / 240
506	(0.4 – 17) / 4	(160 – 225) / 180	(0.004 - 0.02) / 0.01	(50 – 135) / 100
511	(117 – 140) / 130	(240 – 350) / 280	(0.06 - 0.1) / 0.08	(40 – 320) / 160
609	(220 – 360) / 300	(160 – 420) / 290	(0.01 - 0.3) / 0.11	(3 – 165) / 20
802	(0.2 – 14) / 4	(45 – 70) / 55	(0.003 - 0.013) / 0.01	(65 – 185) / 120

A comparison of the table data (1999–2002) and results of long-term monitoring (1999–2008) shows that the character of contamination does not change with time. In other words, the analysis of obtained results did not reveal any considerable seasonal variations in concentrations of ^{137}Cs, ^{90}Sr and $^{239+240}$Pu, and differences in levels can be explained by instability of physical-chemical properties of analyzed nuclides in water.

In addition to the main dose-forming artificial radionuclides, concentrations of ^{99}Tc and ^{36}Cl were determined in tunnel waters. The results of laboratory analysis of their concentrations in water samples are presented in table 7.

Table 7.

^{99}Tc concentration in water samples

##	Sampling place	Sampling site	^{99}Tc		^{36}Cl
			ng/l	mBq/l	Bq/l
1	Degelen	Tunnel 165	0.054±0.027	34±17	0.25 ± 0.05
2	Degelen	Tunnel 511	<0.003	<2	
3	Degelen	Tunnel 504	<0.003	<2	0.21 ± 0.05
4	Degelen	Tunnel 503	<0.003	<2	0.37 ± 0.09
5	Degelen	Tunnel 176	0.013±0.007	8.2±4.1	0.32 ± 0.08
6	Degelen	Tunnel 609	0.035±0.017	22.0±11.0	
7	Degelen	Tunnel 104	0.21±0.10	130.0±65.0	0.31 ± 0.04
8	Degelen	Tunnel 177	0.18±0.09	112.0±56.0	
9	Degelen	Tunnel 385			0.25 ± 0.13
10	Kurchatov	Irtysh river	<0.003	<2	

Table 7 shows that ^{99}Tc concentration in tunnel waters varies in a wide range, from <2 to 130 mBq/l. Maximal concentration reaching 130 mBq/l was registered in tunnel No.104. The monitoring revealed the presence of ^{36}Cl in tunnel waters with concentration not exceeding 0.4 Bq/l, which is undoubtedly a safe level, however, the fact of its presence is important as this factor can be used as an indicator of migration processes.

To study migration properties of artificial radionuclides in surface streams at different distances from tunnel portals water samples were taken. The laboratory analyses showed the following picture – the highest concentrations of ^{137}Cs and ^{90}Sr in water are registered at outflow of streams from tunnels. The radionuclide concentrations decrease as the distance from tunnels increases. Figure 12 shows that maximal $^{239+240}$Pu, ^{137}Cs and ^{90}Sr concentrations are registered near tunnel portals.

As distances from portals increase, radionuclide concentrations decrease which is most probably cause by sorption and precipitation of radioactive particles. However, some radionuclides, probably in dissolved or colloid state, migrate at large distances. The largest distances along stream riverbeds are covered by ^{90}Sr [1].

Therefore, migration properties of artificial radionuclides in water contacting with bottom sediments can be described by the following series ^{3}H>^{90}Sr>^{137}Cs>$^{239+240}$Pu. It should be noted that concentrations of all radionuclides at some distance (rather large distance) fall below the interference level for population [14] if they are taken with water and such water can be used for household needs.

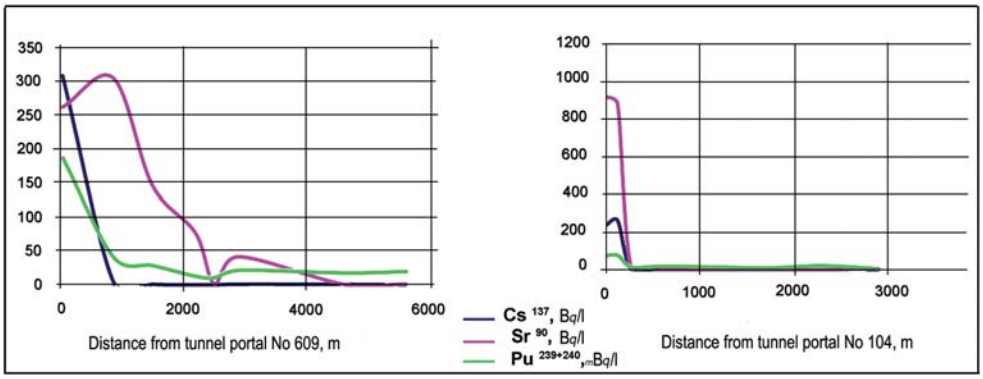


Figure 12. Radionuclide concentration as a function of distance from tunnel portals

Streams. The dynamics of streams discharge and concentrations of artificial radionuclides in surface waters of the main surface water ways: the Uzynbulak, Karabulak and Baitles streams, in the sections where they flow out from Degelen mountains, showed the following picture. As a rule, in the first quarter of a year there is no surface water runoff in this part of the stream because of low temperatures. In April water appears in the Baitles stream and the right confluent of the Karabulak stream.

The most stable surface stream is the Uzynbulak, where maximal flow intensity is observed in May and June due to snow melting and atmospheric precipitations (Table 8).

Table 8.

Radionuclide concentration in surface streams

##	Sampling point	Date	Flowrate, l/min	Specific activity of radionuclides	
				^{137}Cs, Bq/l	^{3}H, kBq/l
1	Uzynbulak stream	04.2001 06.2001 10.2001 06.2002	14,276 3,103 2,948 459	<2 No data 5±2 <2	108±7 91±8.4 173±14 108±9
2	Baitles stream	04.2001 03.2002	3,000 105	3.5±1.9 <2	130±11 142±12
3	Right arm of Karabulak stream	04.2001 03.2002	1,588 1,688	6.6±2 <2	86±8 57±6

Table 9 shows that streams when they have water flows carry intensive ^{3}H contamination. The highest ^{3}H concentration is registered in the waters of the Baitles stream forming on southern slopes of Degelen mountains and the Uzynbulak stream having the largest watershed area on the considered territory. The presence of ^{137}Cs is registered sporadically in amounts below permissible values for drinking water. The absence of ^{137}Cs in water can be probably explained by high absorption of silt and clay particles in alluvial sediments where it can precipitate [7].

2. RADIONUCLIDE MIGRATION OUTSIDE DEGELEN MOUNTAIN MASSIF LIMITS

In 1999 the investigations organized to determine parameters of radionuclide distribution in water beyond the bounds of the mountain massif started. To study characteristics of ground waters in all main valleys going down the mountains in different directions hydrogeological wells were drilled (Figure 13).

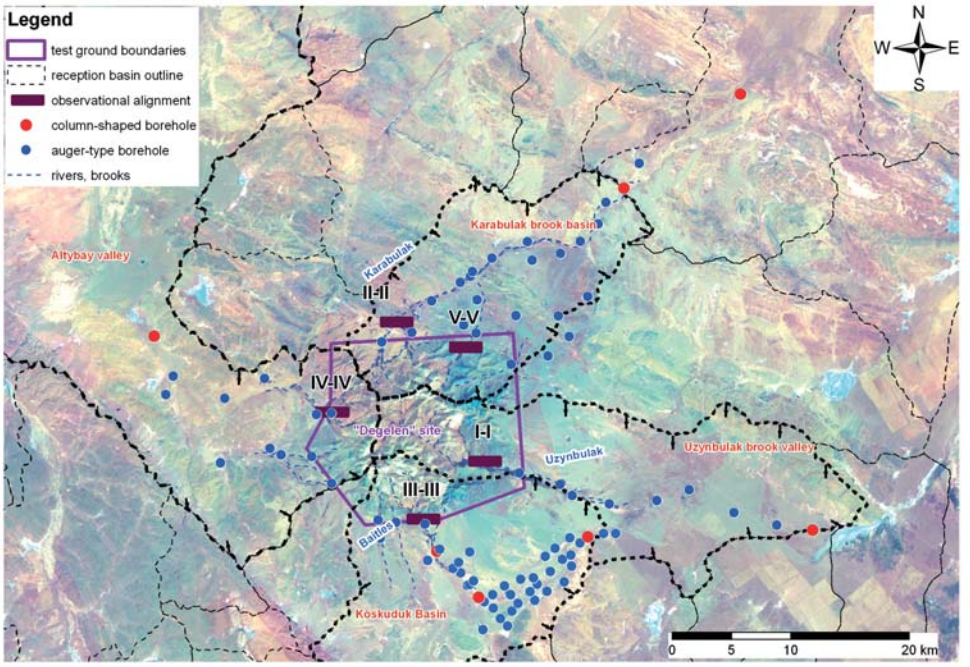


Figure 13. Degelen mountain massif. Location of the main valleys

This chapter describes the results of radioecological investigations of ground waters in the valleys of the Uzynbulak, Baitles, Karabulak and Altybay streams.

2.1. The Uzynbulak stream area

2.1.1. Hydrogeological conditions in the Uzynbulak stream valley

According to the available data the Uzynbulak valley is the main waterway of Degelen mountain massif and, hence, the main channel of migration of UNE radioactive products. In this connection more detailed investigations on radioactive contamination of ground waters were carried out in the Uzynbulak valley.

Section of the Uzynbulak valley inside Degelen mountain massif

Figure 14 shows the scheme of location of wells drilled to study porous waters in the Uzynbulak valley inside Degelen mountain massif.

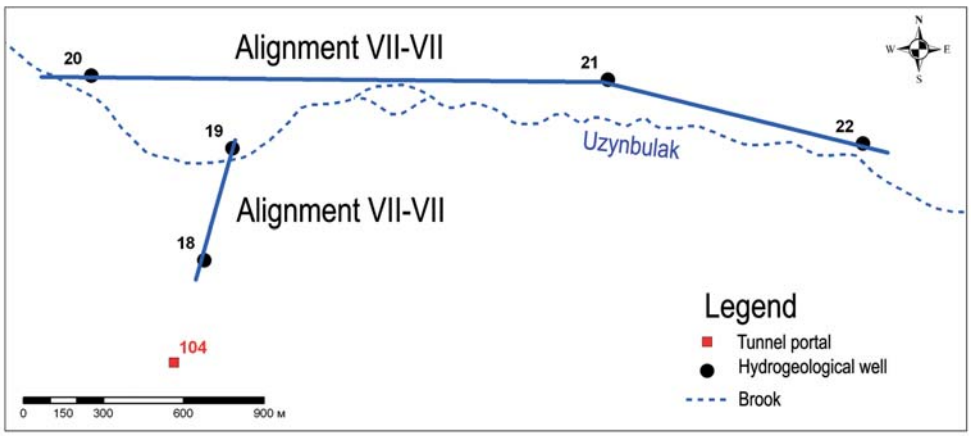


Figure 14. Location of hydrogeological wells in the vicinity of tunnel No. 104

The main task of this research was studying of possible contribution of tunnel waters to radioactive contamination of ground waters. Two hydrogeological observation stations shown in figure 14 were made in the vicinity of tunnel No. 104. The main task of wells in the observation station VI-VI is studying of the water flow in the section between tunnel No. 104 and the Uzynbulak riverbed. Two hydrogeological wells 18 and 19 were drilled into ground waters migrating in the stream direction. Well 18 is located at the end of a broad gully along which water from tunnel No. 104 flows to the Uzynbulak valley. It is the place where the swampy area formed by the stream from the tunnel ends, water goes underground and flows as a stream in alluvial-proluvial sediments. Well 19 is located near the place where the ground waters of the above stream join subsoil waters of the Uzynbulak valley. Station VII-VII is located along the Uzynbulak riverbed; its task is to study subsoil waters in the largest valley of Degelen mountains with vast watershed. 3 wells, 20, 21 and 22, were drilled in the hydrological station. Well 20 was drilled at a distance of 700 m from well 19 upstream. It was drilled to estimate groundwater contamination upstream the inflow of the ground stream from tunnel No. 104. Wells 21 and 22 are located downstream, 1.2 and 2.2 km from the inflow of the side stream from tunnel No. 104.

The depth of wells drilled into groundwater horizons is not more than 4.2 m. The upper part of the section has a soil-vegetation layer with underlying water-bearing loams. At a depth of 1.4-5.0 m underlies the layer of sands with crushed rock. The level of ground waters is located at a depth of 0.6-1.9 m. The data for wells are presented in table 9.

Table 9.

Parameters of hydrogeological wells

River, section No.	Well No.	Well depth, m	Ground water level, m
VI-VI, Uzynbulak riv.	19	4.0	1.9
	18	4.2	1.1
VII-VII, Uzynbulak riv.	20	4.0	1.1
	21	3.8	0.6
	22	3.2	0.6

Section where the Uzynbulak valley outcomes from Degelen mountain massif. To study the state of porous waters in the section where the Uzynbulak valley outcomes beyond Degelen mountain massif, the section I-I was made, it consists of 4 wells: 10, 11, 12 and 13, three of which were drilled into subsoil waters (Figure 15).

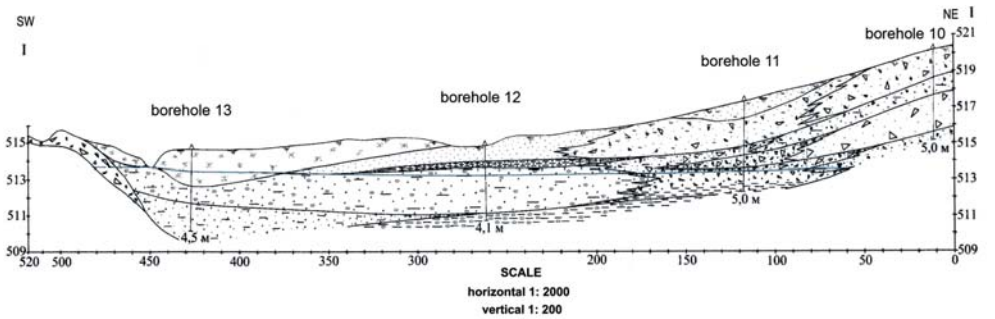


Figure 15. The Uzynbulak stream. Geological section along the section line I-I

The section line I-I is located in the Uzynbulak stream valley, at a distance of 2.5 km to the east from tunnel No. 104 (Figure 14). The section line is directed to the northwest at an angle of 10^0. The section line length is 520 m. The main information about wells is presented in table 10.

Table 10.

Information on hydrogeological wells

River, section No.	Well No.	Well depth, m	Ground water level, m
I-I, Uzynbulak riv.	10	5.0	No water
	11	5.0	3.8
	12	4.1	1.6
	13	4.5	1.3
	16	45.5	2.85
	17	43.0	2.9

The aquifer connected with surface stream waters is formed by rubble-gravel and gravel sediments with sand filler. The section line is formed by the waterproof Neocene clayes which separate subsoil porous waters from the regional basin of fracture waters.

It should be noted that in the vicinity of section line I-I there are waterproof clays which separate subsoil waters from the aquifer of fracture waters. To study the distribution of artificial radionuclides in ground porous and fracture waters, hydrogeological wells 16 and 17 were drilled in section line I-I (Figure 16).

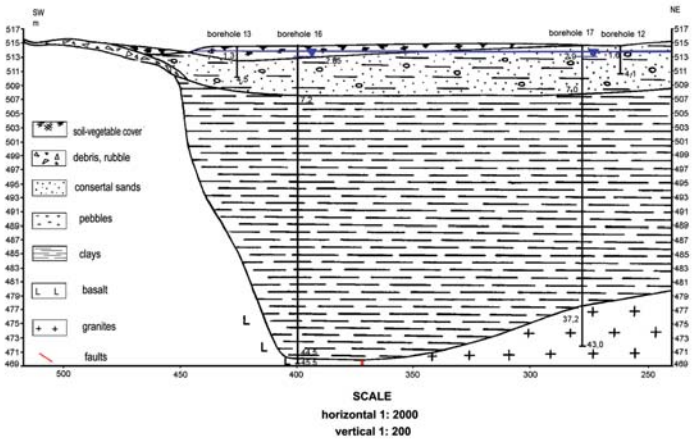


Figure 16. Geological section along the section line of wells 16 and 17

The horizon of porous subsoil waters is isolated by casing pipes to a depth of 8 m. The clay thickness in the well is 30–37.3 m. The level of subsoil waters has a depth of 2.85–2 m. The piesometric level of pressure fracture waters is 1.3–1.55 m below the level of subsoil waters. Along the deepest section of the Paleozoic bedding is observed a fracture zone separating left-side granites from basalts of the right-side part of the Uzynbulak valley. That is the reason why rocks enclosing water-bearing horizon have different composition.

6 auger wells (1U, 2U, 3U, 4U, 5U, 6U) were drilled to study the state of porous waters beyond the bounds of Degelen mountain massif within the Uzynbulak microbasin, and one exploratory well (34P) was drilled to study fracture waters (Figure 17).

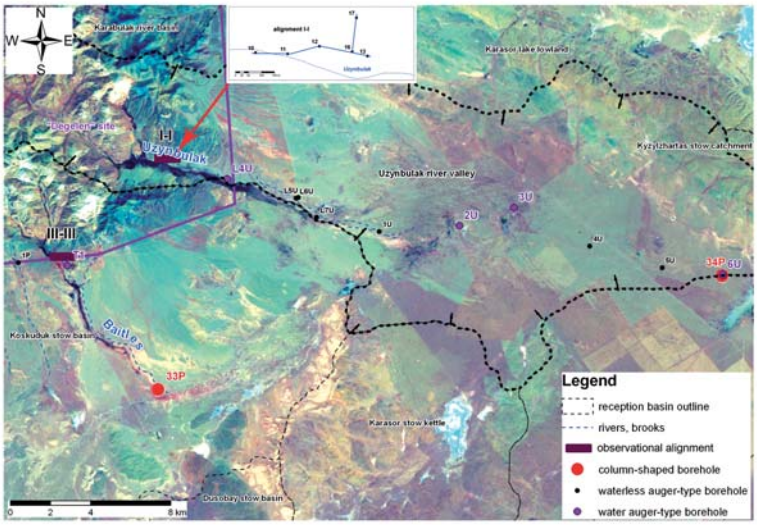


Figure 17. "Degelen" test ground. Wells location in the Uzynbulak microbasin.

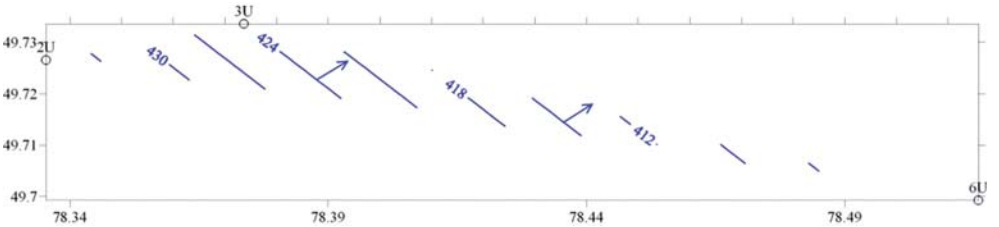
Table 11 presents information about 6 wells drilled to study hydrogeological characteristics.

Table 11.

Degelen mountain massif. Wells drilled to study hydrogeological characteristics of microbasins

##	Well No	Coordinates		Well depth, m	Ground water level, m
		L.N.	L.E.		
1	1U	49° 43' 30,9"	78° 16' 50,0"	15.0	No water
2	2U	49° 43' 34,7"	78° 20' 10,1"	5.0	3.5
3	3U	49° 44' 0,0"	78° 22' 28,0"	4.7	3.0
4	4U	49° 42' 52,2"	78° 25' 32,6"	15.0	No water
5	5U	49° 42' 12,0"	78° 28' 30,2"	15.0	No water
6	6U	49° 41' 56,3"	78° 30' 59,9"	6.0	2.7
7	34P	49° 41' 53,89"	78° 31' 1,62"	52.0	2.0

The experimental data were used to make the map of isohypses of the water-bearing horizon of porous waters (Figure 18).



The arrow shows the direction of groundwater motion

Figure 18. Maps of isohypses of the water-bearing horizon of porous waters in the Uzynbulak microbasin

An analysis of the isohypses map shows that ground waters in the Uzynbulak microbasin are filtered in the southeast direction. To determine characteristics of water-bearing horizon in the Quaternary sediments, in 3 water wells out of 6 auger wells the exploratory-filtration works, i.e. well fillings with water, were carried out. Wells were filled constant discharge (Q = const). Water measurements were used to calculate the filtration coefficient (*K*) and specific water consumption (*q*). The results of exploratory-filtration works are given in table 12.

Table 12.

"Degelen" test ground . Filtration parameters obtained by the results of well-water tests

##	Well No.	K, m/day	Q, m^3/h	q, m^3/h
1	3U	0.10	0.10	0.02
2	6U	0.06	0.07	0.09

Filtration characteristics of the water-bearing rocks can be classified as low-permeated, and in the well 6U the rocks are very low-permeated (*K>0.1 m/day*) [6].

Therefore hydrogeological investigations of ground waters spread within the Uzynbulak valley showed the following pictures:

- two types of ground waters, porous and fracture, are typical in the territory of the Uzynbulak valley;
- the water-bearing horizon of porous waters is presented by rubble-pebble and gravel sediments with sand filler, fracture waters are contained in granites, basalts and alevrolytes;
- porous waters are isolated from fracture waters by clayish sediments, whose thickness goes up to 40 m;
- the groundwater level inside the Degelen mountain massif has depths varying from 0.6 to 2.0 m. The groundwater level beyond the mountain terrain decreases to 3.0 m.
- the fracture water level within the section line I-I is within 4.5 m, beyond the bounds of mountain massif in well 34P it is at a depth of 2 m;
- ground waters in the Uzynbulak microbasin filter in the southeast direction. According to filtration characteristics the water-bearing rocks can be classified as low-permeable and very low-permeable.

2.1.2. Radionuclide contamination of ground waters in the area of the Uzynbulak stream

Table 13 below presents the results of laboratory analyses of ground waters sampled from wells.

Table 13.

"Degelen" test ground . Results of laboratory analyses of groundwater samples

Well No.	^{137}Cs, Bq/kg	^{90}Sr, Bq/kg	$^{239+240}$Pu, Bq/kg	^{3}H, kBq/kg
13 (section I-I)	<2	*	*	130±12
18 (section VI-VI)	<2	*	*	185±17
19 (section VI-VI)	<2	*	*	80±7
21 (section VII-VII)	<2	*	*	140±45
22 (section VII-VII)	<2	*	*	105±9
16 (section I-I)	<2	*	*	40±4
17 (section I-I)	<2	*	*	*
L4U	<0.05	3.00±0.03	0.72±0.02	51±5.1
2U	<0.01	<0.01	0.03	2.2±0.22
3U	<0.01	<0.01	0.05	0.045±0.007
6U	<0.05	2.5±0.2	<0.001	23±2.3
34P	<0.02	<0.01	*	0.06±0.01
* - no measurement				

Studying of the distribution of artificial radionuclides in porous waters in the vicinity of tunnel No. 104 gave the following picture. The ^{3}H concentration in the subsoil water in well 18 located in the same gully as tunnel No. 104 is comparable with its concentration in the tunnel stream. Taking into account the identity of water chemical composition, one can suppose that the main source of subsoil waters in well 18 is the flow from tunnel No. 104. It is

important to note the absence of ^{137}Cs in well 18 though its concentration in the tunnel stream is 190–230 Bq/l. This phenomenon can be explained by high sorption ability of sediments through which the tunnel waters filter. One can suppose that the area of the ^{137}Cs contamination by tunnel flows is limited by the vicinity of the portals.

The sample water in well 19, located in the central part of the valley opposite well 18, has lower ^{3}H contamination. These samples characterize subsoil waters flowing from the upper part of the valley. In the lower parts of the valley ^{3}H concentration in ground waters increases, especially in the vicinity of well 21. It is the place of inflow of the left stream, in the sides if which 10 tunnels including tunnels Nos. 802, 195, 196 and 203 are located. The surface flow from tunnel No. 802 is still occasionally observed. In the other above-mentioned tunnels surface flows were only registered before portal sealing. The tunnel waters have high ^{3}H contamination. As one moves with subsoil waters in the direction of section line I-I, the ^{3}H concentration decreases to the level of ^{3}H concentration in the surface flow of the Uzynbulak stream.

In the groundwater flow outgoing from Degelen mountains the ^{137}Cs concentration in subsoil waters is at the level of maximal permissible activity (MPA). The ^{3}H concentration in ground waters is much higher than the intervention level for population for its intake with water, equal to 7700 Bq/kg, and goes up to 130 kBq/kg. At the same time in the ground waters located under the Neocene clays (wells 16 and 17) in the same area, the ^{3}H concentration varies from 10 to 40 kBq/kg, which is lower than in all other sampled Degelen waters. It is most likely that this area is characterized by low water exchange and stagnation processes.

Studying of carry-over of artificial radionuclides along the Uzynbulak valley with the groundwater flow outside the Degelen mountain massif showed that ^{137}Cs concentration in all studied groundwater samples was below the MPA. The quantitative concentrations of ^{90}Sr were only measured in wells 6U (2.5 Bq/kg) and L4U (3.0 Bq/kg). The presence of ^{90}Sr in the wells is most likely explained by their location within the boundaries of the Degelen mountain massif, where the radionuclide presence is usually registered in ground waters.

The presence of $^{239+240}Pu$ was registered in three wells: 3U, 2U and L4U, its concentration varied from 0.03 to 0.7 Bq/kg. The increase in $^{239+240}Pu$ concentration is most probably caused by closeness of the well to the UNE places on the "Degelen" test ground . It should be noted that the presence of $^{239+240}Pu$ in ground waters of the Degelen mountain massif is not typical, therefore it is necessary to recheck the fact and to study it in more detail.

The main radionuclide causing radioactive contamination is ^{3}H. The ^{3}H concentration in subsoil waters within the main riverbed varied from 185 to 23 kBq/l. The sampling registered general decrease in ^{3}H concentration in the direction from well 3U (0.045 kBq/kg) but considerable increase in its concentration to well 6U (23 kBq/kg). This phenomenon can be explained by two factors. Migration of artificial radionuclides with ground waters along the Uzynbulak valley and in other studied directions (Karabulak and Altybay) probably has a vein (cobweb) structure. In such cases radionuclide concentrations in one direction may change by tens of times and even more. The other reason for sharp increase in ^{90}Sr and ^{3}H concentration may be inflow of contaminated ground waters from the test ground "Telkem" as well 6U is located at a distance of only 4.5 km from the epicenters of excavation nuclear explosions "Telkem-2".

The results of auger drilling in the Uzynbulak region were used to choose the place for hydrogeological well 34P. The well was drilled into fracture ground waters. The ^{3}H concen-

trations in fracture ground waters (0.06 kBq/kg) in that point are much lower than in porous waters (23 kBq/kg), which indicates no connection between the two water types in that area [7, 8].

2.2. The Baitles stream area

2.2.1. Hydrogeological conditions of the Baitles stream area

The Baitles stream valley is located in southeastern part of the Degelen mountain massif. The Baitles riverbed is observed to a distance of 10 km from Degelen mountains. The surface waters in the stream appear seasonally.

To study the state of porous waters beyond the bounds of Degelen mountains the section line III-III consisting of three wells 4, 5 and 6 was made (Figure 19).

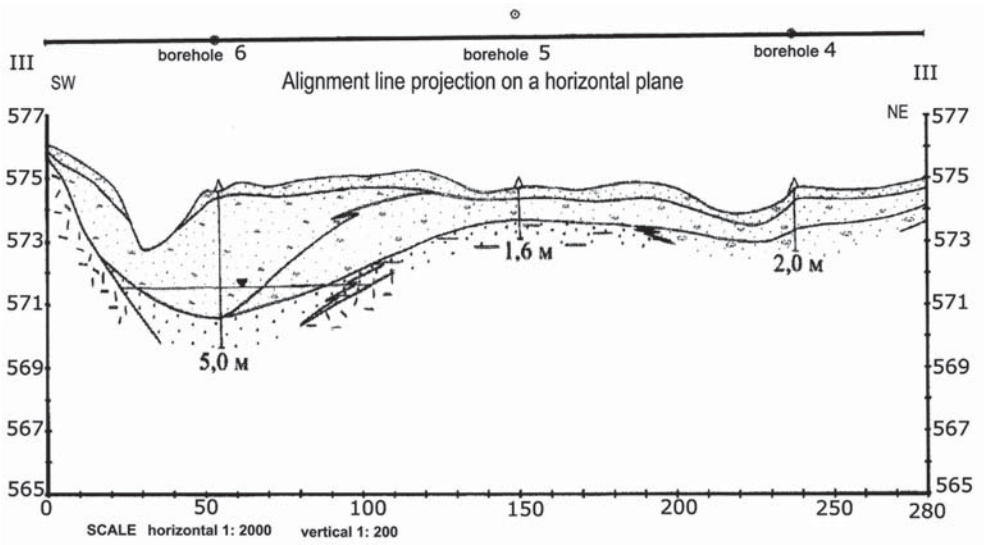


Figure 19. The Baitles stream. Geological section along the section line III-III

Section line III-III is located in the Baitles valley, at a distance of 4 km to the south from tunnel No.609. The section line direction is 55^0 northeast, the length is 280 m. The valley riverbed is well seen in the section and shifted in the direction of southwestern side. The thickness of alluvial sediments in the riverbed is over 5 m (well 6), the ground waters lie at a depth of 3 m. The alluvial sediments are presented by water-bearing loams and sands. Wells 4 and 5 were drilled into boulder-blocky grounds, which do not contain ground waters.

To study the state of ground waters spread within the Baitles stream area, 40 auger wells of a depth up to 6 m (11, 30A, 36) were drilled in the Koskuduk tract (Figure 20). Three observation wells (11, 30A, 36) were equipped for long-term monitoring of porous waters and one exploratory well (33P) was equipped for studying fracture waters. The coordinates of wellheads and the results of measuring of groundwater levels in the wells containing water are presented in table 14.

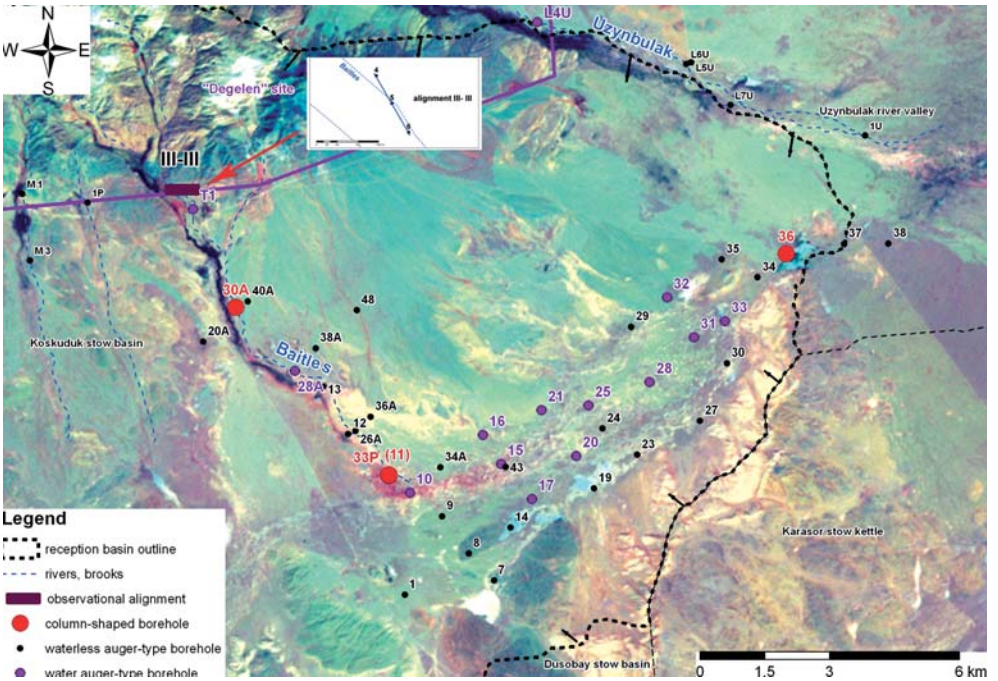


Figure 20. Baitles stream valley. Scheme of wells location

Table 14.

The Baitles section. General information on hydrological wells

##	Well No	Coordinates		Ground water depth, m
		L.N.	L.E	
1	20A	78° 03' 55,6"	49° 41' 17,1"	No water
2	30A	78° 04' 34,7"	49° 41' 41,8"	2.1
3	40A	78° 04' 48,7"	49° 41' 46,0"	No water
4	28A	78° 05' 39,8"	49° 40' 52,7"	2.1
5	38A	78° 06' 05,0"	49° 41' 08,8"	No water
6	48	78° 06' 54,4"	49° 41' 36,0"	No water
7	13	78° 06' 11,9"	49° 40' 40,1"	No water
8	26A	78° 06' 47,0"	49° 40' 06,0"	No water
9	36A	78° 07' 05,0"	49° 40' 16,1"	No water
10	12	78° 06' 38,4"	49° 40' 03,9"	No water
11	11	78° 07' 23,2"	49° 39' 31,7"	3,15
12	10	78° 07' 46,6"	49° 39' 17,9"	2,5
13	34A	78° 08' 23,2"	49° 39' 35,9"	No water
14	16	78° 09' 13,6"	49° 39' 58,9"	5,4
15	21	78° 10' 22,6"	49° 40' 15,7"	3
16	29	78° 12' 10,1"	49° 41' 14,9"	No water

##	Well No	Coordinates		Ground water depth, m
		L.N.	L.E	
17	32	78° 12' 53,1"	49° 41' 35,8"	2,8
18	35	78° 13' 57,9"	49° 42' 02,5"	No water
19	9	78° 08' 22,9"	49° 38' 59,3"	No water
20	15	78° 09' 33,1"	49° 39' 36,4"	2,8
21	43	78° 09' 38,5"	49° 39' 34,2"	No water
22	25	78° 11' 16,8"	49° 40' 17,6"	2,5
23	1	78° 07' 36,0"	49° 38' 01,4"	No water
24	8	78° 08' 51,9"	49° 38' 30,5"	No water
25	14	78° 09' 41,0"	49° 38' 48,5"	No water
26	17	78° 10' 07,2"	49° 39' 09,3"	2,25
27	20	78° 11' 00,4"	49° 39' 40,1"	2,35
28	24	78° 11' 31,9"	49° 39' 59,9"	No water
29	28	78° 12' 28,7"	49° 40' 33,1"	2,8
30	31	78° 13' 22,3"	49° 41' 05,0"	3,2
31	33	78° 13' 58,5"	49° 41' 15,9"	2,5
32	34	78° 14' 38,5"	49° 41' 47,9"	No water
33	36	78° 15' 12,6"	49° 42' 04,6"	1,5
34	37	78° 16' 20,2"	49° 42' 10,3"	No water
35	38	78° 17' 11,4"	49° 42' 09,1"	No water
36	7	78° 09' 19,4"	49° 38' 09,5"	No water
37	19	78° 11' 19,3"	49° 39' 15,4"	No water
38	23	78° 12' 10,8"	49° 39' 39,2"	No water
39	27	78° 13' 25,1"	49° 40' 02,6"	No water
40	30	78° 13' 59,0"	49° 40' 44,8"	No water
41	33P	78° 07' 26,04"	49° 39' 30,73"	4,6

Well 30A is located at the foothills of Degelen mountains, which enables to register the total amount of artificial radionuclides inflowing into the underflow waters of the Baitles stream. Well 11 was drilled at the Baitles outcome to the valley, which enables to control waters inflowing into valley ground waters. Well 36 is located at the outcome from the valley mainly fed by the Baitles waters to the Uzynbulak valley. This observation well enables to sample waters inflowing into the Uzynbulak valley. Therefore, the above wells enable to control radionuclide movement along the Baitles valley to the Koskuduk tract and further to the Uzynbulak valley.

Table 15 shows that out of 41 drilled wells the water horizon was reached only in 16 wells. Most waterless wells are located at the periphery of the studied area.

Exploratory-filtration works using water filling were carried out in 10 wells. The results of calculations are given in table 15.

Table 15.

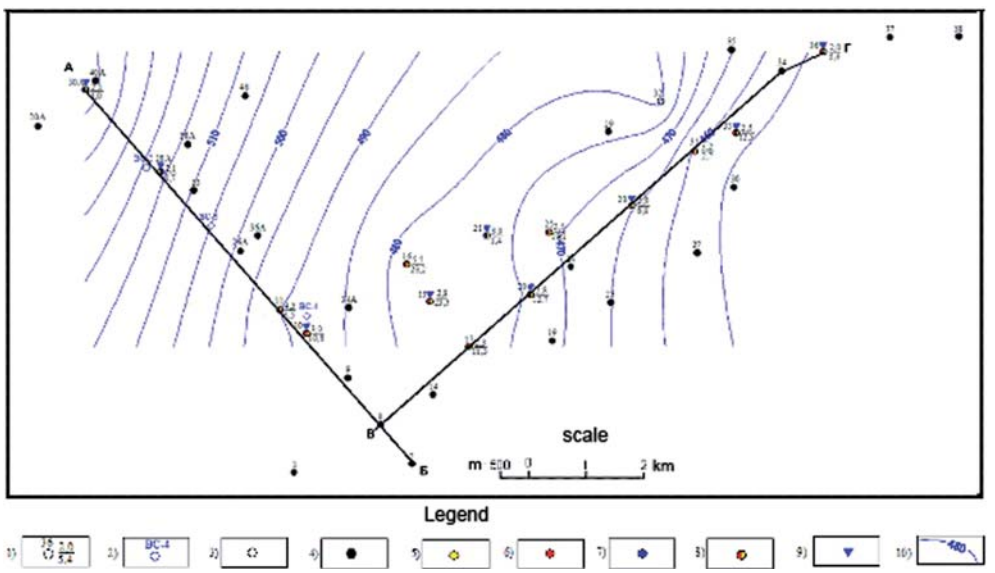
Results of exploratory-filtration works in wells

##	Well No.	K, m/day	q, m^3/h	##	Well No.	K, m/day	q, m^3/h
1	30A	1.2	0.34	6	32	2.0	0.45
2	20	1.4	0.51	7	28	1.6	2.13
3	28A	1.5	0.32	8	15	1.2	3.17
4	21	1.5	0.49	9	10	1.3	0.96
5	36	0.9	0.48	10	33	0.9	0.49

Table 16 shows that filtration rate varies within 0.9-2.0 m/day, with different values for northwest and south parts of the studied area. The values of specific water absorption vary in a wide range: from 0.45 m^3/hour (well 32) to 3.17 m^3/hour (well 15).

These data were used to make a schematic hydrogeological map (Figure 21).

The ground waters are located in the Upper Quaternary diluvial-proluvial sand-gravel sediments. The water-bearing horizon is overlaid by Quaternary loamy sands and loams, from the underneath it almost everywhere has waterproof clays, supposedly of Neocene age, which isolate horizon of porous waters, penetrated by wells, from the underlying horizon of fracture waters. The depth of subsoil waters ranges from 1.5 to 3.2 m (in well 16 – 5.4 m). Thickness of the water-bearing horizon varies from 1.0 to 2.0 m, the ground waters are unconfined, only in places with clayish lenses they have a small head (up to 0.5 m).



Well: 1 – well number (at the top); water level, m (numerator), water mineralization in g/dm^3 (denominator); 2 – well drilled in 2007; 3 – well without determination of chemical composition; 4 – a waterless well. Chemical composition of ground waters: 5 – prevalence of sulphate anion, 6 - prevalence of chloride anion, 7 - prevalence of hydrocarbonate anion, 8 – mixed composition; 9 – test water filings, 10 - hydroisohypses with absolute line altitude in m

Figure 21. The Baitles stream microbasin. Schematic hydrogeological map

Schematic hydrogeological sections were constructed along lines A-B and C-D denoted in figure 21 (Figure 22).

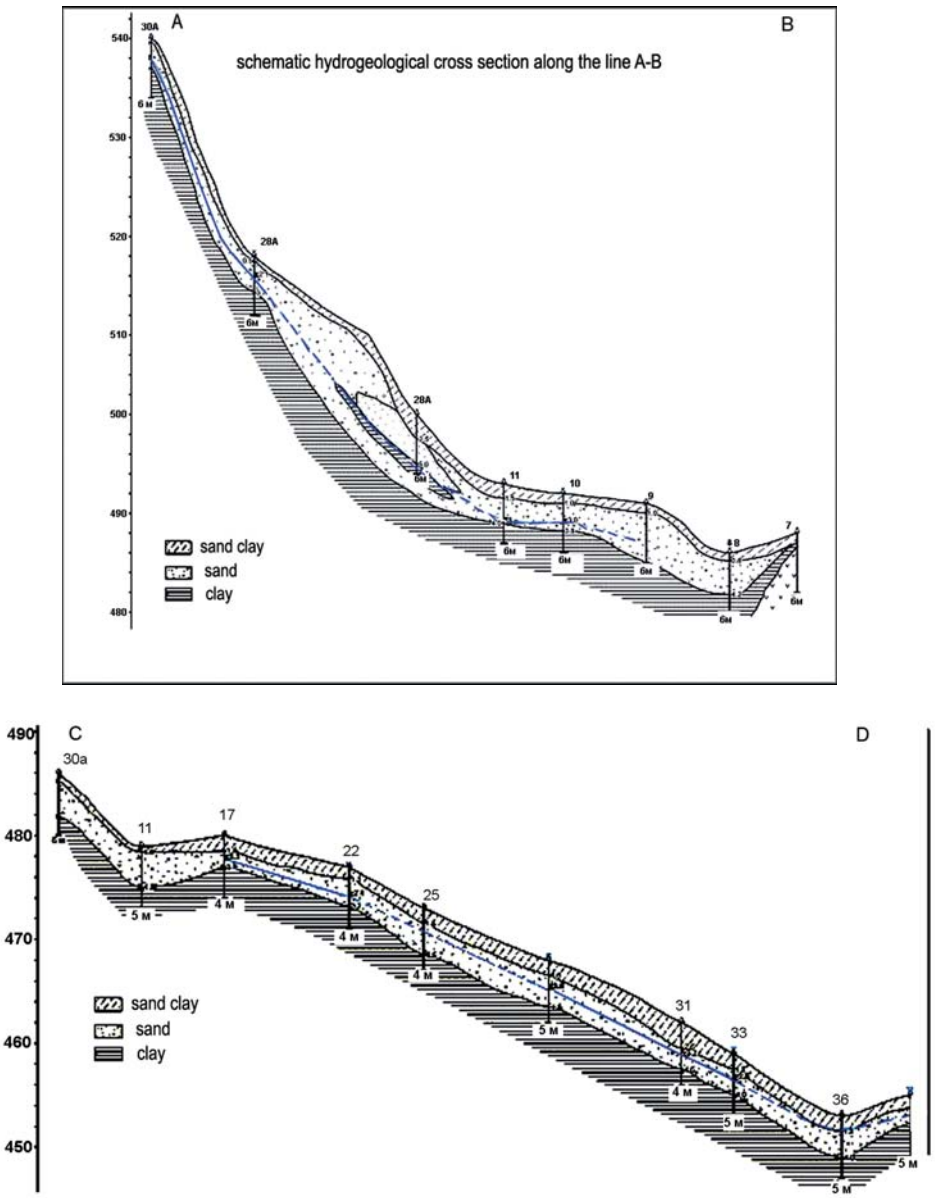


Figure 22. The Baitles stream area. Schematic hydrogeological sections

The obtained data were used to construct a three-dimensional model of the ground flow surface (Figure 23).

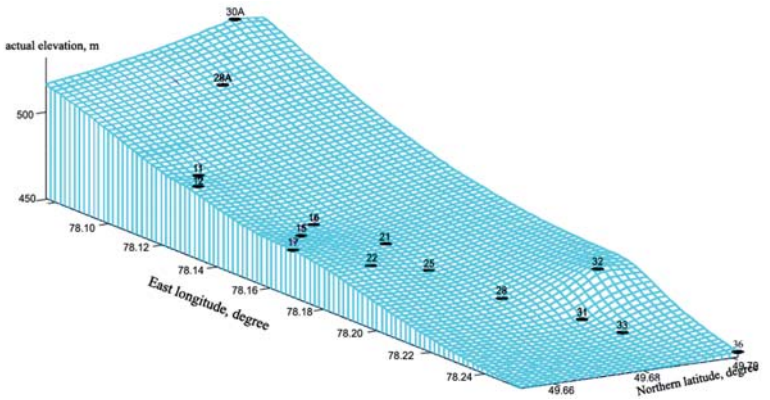


Figure 23. The Baitles stream area. Three-dimensional model of the ground flow surface

The results of hydrogeological explorations of ground waters spread in the Baitles valley enable to make the following conclusions:

- two types of ground waters, porous and fracture, are widely spread in the territory of the Baitles valley;
- The porous waters are located in the Upper Quaternary diluvial-proluvial sand-gravel sediments, fracture waters are located in sandstones;
- The depth of subsoil waters ranges from 1.5 to 3.2 m (in well 16 – 5.4 m). The thickness of the water-bearing horizon varies from 1.0 to 2.0 m, the ground waters are unconfined, only in places with clayish lenses they have a small head (up to 0.5 m). The fracture waters in well 33P are at a depth of 4.6 m;
- The filtration rate of porous waters ranges from 0.9 to 2.0 m/day, having different values in northwestern and southern part of the studied area.

2.2.2. Radionuclide contamination of ground waters in the Baitles section

In laboratory analyses of groundwater samples the concentrations of radionuclides ^{137}Cs, ^{90}Sr and ^{3}H were determined. Table 16 presents the results of determination of tritium concentration in water.

Table 16.

Baitles section. Results of laboratory analyses of groundwater samples

##	Well No.	^{137}Cs, Bq/kg	^{90}Sr, Bq/kg	$^{239+240}$Pu, Bq/kg	^{3}H, kBq/kg
1	10	<0.02	0.13±0.03	*	1±0.1
2	11	<0.05	0.03±0.01	*	10±1
3	15	0.04±0.02	<0.015	*	0.01±0.001
4	16	<0.06	<0.01	*	0.02±0.002
5	17	<0.03	<0.019	*	10±1
6	20	0.04±0.02	<0.01	*	0.02±0.002
7	21	*	*	*	0.011±0.001
8	25	<0.03	<0.05	*	2.7±0.25
9	28	<0.05	<0.02	*	0.015±0.003

##	Well No.	^{137}Cs, Bq/kg	^{90}Sr, Bq/kg	$^{239+240}$Pu, Bq/kg	^{3}H, kBq/kg
10	28A	<0.03	<0.03	*	50±5
11	30A	<0.02	<0.03	<0.0034	100±10
12	31	<0.03	<0.02	*	0.015±0.003
13	32	*	*	*	0.011±0.001
14	33	<0.03	<0.007	*	1±0.1
15	36	<0.04	<0.006	*	10±1
16	6 (section III-III)	<2	0.19±0.09	*	260±24
17	33P	<0.02	1.0±0.01	*	9.5±0.5
18	T1	<0.01	0.7±0.01	*	80±1
* - no measurements performed					

Study of carry-over of artificial radionuclides with the porous water flow along the Baitles valley beyond the bounds of Degelen massif showed that in most samples ^{137}Cs concentration was below the MPA. Quantitative values for ^{90}Sr were obtained only in wells 10 (0.13 Bq/kg), 14 (0.03 Bq/kg) and 6 (0.19 Bq/kg).

The main element causing radioactive contamination of ground waters is ^{3}H. The obtained data show that ^{3}H concentration in ground waters in the studied area varies from 0.1 to 260 kBq/kg. The highest values were registered in the stream underflow along all its flow from Degelen mountains to the Koskuduk tract. Maximal ^{3}H concentration was registered in well 6 located at the border of Degelen mountain massif.

As the distance from the mountains increases, the ^{3}H concentration in water decreases but, nevertheless, in the place where the riverbed disappears in the Koskuduk valley the ^{3}H concentration in the water sample, in subsoil water from well 17, is as high as 10 kBq/ kg (8 km away from well 30A). The available information is insufficient to determine ^{3}H distribution in subsoil waters in the territory of Koskuduk tract. For example, in some wells (16, 15, 20) drilled at a distance of 1 km from each other ^{3}H concentration was as low as 0.01–0.02 kBq/kg. The two wells drilled at the Kii foothill turned out to be waterless. At the same time, in point 28 located in the center of the unstudied area the ^{3}H concentration in the plant free water was as high as 1.5 kBq/kg, which is an indirect proof of the fact that subsoil waters in that area have increased ^{3}H concentration. Of special attention is presence of rather high ^{3}H concentrations in subsoil waters spreading within northeastern boundary of the Koskuduk tract (well 36, ^{3}H – 10 kBq/kg). Sharp increase in ^{3}H concentration may be caused by different factors. Firstly, in such a case we can have different flows of ground waters inflowing into the tract from Degelen mountains. The character of ^{3}H distribution in the waters of the regional basin is still not studied. In this area the subsoil waters may be connected with ground waters from the zone of exogenous fracturing with ^{3}H concentration up to 10 kBq/kg.

According to the data of well 33P, concentration of artificial radionuclides ^{137}Cs, ^{90}Sr and ^{3}H in fracture waters in the considered area is equal to <0.01, 1.0 and 9500 Bq/l, respectively. Practically the same values were obtained for porous waters in the nearest well 11, which indicates possible interrelation between porous and fracture waters in the section. The presence of ^{90}Sr in fracture waters calls for a more detailed studying of specific features of this radionuclide distribution [7, 8].

2.3. The Karabulak stream area

2.3.1. Hydrogeological characteristics of the Karabulak stream valley

The Karabulak valley stretches in the northeast direction. The riverbed is traced to a distance of 40 km from Degelen mountains. To study characteristics of porous water flows flowing beyond the bounds of Degelen mountain massif the hydrogeological section lines II-II (Figure 24 and Fogure 26) and V-V (Figure 25 and Figure 26) were made.

The section line II-II is located 4 km to the north from tunnel 511. The section line is directed 300° to the northeast. Two wells, 1 and 2, which reached subsoil waters, and well 3, which did not reach subsoil waters, were drilled in the main riverbed. The section line is located at a distance of 4 km from the mountains, therefore the section is mainly presented by fine-sized crushed alluvial sediments consisting of loam sands and gravel with sand filler. The subsoil waters lie at a depth of 0.5-0.8 m.

The section line V-V is located in the valley of the right confluent of the Karabulak stream, at a distance of 2.5 km to the northeast from tunnel 504 (Figure 25 and Figure 26).

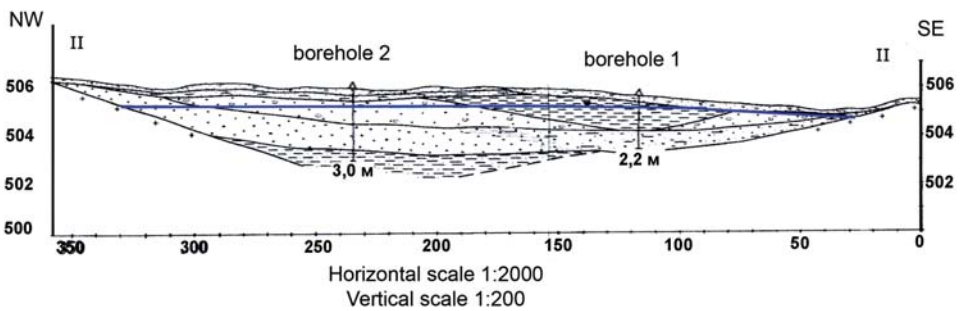
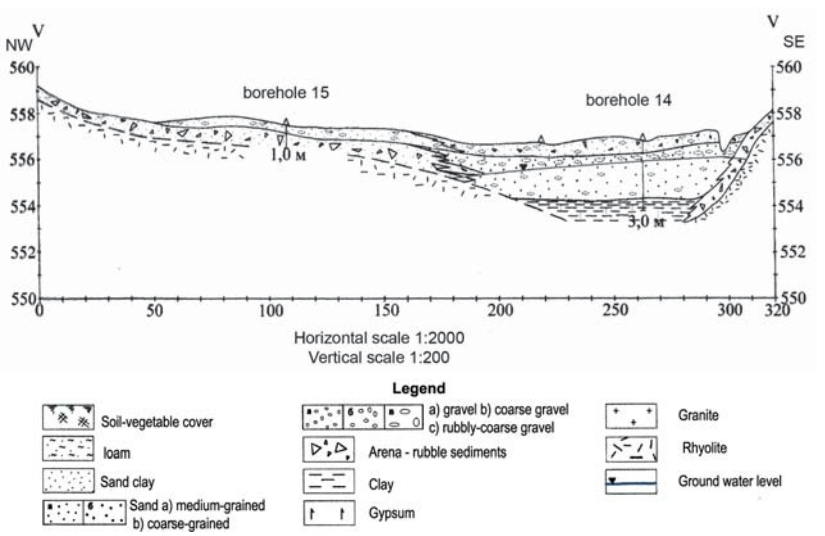


Figure 24. Karabulak stream basin. Geological section along the section line II-II



Figue 25. Karabulak stream basin. Geological section along the section line V-V

The main data on the wells are presented in table 17.

Table 17.

Information on hydrogeological wells

Section, river name	Well No.	Well depth, m	Ground water level, m
II-II, Karabulak riv.	1	2.2	0.5
	2	3.0	0.8
	3	2.9	No water
V-V, right tributary of Karabulak riv.	14	3.0	0.9
	15	1.0	No water

The necessity of making the second profile in the Karabulak river system is explained by the fact that the section line II-II located in the main valley enables to control only northwestern part of the mountains. The head of the right confluent is located in the vicinity of the most radioactively contaminated tunnels.

To study the state of porous waters beyond the bounds of Degelen mountains, in the Karabulak valley, 17 auger wells (1K, 2K, 3K, 4K, 5K, 6K, 7K, 8K, 9K, 10K, 11K, 12K, 13K, 14K, 15K, 16K, 17K) were drilled, and to study fracture waters two exploratory wells (31P and 32P) were drilled (Figure 26).

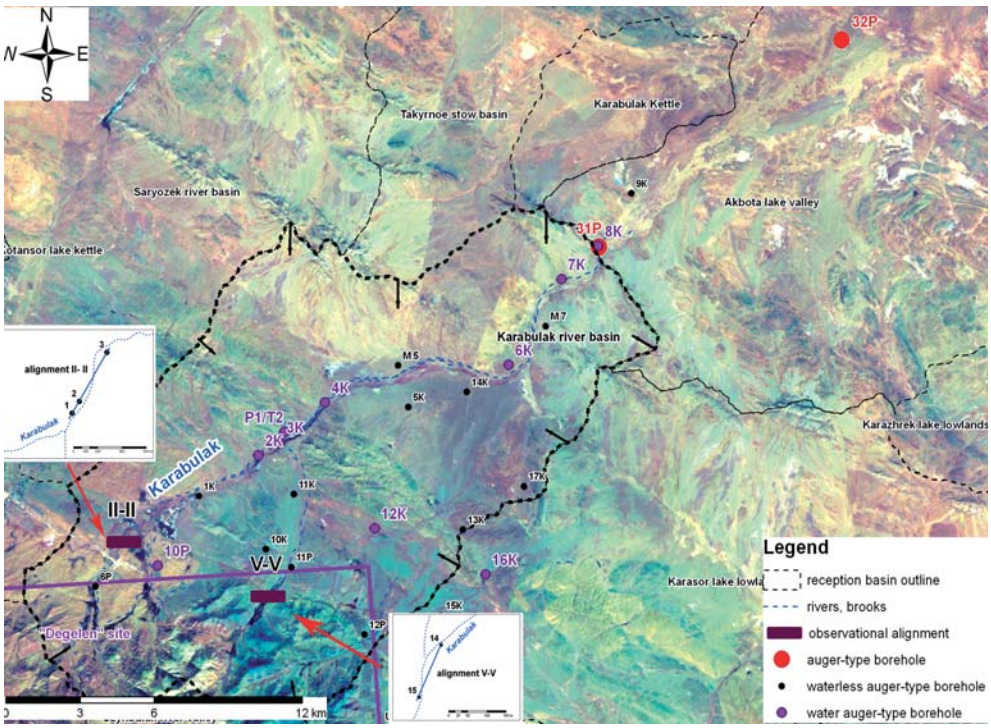


Figure 26. Degelen mountain massif. Karabulak microbasin area.

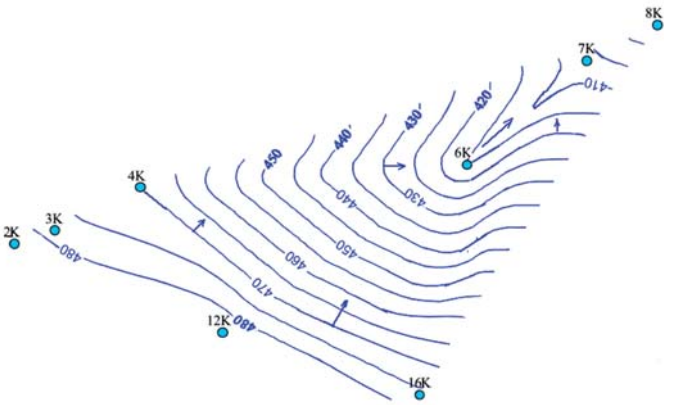
Information about drilled wells is presented in table 18.

Table 18.

Degelen mountain massif. General information on wells.

##	Well No.	Coordinates		Well depth, m	Ground water level, m
		L.N.	L.E.		
1	1K	49º 53' 05,1"	78º 04' 56,9"	2.5	no water
2	2K	49º 53' 58,4"	78º 07' 05,9"	1.5	0.7
3	3K	49º 54' 07,3"	78º 07' 45,4"	6.0	2.6
4	4K	49º 54' 28,9"	78º 09' 38,3"	3.0	2.1
5	5K	49º 54' 44,4"	78º 12' 09,3"	1.0	no water
6	6K	49º 55' 30,4"	78º 15' 36,1"	6.5	5.5
7	7K	49º 57' 13,9"	78º 17' 30,8"	3.0	1.5
8	8K	49º 57' 36,8"	78º 18' 49,8"	2.0	1.3
9	9K	49º 58' 56,9"	78º 20' 00,0"	3.5	no water
10	10K	49º 51' 55,3"	78º 07' 08,7"	0.5	no water
11	11K	49º 53' 02,1"	78º 08' 09,9"	1.0	no water
12	12K	49º 52' 23,1"	78º 10' 58,8"	6.0	2.2
13	13K	49º 52' 08,6"	78º 13' 50,1"	1.0	no water
14	14K	49º 54' 59,9"	78º 14' 09,4"	15.0	no water
15	15K	49º 50' 23,0"	78º 12' 58,2"	0.5	no water
16	16K	49º 51' 06,6"	78º 14' 26,5"	No data	0.5
17	17K	49º 52' 59,3"	78º 15' 57,2"	8.0	no water
18	31P	49°57' 50,79"	78°18' 52,59"	40.0	2.0
19	32P	50°01' 53,78"	78º 27' 24,06"	42.0	12.1
20	10П	49º 51' 46,6"	78º 3' 27,4"	0.2	3
21	P1/T2	49º 54' 20,2"	78º 07' 54,9"	4.0	3.2

The results of explorations were used to make the map of isohypses of porous waters aquifer in the Karabulak stream valley, which is shown in the figure (Figure 27).



The arrow denotes the direction of groundwater flow.

Figure 27. Isohypses of porous waters aquifer in the Karabulak stream valley

The map shows that the ground waters filter in northeastern direction. To determine characteristics of water-bearing horizon in the Quaternary sediments, the exploratory-filtration works, consisting in well filling with water, were carried out in 4 water wells. Wells were filled at constant discharge (Q = const). Water characteristics were used to calculate the filtration coefficient (*K*) and specific water consumption (*q*). The results of exploratory-filtration works are given in table 19.

Table 19.

"Degelen" test ground . Filtration parameters calculated by the results of well fillings

##	Well No.	K, m/day	Q, m^3/h	q, m^3/h
1	3K	0.08	0.09	0.01
2	4K	0.28	0.10	0.06
	7K	0.25	0.06	0.06
	8K	0.25	0.05	0.06
	12K	0.06	0.07	0.01

According to filtration characteristics the water-bearing rocks can be classified as low permeable, and in well 12K as very low permeable (*K>0.1 m/day*). The results of hydro-geological explorations of ground waters spread in the Karabulak valley enable to make the following conclusions:

- two types of ground waters, porous and fracture, are spread in the territory of the Karabulak valley;
- the porous waters are located in fine-crushed alluvial sediments presented by loamy sand and gravel with sand filler, the fracture waters are contained in alevrolytes;
- the groundwater level varies from 0.5 to 0.9 m. The fracture water level is at a depth of 4.5 m and decreases to 12.0 m beyond the mountain terrain.
- the ground waters in the Karabulak microbasin filter in the northeast direction. According to filtration characteristics the water-bearing rocks can be classified as low-permeable and in wells 12K as very low-permeable.

2.3.2. Radionuclide contamination of ground waters in the Karabulak stream valley

The results of laboratory analyses of groundwater samples are presented in table 20.

Table 20.

Results of laboratory analyses of groundwater samples

##	Well No.	^{137}Cs, Bq/kg	^{90}Sr, Bq/kg	^{3}H, kBq/kg
1	2 (section II-II)	<2	0.8	16±6
2	14 (section V-V)	<2	9.0±0.01	110±5
3	7K	<0.02	<0.01	1.2±0.12
4	3K	<0.02	<0.01	30±3
5	16K	<0.02	<0.01	<0.007

##	Well No.	^{137}Cs, Bq/kg	^{90}Sr, Bq/kg	^{3}H, kBq/kg
6	2K	<0.01	<0.01	2.1±0.1
7	4K	<0.01	<0.01	0.7±0.05
8	12K	<0.01	<0.01	0.015±0.005
9	8K	<0.02	2.4±0.02	21±2
10	6K	<0.01	7.6±0.01	30±3
11	31P	<0.01	<0.01	<0.012
12	32P	<0.02	<0.01	0.06±0.007
13	10П	<0.01	0.6±0.01	95±1
14	P1/T2	<0.01	<0.01	0.9±0.05

Table 20 shows that ^{137}Cs concentration in the ground waters in the vicinity of observation points II and V does not exceed MPA. An excess up to 9.0 Bq/l is registered for ^{90}Sr in the right confluent of the Karabulak stream, which intakes waters from the most contaminated tunnels. The most widely spread radionuclide is ^{3}H. Its concentrations in both valleys are above permissible levels for drinking water and are as high as 16 kBq/kg in northern valley and go up to 110 kBq/kg in the valley of the Karabulak right confluent.

To explore spreading of artificial radionuclides with ground waters, we studied the area adjacent to the main Karabulak at a distance of 36 km from northern boundary of Degelen mountains. Of 17 wells only 8 were filled with water. Table 21 presents the results of laboratory analyses made to determine concentrations of artificial radionuclides in groundwater samples. The table data show that ^{137}Cs specific activity in ground waters in all studied area did not exceed MPA, equal to 0.02 Bq/l. Quantitative concentrations of ^{90}Sr were determined only in wells 8K, 6K and 10P. No reasonable explanation can be suggested to describe the ^{90}Sr distribution. To explain ^{90}Sr behaviour it is necessary to carry out explorations with detailed study of migration routes in the area.

The main element causing radioactive contamination is ^{3}H. On the background of continuous decrease in ^{3}H concentrations in subsoil waters (from 95 to 21 kBq/l), there are some points registering considerable variations in ^{3}H concentration. Most probably, these deviations can be explained by gradients and configurations in the contamination front and by vein type of radionuclide migration routes in the area. It is quite likely that some wells penetrated into the head part of the flow with high concentrations, whereas others penetrated into peripheral parts with low concentrations.

Two hydrogeological wells for groundwater monitoring are made in this direction. Well 31P is drilled at a distance of about 50 m from the auger well 8K. ^{137}Cs, ^{90}Sr and ^{3}H concentrations in the water in well 31P do not exceed MPA, which indicates the absence of connection between subsoil waters with the groundwater horizon located in the zone of exogenous fracturing. The well was made to monitor the groundwater contamination in the vicinity of the Karabulak stream. Well 32P was drilled 14 km away from the auger well 8K, downstream the Karabulak riverbed. In the well water ^{3}H concentration is 60 Bq/l, whereas ^{137}Cs and ^{90}Sr concentrations do not exceed MPA. The well was drilled to control possible inflow of contaminated waters in the zone of influence of the Kalba-Chigiz fault [7, 8].

2.4. Altybay stream

2.4.1. Hydrogeological characteristics of the Altybay valley

The valley of Altybay microbasin is located in northwestern part of the Degelen mountain massif.

To study the groundwater flows of western direction from Degelen mountains, the section line IV-IV was made (Figure 28). It is located in the valley of the Sary-Uzen right confluent, 3.5 km to the north-west from tunnel No.503. The section line is directed to the northwest, at the angle of 70°. The length of the section line is 480 m. The valley has a hardly-defined riverbed in the central part. Three wells 7, 8 and 9 were drilled in the central side parts of the valley (Figure 28).

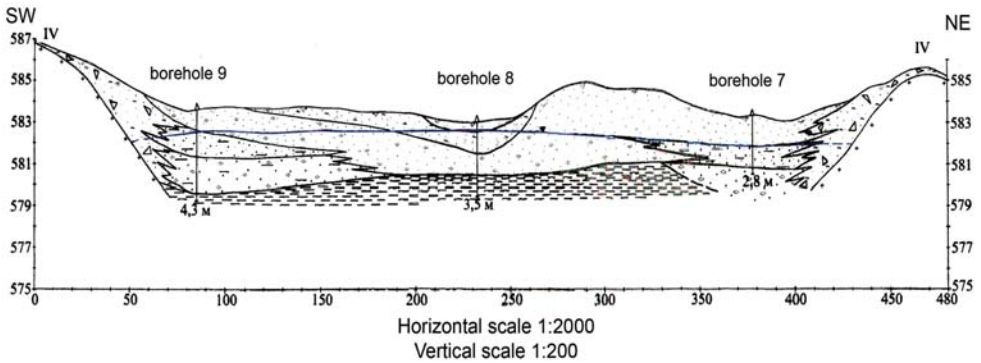


Figure 28. "Degelen" test ground. Geological section along the section line IV-IV

The main data on the wells are presented in table 21.

Table 21.

Information on hydrogeological wells

Section No., river name	Well No.	Well depth, m	Ground water flow, m
IV, Sary-Uzen riv.	7	2.8	no water
	8	3.5	1.5
	9	4.3	no water

The section contains proluvial and alluvial sediments presented by gravel, sands and loamy sands. The thickness of sediments is 2.5 – 4.0 m. The ground waters were detected at a depth varying from 0.3 to 1.5 m. The underlying level is formed by Neocene waterproof clays.

To study the state of porous waters beyond the bounds of Degelen mountains, in the direction of Sary-Uzen test ground, the area adjacent to Altybay microbasin was studied to a distance of 15 km from Degelen mountains. To obtain information on ground waters 7 auger wells (7A, 8A, 9A, 10A, 11A, 12A, 13A) and one exploratory well (35P) were drilled (Figure 29).

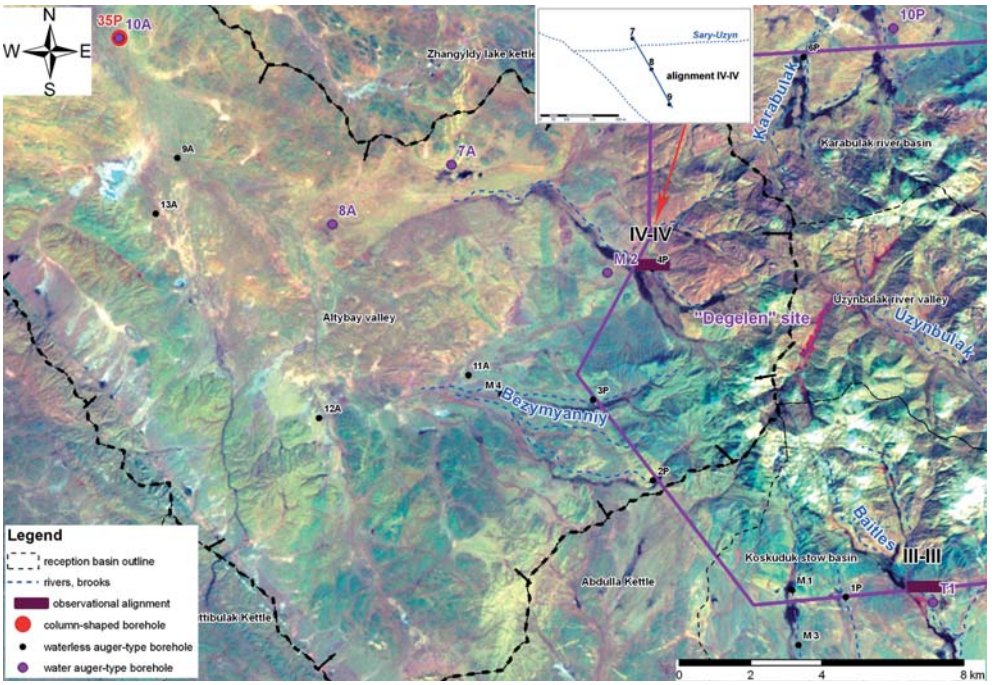


Figure 29. "Degelen" test ground . Altybay microbasin

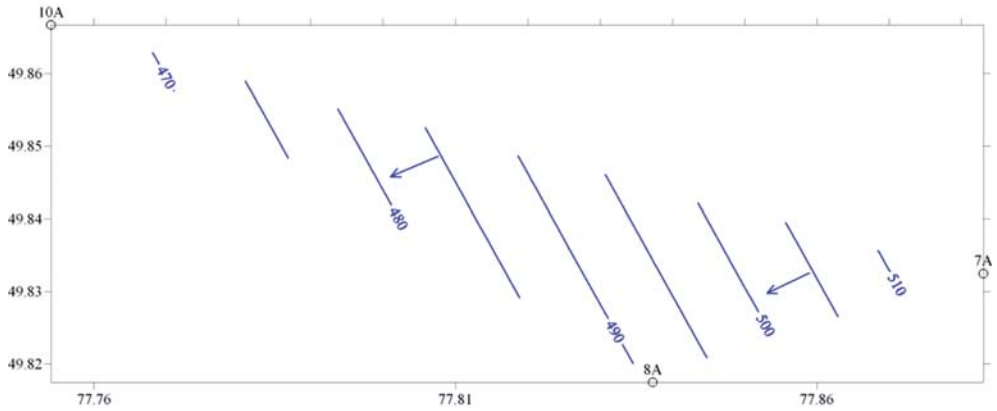
The data on drilled wells are presented in table 22.

Table 22.

"Degelen" test ground. General information on wells

##	Well No.	Coordinates		Well depth, m	Ground water level, m
		L.N.	L. E.		
1	7A	49º 49' 55,8"	77º 53' 01,7"	2.5	2.0
2	8A	49º 49' 01,8"	77º 50' 17,0"	4.0	2.4
3	9A	49º 50' 08,0"	77º 46' 33,5"	15.0	no water
4	10A	49º 51' 59,0"	77º 45' 17,3"	10	5.6
5	11A	49º 46' 40,5"	77º 53' 10,6"	15.0	no water
6	12A	49º 46' 06,7"	77º 49' 39,0"	15.0	no water
7	13A	49º 49' 18,1"	77º 45' 59,9"	15.0	no water
8	35P	49º 51' 57,97"	77º 45' 21,0"	38.0	3.3
9	M2	49º 48' 8,8"	77º 56' 31,5"	3.0	2.2

The results of explorations were used to make the map of isohypses of porous waters aquifer in the Karabulak valley, which is shown in the figure (Figure 30).



The arrow denotes the direction of groundwater flow.

Figure 30. Map of water-bearing horizon of porous waters in the Altybay valley

The map analysis enables to make a conclusion that ground waters filter in northwestern direction. To determine characteristics of water-bearing horizon in the Quaternary sediments, exploratory-filtration works, consisting in well filling with water, were carried out in 3 wells. The wells were filled at constant discharge (Q = const). The results of experiments were used to calculate the filtration coefficient (*K*) and specific water consumption (*q*). The results of exploratory-filtration works are given in table 23.

Table 23.

"Degelen" test ground . Filtration parameters calculated by the results of well fillings

##	Well No.	K, m/day	Q, m^3/h	q, m^3/h
1	7A	0.27	0.07	0.09
2	8A	0.27	0.10	0.05
3	10A	0.16	0.17	0.07

According to filtration characteristics the water-bearing rocks in the valley area can be classified as hardly permeable.

The results of hydrogeological explorations of ground waters spread in the Altybay valley enable to make the following conclusions:

- two types of ground waters, porous and fracture, are spread in the territory of the Altybay valley;
- The porous waters are located in the diluvial-proluvial sediments presented by gravel, sand and loamy sands. The subsoil waters were drilled into at a depth varying from 0.3 to 1.5m. Below this layer the Neocene waterproof clays are located.
- The depth of subsoil waters ranges from 0.3 to 1.5 m. The level of fracture waters beyond the mountain terrain in well 35P is at a depth of 3.3 m;
- the ground waters in the Altybay area filter in the northwest direction. According to filtration characteristics the water-bearing rocks can be classified as low-permeable.

2.4.2. Radionuclide contamination of ground waters in the Altybay valley

Table 24 presents results of laboratory analyses of groundwater samples.

Table 24.

"Degelen" test ground. Radionuclide concentration in porous waters

##	Well No.	^{137}Cs, Bq/l	^{90}Sr, Bq/l	$^{239+240}$Pu, Bq/l	^{3}H, kBq/l
1	8 (section IV-IV)	3.0	0.2	*	170±17
2	10A	0.04±0.01	<0.01	*	17±1.7
3	8A	<0.03	<0.01	*	0.07±0.01
4	7A	<0.01	<0.01	0.02	19±1
5	35P	<0.01	<0.01	*	14±1.4
6	M2	<0.01	<0.01	*	41±1
* - no measurements performed					

Analysis of the table data shows that in the place where the observation point is located the ^{137}Cs and ^{90}Sr concentrations in ground waters are 3.0 and 0.2 Bq/l, respectively, which does not exceed the permissible values for drinking water.

Of 7 auger wells drilled to study the distribution of artificial radionuclides in the chosen direction only 4 wells had water. Table 25 presents the results of laboratory analyses. The available data show that concentrations of artificial radionuclides ^{137}Cs and ^{90}Sr do not exceed MPA in the studied area. In well 10 the ^{137}Cs concentration was 0.04 Bq/l and in well 7A concentration of $^{239+240}$Pu was 0.02 Bq/l. These concentrations are not hazardous and much lower than permissible values for drinking water. The ^{3}H concentration in subsoil waters within the main riverbed varies from 41 to 17 kBq/l.

To study fracture waters in the Altybay area one hydrogeological well 35P was drilled near the auger well 10A. The ^{3}H concentration in the water from well 35P is 14 kBq/l. This value is within the values of ^{3}H concentration in porous waters (well 10A), which indicates close interrelation between porous and fracture waters in this area.

To study the character of ^{3}H contamination of ground waters in this direction it is necessary to carry out additional investigations. The data from well 8A enable to make a preliminary conclusion that radioactive contamination of ground waters in this direction is very low. High tritium concentrations in wells 10A and 35P may be explained by the fact that the above wells were drilled on the boundary of Sary-Uzen test ground. Therefore it is possible to conclude that the data from wells 10A and 35P mainly characterize not flows of ground waters from Degelen mountain but give the ^{3}H concentration in ground waters spread on Sary-Uzen test ground [7, 8].

2.5. Radionuclide contamination of springs

2.5.1. Output of springs

Before nuclear tests there were more than 30 springs in Degelen mountains at different height marks. According to results of surveys in 2005, there were discovered 17 active springs (Figure 31).

From the start of nuclear tests the mountain massif underwent the destructive damages. Hydrostatic pressure is reduced due to expansion of fractures and it results in lowering of underground water mound roof. Water-bearing tectonic structures are renewed, and linear circulation of underground waters is increased there. Redistribution of linear streams and reorientation of stream courses of underground waters takes place. Springs located at high hypsometrical levels are disappearing. Absolutely new springs are arising. Some springs become pulsative. Judging by lush vegetation, there are seasonal springs in several places.

General features of springs output can be presented according to results of observations carried out in 2002. In 2002 two springs and artesian well 385 were monitored (Figure 32). Water streams were relatively stable in first six months. In this period of time the intensive stream run from spring 251/252. Most stable output was from spring 3089 and well 385. Water regime has stabilized in May and total stream has reduced to values observed in winter. However streams have intensified in June due to increase of atmospheric fallouts. Peak of atmospheric fallouts in 2002 fell on the end of spring and first two summer months. Amount of precipitations in May-June considerably exceeds the values of long-term observations and this directly effected on increase of streams output. Increase of output has continued in July. Output of spring 251/252 was 1528 l/min. This value is almost 4 times greater than output value in June. Output of spring 3089 and well 385 has increased 2 times as much.

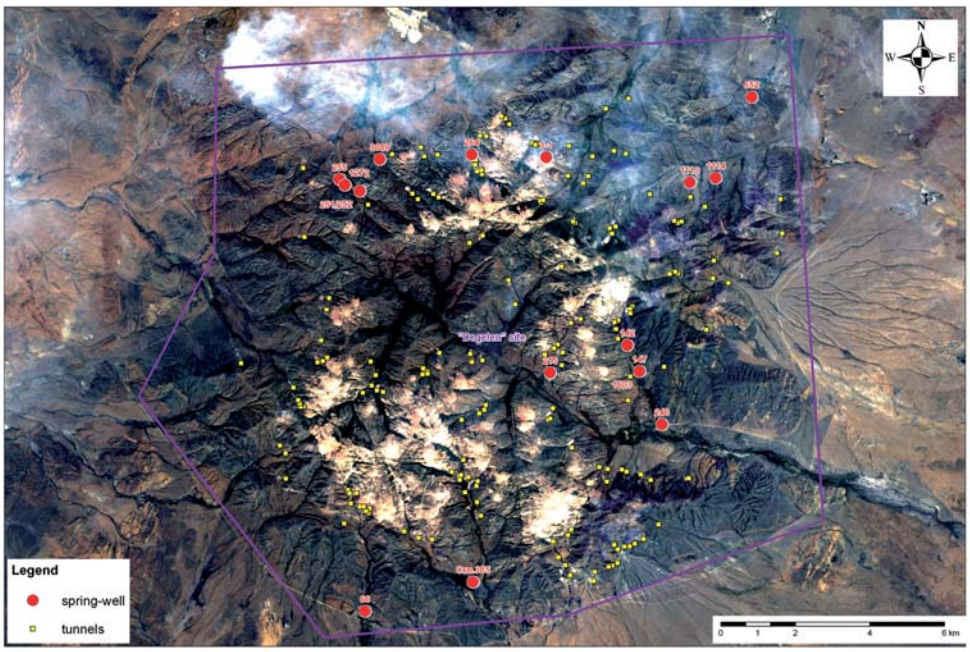


Figure 31. Location of springs on "Degelen" site

In Figure 32 the mistiming of peaks of maximum amount of precipitations and maximum total output of water running from springs and well 385 can be clearly traced.

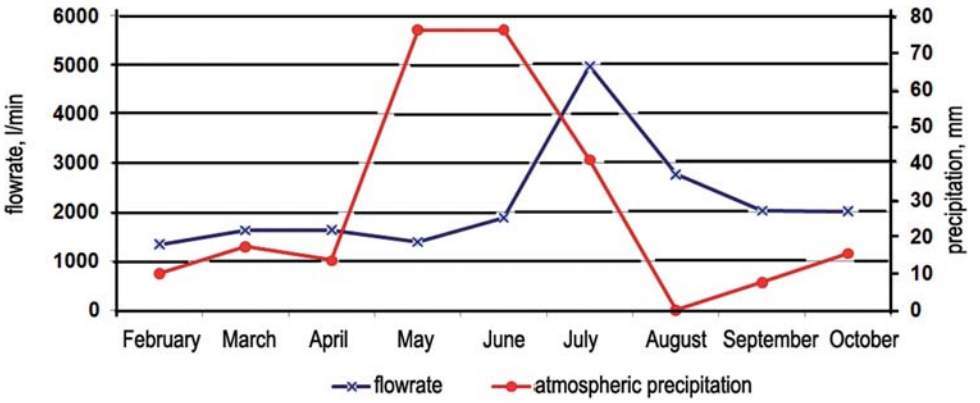


Figure 32. "Degelen" site. Total inflow and atmospheric fallouts on "Degelen" site

The figure shows that maximum total output of streams is almost 2 months away from the start of intensive atmospheric fallouts. This period can be taken for the time of precipitation infiltration through fracture systems in Degelen mountains. Output of all water-developments is reducing in August, and it is normalized in September and October reaching the usual level.

As for other springs, only fragmentary data are available. According to results of inspection in 2005, general information about these springs is presented in Table 25.

Table 25.

Springs inspected at "Degelen" site

##	Sampling point	^{90}Sr, Bq/l	^{3}H, kBq/l	^{137}Cs, Bq/l	Flowrate, l/min
1	Spring 3089	8	74	1	36
2	Spring 652	<0.1	0.4	0.49	69
3	Spring 147	56	15	<0.36	*
4	Spring 3-1	60	24	<0.36	300
5	Spring 264	125	200	1.3	270
6	Spring 255	<0.1	64	<0.36	174
7	Spring 66	<0.1	142	<0.36	450
8	Spring 142	12	153	<0.36	*
9	Spring 215	3	33	<0.36	*
10	Spring 1114	<0.1	1.3	<0.36	*
11	Spring 1113	13	60	<0.36	300
12	Spring 246	1	165	1.25	2680
13	Spring 1603	40	19	<0.36	1040
14	Spring 303	58	*	1	*
15	Spring 251/252	<0.1	65	<0.36	120
16	Spring 1272	7	*	<0.36	*
17	Well 385	1.4	180	0.4	70
*- no measurements performed					

Table data demonstrate that output of most springs does not exceed 1000 l/min and does change from 36 to 1040 l/min. Maximum output values reaching 2680 l/min were registered for spring 1603 [7, 13].

2.5.2. Pattern of radionuclide contamination of spring water streams

Results of laboratory analyses of spring water samples taken in 2005 are presented in Table 26. According to data presented it can be noticed that concentration of the artificial radionuclides (^{90}Sr, ^{137}Cs, ^{3}H) only in springs 652 and 1114 does not exceed the permissible values for drinking water and is equal to 5, 11 and 7700 Bq/kg respectively. At the same time, concentration of ^{137}Cs in water of most springs is lower than MPA. Concentration of ^{90}Sr in water of springs varies from values below MPA to 125 Bq/kg. Presence of high concentrations of ^{90}Sr in water of springs can not be explained for the present and requires more detailed study. This phenomena is both of purely scientific importance to study the pattern of migration of the given radionuclide with water streams and, at the same time, represents a problematic issue of development of radioecological situation. This is connected with sorption of ^{90}Sr by bottom sediments and its gradual accumulation in spring areas in considerable amounts representing radiation hazard to people and animals.

Primary radioactive contaminant of spring water is ^{3}H. Concentration of ^{3}H does not exceed the permissible values for drinking water only in springs 652 and 1114. Content of ^{3}H in other springs varies from 15 to 200 kBq/l what is more than 25 times greater than the permissible level. It is necessary to note that the specified sources are characterized by low values of mineralization and hardness and have low temperature and high taste properties at any time of year. Therefore it is necessary to undertake protective activities to exclude the use of this water for drinking purposes.

2.6. Chemical composition of underground waters

Analyses of chemical composition of underground waters have demonstrated that at any intensity, water streams arise from the same geological structures, where it acquires certain mineralization and chemical composition. Since chemical water composition is formed in different fracture systems, some particularities in composition of streams running from different tunnels are observed. For example, intensively sulphate water runs from tunnel No. 504, moderately sulphate water runs from tunnel No. 511, weakly sulphate water runs from tunnel No. 506, and hydrocarbonate water runs from tunnel No. 152. Sulphate waters predominate, and sulphate-hydrocarbonate and hydrocarbonate waters are rarely traced. Calcium usually predominates among cations, and predominance of alkali cations: sodium and potassium is rarely traced. Chemical composition of water running to portals does not depend from attitude position of tunnels.

Dependence of the general mineralization value from chemical composition is clearly observed. Mineralization in sulphate waters is considerably higher (up to 1,0–1,2 g/l) than in hydrocarbonate waters. Highly mineralized fracture waters are probably formed in geological systems containing minerals having easily leachable sulphur. Within Degelen there are such mineralized areas containing considerable amounts of pyrite. Thus, fracture waters formed in local structures are predominately inflow in tunnels, and only in some cases the penetration of water from the regional system can be assumed.

Composition of water of the regional system can be characterized by composition of water in self-effusive well 385 and springs representing the ascending sources of fracture waters. Water of these sources is characterized by extremely low mineralization (0,1–0,2 g/l) and sulphate-hydrocarbonate or hydrocarbonate composition, as well as by rather low hardness of water while removable hardness predominates over permanent one. Springs 3089 and 251 are located in the north-west part of the site, spring 163 is located in the central part, and self-effusive well 385 characterizes the southern flanks of mountains. Thus sources of water coming from the regional system are located in different parts of the area. However chemical composition of water in all sources is rather similar, although there are some nuances. Thus content of hydrocarbonate anion is gradually decreased in water of well 385, it is probably related to the gain of flow from the filtration zone of Degelen mountains.

Water of similar chemical composition runs in tunnel No. 176 and composition of water streams running from tunnels No. 506, 707 and 609 becomes slightly similar. Water of major streams running from tunnels is characterized by sulphate component. At the same time, the higher general mineralization the higher content of sulphate anion.

Thus, mineralization and chemical composition of water running in tunnels are formed in local fracture structures. Under conditions of rock relief and irregular soil-vegetable layer, chemical composition of water completely depends from mineralogical composition of host rocks. When these waters inflow in the regional basin, leveling of chemical composition takes place.

In most cases water of surface streams and underground waters of valleys are of sulphate type and their chemical composition is close to composition of streams running from tunnels. Chemical composition of underground waters in every valley is characterized by some distinctive features. Underground and surface waters in valley of Uzynbulak rivulet are related to the sulphate type with predominance of cation Ca^{2+}. Mineralization of underground water varies within 0.7–1.2 g/l, and waters of surface waterway have mineralization 0.5 g/l. Directly under the bed located near the right side of the valley (well 13), mineralization of underground water makes up 0.9 g/l, and in the central part of the valley – 0.7 g/l. Near the left side of the valley, the well 11 has unsealed stagnant underground waters of highest mineralization (2.87 g/l) and content of sulphate anion up to 87 % mg-eqv.

Underground waters of the valley of Karabulak rivulet (wells 1 and 2) and its right inflow (well 14) are of high mineralization level (1.0–1.28 g/l). These waters are related to the sulphate type. Wells subjected to sampling are located far (2.5 – 4 km) from the northern foot of mountains. In low sections of relief the salinization of soil is observed, and it caused high mineralization of waters.

Most fresh underground waters (0.47 g/l) were observed in the valley of Baitles rivulet (well 6) that springs from the southern part of Degelen mountains. Level of the regional basin of underground waters is located here close to the day surface and the presence of self-effusive well indicates this. Like fracture waters of the regional basin, underground waters are related to hydrocarbonate type. The regional basin probably serves as a main source of underground waters.

The right inflow of Sary-Uzen rivulet springs from the western flanks of Degelen mountains. There is no surface flow in this valley. Underground waters were detected in all three wells located across the valley. Mineralization of water varies in the small range 0.72–0.75 g/l and is related to the sulphate type (wells 7 and 8) and rarely to the hydrocarbonate-sulphate type (well 9). Chemical composition of underground waters of this valley corresponds to chemical composition of water streams running from tunnels.

Considerable differences were detected in microelement composition of tunnel waters presented in Table 26.

Table 26.

Microelement composition of surface waters of some tunnels having water-effects

Sampling point	Fe	Cu	Zn	Mo	Sr	Be	Al	Mn	Ba	Li
	µg/l									
tunnel 165	95	20	62	76	266	<DT	<DT	20	16	48
tunnel 176	36	3	52	207	139	<DT	<DT	<DT	<DT	24
tunnel 609	26	3	<DT	202	145	<DT	<DT	<DT	19	26
tunnel 104	25	<DT	<DT	991	384	10	<DT	16	14	58
tunnel 177	55	<DT	<DT	531	347	<DT	<DT	<DT	16	48
tunnel 511	64	32	1000	<DT	448	61	<DT	3000	27	44
tunnel 504	274	<DT	8000	18	784	445	1500	87000	57	119
tunnel 503	53	11	180	21	598	14	<DT	1200	30	53
tunnel A-1	342	25	1500	42	878	108	1600	12000	51	78
MPC, µg/l	300	1000	5000	240	7000	0.2	500	100	100	3

Considerable concentrations of zinc, molybdenum, manganese were detected in many water samples. Maximum microelement concentration values were detected in water of tunnels A-1 and 504. Very high concentrations of beryllium exceeding the maximum permissible concentration by several digits [15] are to be mentioned particularly. Considerable concentrations of some microelements probably do reflect to a considerable degree the influence of mountain massif geology features than design features of the tested nuclear devices. Analysis of the obtained results on elemental, anion-cation and radionuclide composition of water has not exposed the correlation dependence between them so far [1, 7, 8].

3. FORECAST FOR UNDERGROUND WATER STATE DEVELOPMENT BEYOND DEGELEN MOUNTAIN MASSIF

Analysis of hydrogeological and geodynamic situation of Degelen complex allows us to make the following conclusions about conditions of migration of radionuclides with underground waters.

Hydrogeological conditions of Degelen complex are typical for the territory of central-Kazakhstan hummocky topography. Even if there is no continuous cover of loose low-permeable sediments (favorable conditions of infiltration), small amount of atmospheric fallouts (170–460 mm a year, average annual – 263 mm, in high-water year – 382 mm, in low-water year – 221 mm) forms the limited amount of natural resources of underground waters. Developed everywhere in Paleozoic rocks, fracture underground waters do not form considerable natural reserves (static, capacitive), including conditions of increased man-induced fracturing. Tectonic deformation zones and rock contact zones having different resistance to deformations are characterized by the increased water permeability under natural conditions. Under conditions of powerful man-induced impact such zones became more deformed and water-permeable. As a result, these zones act as drains for fracture waters and thinning of underground streams towards ground surface with formation of rather large

areas with hydrophilous vegetation takes place in the lowered sections of zones. Underground water consumption for plant transpiration and evapouration takes place right here under conditions of dry windy weather.

Currently existing predictive estimate for tunnels having stable water-developments (Nos. 503, 504, 511, 609, 165, 104, 176 and 177) regarding the migration of radioactive products from central underground nuclear explosion (UNE) zones with underground waters looks in the following way.

Upon completion of a phase of gas migration of UNE radionuclides, underground waters become main migration agent and carrier of radionuclides moving them out of central UNE zone. There are main sources of radionuclides. The first one is radioactive aerosol-dust component localized on broken rock and in fractures of the complex. The second one is radioactive glassy-slaggy rock containing major portion of ^{90}Sr, ^{137}Cs and almost all $^{239+240}Pu$. Long-term observations over tunnels of Semipalatinsk test site and other tunnels of UNE objects demonstrate that concentration of radionuclides in water goes up during the first several years after explosion, and then it gradually goes down due to both depletion of aerosol-dust source and decay of radionuclides. In comparison with migration of radionuclides from the first source, migration of radionuclides from the solidified radioactive rock melt is practically minor during the first years and probably decades, since radionuclide leaching runs with coefficient about 10^{-3}–10^{-7} g/cm^2×day, taking into account that specific surface area of the given source is also lower by several digits than the one of the first source (rocks in shatter zone).

The following conditions were accepted for the given model of migration of radionuclides from the mountain massif:

- radionuclides are concentrated in the solidified rock melt and distributed on surface of fractures formed in the fracture zone and on surface of rock fragments of the caving column;
- underground waters and atmospheric fallouts penetrate in the shatter, fracturing zone and directly in the cavity through fracture systems.

At first radionuclides are redistributed in rocks due to its sorption on walls of channels, where water runs. If water channels do not change, surface of channels in due course becomes saturated with radionuclide and quasi-equilibrium state between aqueous and solid phases is settled.

System of equations for the accepted model:

$$\begin{cases} \dfrac{dA_p}{dt} = -\left[\lambda + \dfrac{k \cdot \dfrac{M_h}{V}}{1 + k \cdot \dfrac{M_h}{V}} \cdot \dfrac{v_i}{V}\right] \cdot A_p \\ \dfrac{dA_m}{dt} = -(\lambda + k_m) \cdot A_m \\ \dfrac{dA_c}{dt} = -\left(\lambda + \dfrac{v_o}{V}\right) \cdot A_c + \left[\dfrac{k \cdot \dfrac{M_h}{V}}{1 + k \cdot \dfrac{M_h}{V}} \cdot \dfrac{v_i}{V}\right] \cdot A_p + \dfrac{k_m}{1 + k \cdot \dfrac{M_h}{V}} \cdot A_m \\ \dfrac{dV}{dt} = v_i - v_o \end{cases}$$

, where:

t – time;
A_p – activity of a nuclide sorbed by material of the caving column and shatter zone;
A_m – activity of a nuclide contained in melt;
A_c – activity of a nuclide contained in liquid phase;
λ – radioactive decay constant;
M_p – mass of rock in the caving column;
k – nuclide distribution coefficient (m^3/kg);
k_m –coefficient of nuclide leaching from melt (1/year);
V – volume of liquid phase (m^3);
v_o – cavity water outflow rate (m^3/year);
v_i – cavity water inflow rate (m^3/year).

First equation of the system expresses the change of nuclide activity in the caving column and shatter zone, second one – decrease of melt activity due to nuclide decay and leaching, third one – change of activity of nuclides located in liquid phases of the cavity, forth one – rate of increase of the liquid phase volume in the cavity due to difference between water inflow and outflow.

A model of radionuclide carry-over by tunnel waters was realized using program language Visual Basic 6 then using COM-technologies of ArcObjects it was introduced in geographic information system (GIS) ArcView/ArcInfo in application ArcMap 8.3. A system of differential equations was solved using Runge-Kutt method. Software version of the model includes two modules: calculation of radionuclide carry-over from tunnels and comparison of the obtained results with experimental data.

The following was taken as primary data for conduction of calculations: date of test, explosion power, average annual water flow rate and forecast date. All model parameters are stored in GIS database of NNC RK, can be initialized at startup and changed right before calculations using form "Model parameter editing".

Average outflow rate values beginning from 1996 and available average annual experimental data were used in calculations. Calculations show that concentrations of ^{90}Sr and ^{137}Cs have maximum values at the initial time (physically is corresponds to completion of initial filling of explosion cavity with water) and then they gradually go down. Such behaviour of radionuclides is conditioned by processes of their desorption from the caving column and fracture zone and as a result by depletion of dust-aerosol source. Such phenomenon can be observed experimentally during the first years after explosion, when cavity is filled with water.

The obtained results can be divided into two groups (Figure 33). Simulation results agree well with experimental data for tunnels No. 104, 165, 503 and 609. This can be explained by the fact that practically all volume of water running out of tunnel has washed the shatter column and fracture zone. As for other tunnels (No. 176, 177, 511), results of calculations differ from experimental data.

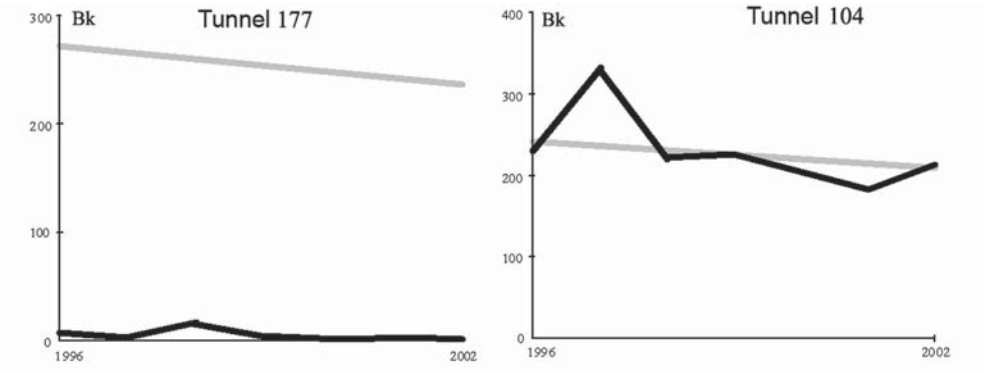


Figure 33. Comparison of experimental (black line) and calculation data

Disagreement of simulation results and experimental data can be caused by insufficiently studied mechanism of formation of water circulation system being formed by fracture waters and atmospheric fallouts. Formation of this system by now is not completed, therefore disappearance (or appearance) of water streams and change of their intensity is possible.

Thus, preliminary predictive estimates made on basis of up-to-date knowledge and data show that in case of no abrupt changes, concentrations of ^{90}Sr and ^{137}Cs will slowly decrease in Degelen underground waters running out of central UNE zones, and concentrations of $^{239+240}Pu$ will feebly increase during 100 years [1, 2].

Thus, migration of the artificial radionuclides with surface and underground waters will result in gradual redistribution of radioactive products from the central UNE zones to peripheral part of the mountain massif. At the same time, migration of radionuclides for long distances beyond Degelen complex with fracture-vein waters through deformation zones is probable. Thus, at present a process of radionuclide carry-over by streams of underground waters beyond Degelen mountain massif becomes one of key factors of formation of radioecological situation both within Degelen mountain massif and beyond its boundaries. At the same time it is clear that once the character of ^{3}H migration with underground waters beyond Degelen mountain massif is learned, possible main ways of migration of such radionuclides as ^{137}Cs, ^{90}Sr and $^{239+240}Pu$ can be estimated [2, 3].

CONCLUSION

The following main water systems present within Degelen mountain massif:

- fracture waters of the regional basin;
- fracture waters of atmospheric precipitation filtration zone;
- pore waters of alluvial deposits;
- surface flow waters.

Fracture waters of the regional basin are widespread in subsoil and represent a main source of water in the region. Level of these waters within Degelen is higher than in ambient area. Therefore the regional basin is predominately recharged here.

Atmospheric precipitation filtration zone is confined to central mountain part of Degelen and takes a position between the regional water basin level and surface. This zone

includes tunnel complex from which considerable amount of water recharging the regional basin has run out on surface. Underground waters of alluvial deposits are widespread in valleys originating from Degelen mountains. Their sources are atmospheric fallouts and fracture waters.

UNEs have resulted in essential deformation of rock massif with formation of multiple shatter zones, craters and gaping fractures. As a result, rock permeability has increased considerably and it caused the increase of descending filtration and partial diversion of surface waterway into underground one.

As a result of UNEs, essential redistribution of linear streams and reorientation of underground water running directions took place. Some springs have disappeared and absolutely new springs have appeared, as well as springs of seasonal activity. As a result of tunnel closure, outflow of water on the day surface from the contaminated tunnels has reduced, and this fact makes a contribution in improvement of environmental conditions of the site. At present stable water streams are observed in portals of 8 tunnels. Main surface waterways (Uzynbulak, Karabulak and Baitles) are seasonal.

Surveys have demonstrated that underground waters are filtered in Uzynbulak area to the south-east, in Karabulak area to the north-east and in Altybay area to the south west directions. According to results of research-filtration works in water-bearing horizon of quaternary deposits, water-bearing rocks can be classified as low-permeable and very low-permeable. Underground water level in intermountain valleys is located at depths from 0 to 6 m. Beyond mountain terrain the level goes down to 12 m.

Mineralization and chemical composition of water running from tunnels are characterized by high constancy in every individual stream and indicate that mineralization is formed in local geological structures in the course of infiltration. Fracture waters of the regional basin have low mineralization and hydrocarbonate-sulphate composition, and this fact distinguishes them from water of streams running from majority of tunnels.

Underground and surface water radioactive contamination process at "Degelen" site continues and, according to radionuclide monitoring data, it has relatively stable character. Propagation of the artificial radionuclides by streams of underground waters beyond Degelen mountain massif has ambiguous character. In most observed cases the migration paths have the vein (cobweb) appearance, contamination of underground waters in local areas occurs rarely. At the same time, main streams of the contaminated underground waters are confined to beds of temporary and permanent surface waterways.

Main radioactive contaminant of aqueous medium both at "Degelen" site and beyond its borders is ^{3}H. Levels of concentration of the artificial radionuclides (except ^{3}H) beyond the borders of Degelen mountain massif do not represent radiation hazard and do not exceed the permissible values for drinking water. Pore waters are contaminated to a greater degree, especially by tritium. Level of contamination of fracture waters by tritium substantially depends from geology and hydrogeological conditions in areas of their interaction with pore waters.

It is necessary to draw a special attention on high concentrations of ^{90}Sr in water of springs. In this case we deal with "integral" water streams, but not with streams confined to any specific tunnels. At the same time, the level of specific activity of ^{90}Sr is commensurable with levels that we detect in water-developments of tunnel portals, where rather high concentrations of ^{137}Cs occur, and water of springs does not contain ^{137}Cs. Besides springs, high migration features of ^{90}Sr were observed in series of wells drilled both in pore and fracture wa-

ters. From the radiation safety point of view, the observed levels do not represent hazard, but from a position of ^{90}Sr migration predictive estimates these levels are very high. The above mentioned features give grounds to assume that ^{90}Sr migrates with fracture-vein waters. This type of underground waters is confined to deformation zones and has higher travel speed than fracture waters of ambient rocks. Taking into account the fact that sorptive capacity of hard rocks is considerably lower than the one of loose rocks, it is expected that this radionuclide will appear far from borders of Degelen Mountains.

Predictive estimates have showed that in case of no abrupt changes, concentrations of ^{90}Sr and ^{137}Cs will slowly decrease in Degelen underground waters running out of central UNE zones, and concentrations of $^{239+240}Pu$ will feebly increase.

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ДЕГЕЛЕҢ ТАУЛЫ МАССИВІНЕН ТЫС ЖЕРЛЕРГЕ ЖАСАНДЫ РАДИОНУКЛИДТЕРДІҢ ЖЕРАСТЫ ЖЫЛЫСТАУЫ

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Бұл мақалада "Дегелең" алаңындағы ядролық сынақтар өткізілген жерлерден жерасты суларының ағысымен техногенді радионуклидтердің шығу сипатын зерттеу нәтижелері келтірілген. Негізгі ағын суларда радиоэкологиялық зерттеулер жүргізілді және жерасты суларының жай-күйін ұзақ мерзімді мониторинг жүргізу үшін бақылау ұңғымалары жабдықталды. Дегелең таулы массивінің шегінде, сонымен бірге оның шекарасынан тыс жерлердегі жерасты суларының радиоактивті ластануының деңгейі жайлы жаңа деректер алынды. Алынған деректердің негізінде жерасты суларының ағысымен техногенді радионуклидтердің жылыстау сипаты туралы қорытынды жасалды, Дегелең таулы массивінің ауданындағы радиоэкологиялық жағдайының даму мүмкіндігінің барысы жайлы болжамдық бағалау жасалды.

Түйінді сөздер: жерасты сулары, ядролық сынақтар, ССП, Ұзынбұлақ, Байтілес, Қарабұлақ, Алтыбай, радионуклидтердің жылыстауы, Дегелң таулы массивінің геологиялық құрылысы және гидрогеологиялық жағдайы.

ПОДЗЕМНАЯ МИГРАЦИЯ ИСКУССТВЕННЫХ РАДИОНУКЛИДОВ ЗА ПРЕДЕЛЫ ГОРНОГО МАССИВА ДЕГЕЛЕН

С.Б. Субботин, С.Н. Лукашенко, В.В. Каширский, Ю.Ю. Яковенко, Л.В. Бахтин*

В статье представлены результаты исследования характера выноса техногенных радионуклидов с потоками подземных вод из мест проведения ядерных испытаний на площадке "Дегелен". На основных водных потоках проведены радиоэкологические исследования и оборудованы наблюдательные скважины для долгосрочного мониторинга состояния подземных вод. Получены новые данные об уровнях радиоактивного загрязнения подземных вод как в пределах горного массива Дегелен, так и за его границами. На основе полученных данных сделаны выводы о характере миграции техногенных радионуклидов с потоками подземных вод, проведена прогнозная оценка возможного хода развития радиоэкологической обстановки в районе горного массива Дегелен.

Ключевые слова: подземные воды, ядерное испытание, СИП, Узынбулак, Байтлес, Карабулак, Алтыбай, миграция радионуклидов, геологическое строение и гидрогеологические условия горного массива Дегелен.

УДК 577.4:504.064:539.16

EFFECT OF RESEALING OPERATIONS ON RADIOLOGICAL CONDITIONS OF DEGELEN TUNNEL NEAR-PORTAL AREAS

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Resealing of nine tunnels located in Degelen massif of the Semipalatinsk test site (STS) was undertaken in order to reduce the threat of nuclear proliferation. Radioecological surveys have been carried out to evaluate radiological conditions of the environment after the resealing operations and understand potential negative impacts of such operations on the tunnel mouth areas. The paper presents data describing changes in radiological conditions of the monitored tunnel mouth areas.

Key words: Semipalatinsk test site, Degelen Massif, tunnels, supplementary protection of engineering facilities, radiological survey, radiological conditions, EDR, flux density of α- and β-particles, radionuclides, specific activity, areal contamination.

INTRODUCTION

During 1996-2000, certain actions were taken at all the engineering facilities of the Semipalatinsk nuclear test site. These actions were aimed at abandoning the nuclear test infrastructure, including avoidance of any unauthorized access to the facilities. The 2001-2005 surveys of Degelen test site showed that a number of protective barriers at Degelen engineering facilities had been broken or damaged. Traces of nuclear waste (NW) were registered on some broken metal structures.

To reduce the threat of NW dissipation/proliferation, which could result from unauthorized access, it was decided to reinforce the existing protection barriers. Resealing of 9 tunnels was undertaken from September 2006 to December 2009.

The first stage of the operations was to conduct a preliminary radioecological survey within the proposed work site and in the field camp area. Radiation surveys were done at each tunnel mouth. The results obtained were used to plan works for each individual tunnel in order to avoid excessive exposure of the personnel to radiation.

Final survey was conducted similarly to the preliminary one in order to assess potential changes in the radiation environment, which could be caused by engineering works planned for each tunnel.

1. BACKGROUND INFORMATION

The operations area is Degelen massif, which is a dome-like upheaval having the diameter of 18 km and 300 m difference in elevations.

Geographically, Degelen massif is situated within the area of the former Semipalatinsk nuclear test site in the northeast part the Kazakh upland (hummocky topography).

Degelen facilities layout and photos of the tunnel mouths before the beginning of works are given in Figure 1.

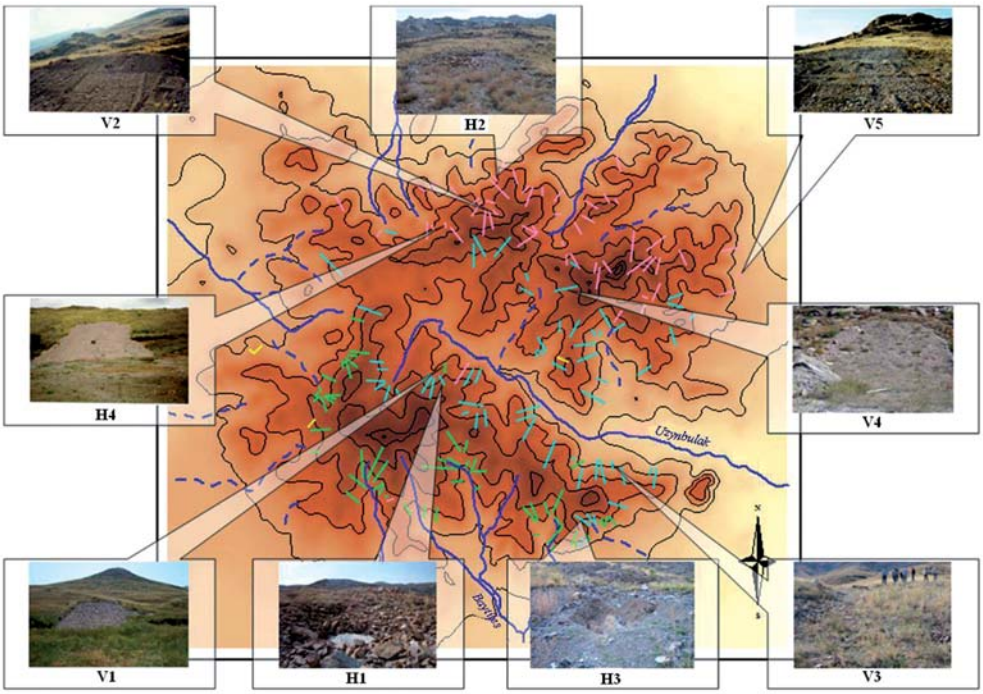


Figure 1. Tunnel layout at Degelen Site

1.1. Methods of building supplementary protection for the engineering facilities (2006–2009)

Supplementary protection of engineering facilities was provided by filling cavity pockets with binding material, which would prevent any unauthorized withdrawal/recovery of nuclear waste from the tunnels. Depending on the design basis, two methods were used for providing supplementary protection:

- Supplementary protection provided through vertical boreholes drilled from the surface – vertical method (Fig. 2);
- Supplementary protection of engineering facilities provided by opening the tunnel mouths and excavating the tunnels – horizontal method (Fig. 2).

Depending on the method chosen for supplementary protection, tunnels were assigned the following identification codes:

- H1, H2, H3, and H4 in case of the horizontal method of supplementary protection; and
- V1, V2, V3, V4, and V5 in case of the vertical method of supplementary protection.

a) b)

Figure 2. Vertical (a) and Horizontal (b) methods of supplementary protection provided for engineering facilities

1.2. Description of the sources of radioactive contamination

Table 1 shows data derived from literature [1, 2] on the types of tests conducted in the surveyed tunnels. The tunnels designated H1, H3, H4, V3, and V4 were designed for serial explosions using several charges. Such explosions were conducted for the following purposes: creating/improving nuclear weapons (CNW), conducting fundamental and applied research (FAR), studying equipment malfunctions and emergency situations (EMES), and studying destructive/damage agents of nuclear weapons (DANW) and their impacts on military and civil facilities.

Table 1.

Data on tests conducted in the H1-H4 and V1-V5 tunnels of Degelen massif

Tunnel code	Test date	Nuclear explosion yield, kt	Description of explosion	Test purpose	Radiological conditions
H1	1988	< 20	A series of 3 nuclear explosions	CNW, FAR, EMES	CCE
H2	1987	< 20		FAR	CCE
H3	1981	< 20	A series of 2 nuclear explosions	CNW	ICE
H4	1974	< 20	A series of 3 nuclear explosions	DANW	CCE
	1980	< 0.001		EMES	ICE
	1981	< 0.001		EMES	CCE
	1981	< 0.001		EMES	CCE
V1	1983	< 20		CNW	CCE
V2	1989	< 20		CNW	CCE
V3	1987	< 20	A series of 3 nuclear explosions	CNW	CCE
V4	1984	< 20	A series of 4 nuclear explosions	DANW	CCE
	1987	< 20		DANW, EMES	ICE

Tunnel code	Test date	Nuclear explosion yield, kt	Description of explosion	Test purpose	Radiological conditions
V5	1976	< 20		CNW	CCE
	1979	< 0.001		EMES	CCE
	1979	< 0.001		EMES	CCE
	1979	< 0.001		EMES	CCE

CNW – creating/improving nuclear weapons
FAR – fundamental and applied research
EMES – research on equipment malfunctions and emergency situations
DANW – studying destructive/damage agents of nuclear weapons and their impacts on military and civil facilities
CCE – complete camouflet explosion, ICE –incomplete camouflet explosion

Residual activity of radionuclides was estimated based on the assumption that all explosions in each individual tunnel yielded in 20 kt (for two explosions of 20-40 kt each). However, it could happen that only one 20-kt charge was blasted, while the two other charges were destroyed and spread around by the first blast. In such cases, fission fragments activity can exceed the values given in the table.

Since none of the tunnel surveys showed ^{235}U and ^{238}U activity higher than natural background, it may be reputed that all nuclear charges were made of ^{239}Pu. In this case, sources of radioactive contamination in the tunnels are ^{239}Pu fission fragments, remaining fissionable materials and activation products.

Fission yields of ^{239}Pu have been thoroughly studied and published elsewhere [3]. Using the published data and original information on blasts, we calculated activity levels with regard to half-life of ^{239}Pu fission fragments, which contribute quite tangibly to the overall activity level (Figure 3).

Quantity of the remaining fissionable material at the destruction point of a nuclear charge structure is a function of the nuclear blast efficiency, which can vary in the range from 1 to 30% depending on the time of a test, type and structure of the nuclear device. Since no information was available on this very explosion, we took the efficiency equal to 20% in order to calculate quantity of the remaining fissionable material.

According to the theoretical calculations for a 1-kt explosion, ^{239}Pu activity is $4.8 \cdot 10^{11}$ Bq/kt at the point of time t = 0 after the explosion. As a next step, knowing typical isotopic composition of weapon-grade Pu, activity of its other isotopes contained in the charge was calculated (Figure 3).

It is well known that many isotopes produced in the process of neutron activation have quite a short half-life. Given that it has been a long time after the explosion, it can be taken that contribution of activation products in the total gamma activity is negligible. Only ^{60}Co, ^{152}Eu, and ^{154}Eu can be an exception, but is not feasible to calculate their activity at this point of time. Therefore, when testing the tunnel samples in a lab, it must be borne in mind that this radionuclide is likely to be present and can influence the overall radiological condition in a tunnel.

Thus, high concentrations of ^{90}Sr, ^{99}Tc, ^{137}Cs, ^{151}Sm, and ^{241}Am, as well as plutonium isotopes can be found in the tunnel areas. Basic artificial radionuclides, such as ^{90}Sr, ^{137}Cs,

and ^{241}Am, can be identified in a laboratory equipped with highly sensitive gamma- and beta-spectrometers. To determine $^{239+240}$Pu, alpha-spectrometry facilities with primary radiochemistry should be used. ^{99}Tc and ^{151}Sm have not been covered yet by the STS survey programs, as there are no standard techniques for estimating their concentrations, although these radionuclides contribute significantly to the overall radiation level. NNC is currently developing standard techniques for determining concentrations of these radionuclides in the environment, which very soon will be available for estimating ^{99}Tc and ^{151}Sm concentrations at STS and Degelen complex, in particular.

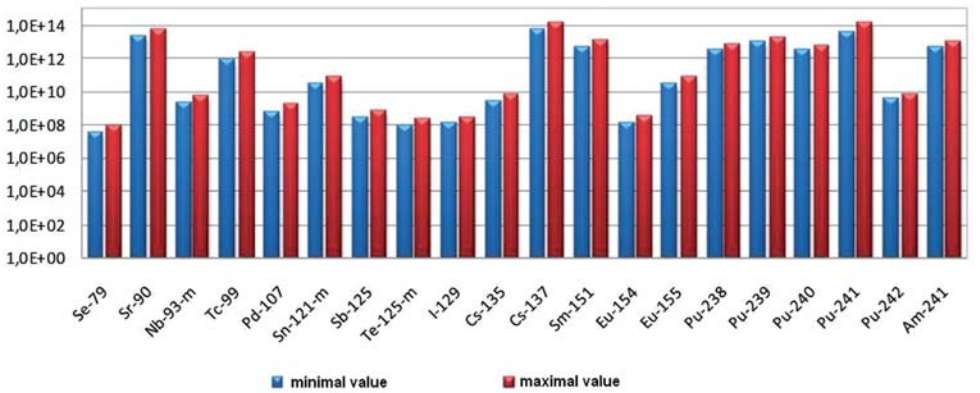


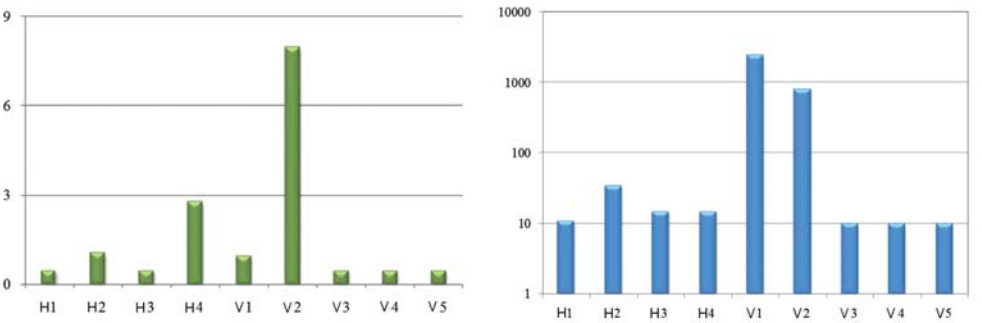
Figure 3. Degelen tunnels H1-H4 and V1-V5: Estimated activity of ^{239}Pu fission fragments and remaining fissile material, *Bq*

Radiological condition of the tunnel areas per 1999 survey findings

Decommissioning activities on these tunnels were taken during the period from 1996 through 1998. Radiometric survey of the tunnel near-portal areas was carried out in 1999.

To survey the areas, a 20m x 20m grid was used to measure EDR and α- and β-particle flux density at each point. Five soil samples were taken from each tunnel.

The survey results are given in the figures below (Figure 4 and Figure 5).



a) b)

Figure 4. Maximum flux densities of α-particles (a) and β-particles (b) obtained during 1999 survey, particles/(min*cm^2)

As follows from the 1999 survey data, the radiological hazard was highest at the tunnels V1 (with β-particle flux density as high as 2,500 particles/(min*cm^2)) and V2 (EDR up to 45 μSv/hour and α-particle flux density amounting to 8 particles /(min*cm^2)). Measured radiological parameters were within the allowable limits for tunnels H1, H2, H3, H4, V3, V4, and V5 [4].

Laboratory tests showed that the specific activity of radionuclides ^{137}Cs, ^{90}Sr, ^{241}Am and $^{239+240}$Pu in the samples taken after completing the works at the H1, H2, V3, V4 and V5 tunnel mouths did not exceed the standard levels of Minimal Significant Specific Activity (MSSA) stipulated for these radionuclides by the document NRB-99 (Radiation Safety Standards). As per NRB-99, the standard MSSA levels shall be as follows: $1 \cdot 10^4$ Bq/kg for ^{137}Cs, $1 \cdot 10^5$ Bq/kg for ^{90}Sr, $1 \cdot 10^3$ Bq/kg for ^{241}Am, and $1 \cdot 10^3$ Bq/kg for $^{239+240}$Pu. High concentrations of transuranium radionuclides were found in the near-portal areas of V1, V3 and H4 tunnels. Figure 6 shows maximum specific activities of radionuclides as measured after sealing the tunnels.

As per the 1999 final survey, radiological hazard was the highest at the V2 tunnel mouth, with maximum concentrations being as follows: $9.7 \cdot 10^4$ Bq/kg for ^{137}Cs, $2.3 \cdot 10^6$ Bq/kg for ^{241}Am, and $2.0 \cdot 10^8$ Bq/kg for $^{239+240}$Pu. These activities are way above the standard MSSA level.

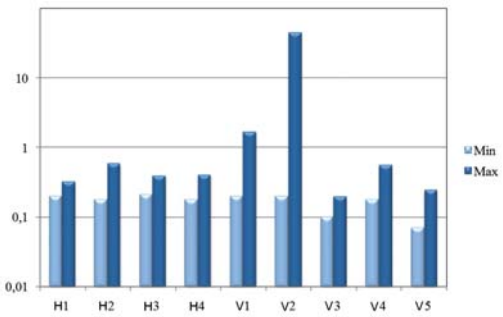


Figure 5. Maximal and minimal EDR values obtained during the 1999 final survey, *μSv/hour*

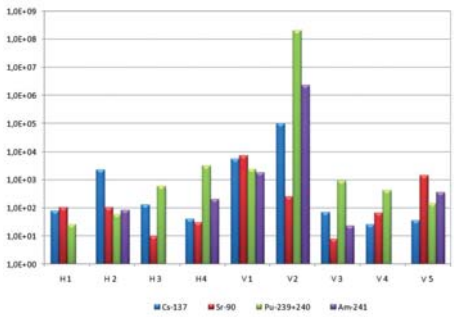


Figure 6. Maximum specific activities of radionuclides obtained during the 1999 final survey, *Bq/kg*

1.3. Findings of the 1999 tunnel survey

High concentrations of ^{90}Sr, ^{99}Tc, ^{137}Cs, ^{151}Sm, ^{241}Am and plutonium isotopes would be expected within the near-portal areas of the surveyed tunnels.

As follows from the 1999 field survey, the radiological hazard was highest at the tunnels V1 (with β-particle flux density as high as 2,500 particles/(min*cm^2)) and V2 (EDR up to 45 μSv/hour and α-particle flux density amounting to 8 particles /(min*cm^2)). Measured radiological parameters were within the allowable limits for tunnels H1, H2, H3, H4, V3, V4, and V5 [4].

Laboratory tests ascertained that the specific activity of radionuclides ^{137}Cs, ^{90}Sr, ^{241}Am and $^{239+240}$Pu in the samples taken after completing the works at the H1, H2, V3, V4 and V5 tunnel mouths did not exceed the standard levels of Minimal Significant Specific Activity (MSSA) [4]. High concentrations of transuranium radionuclides were found in the near-portal areas of V1, V3 and H4 tunnels. The 1999 final survey showed that radiological hazard was the highest at the V2 tunnel mouth.

2. KEY FACTORS AFFECTING RADIOLOGICAL CONDITIONS WHILE BUILDING SUPPLEMENTARY PROTECTION

Factors affecting radiological conditions can be subdivided into 3 groups:

- common factors, which do not depend on the type of operations for building supplementary protection;
- factors specific to the vertical method; and
- factors specific to the horizontal method.

2.1. Common factors

Common factors affecting radiological conditions include the existing radioactive contamination of the work site. This type of contamination is a legacy of nuclear tests conducted many years ago and has been identified during the preliminary survey. To reduce the personnel exposure, specific actions are to be taken, such as radiological decontamination, backing with a layer of uncontaminated soil, etc. In addition, this type of contamination can be caused when conducting site-leveling operations if any highly contaminated piles (stacks) have been disturbed.

Radiological conditions can also be affected by highly radioactive objects located in a close proximity to the work sites (contaminated mouths of other tunnels and piles/stacks with high radioactive contamination of their surfaces). Secondary transport of contaminants (air-borne dust having high specific activity of radionuclides and/or soil ablation) can bring such contaminants into the surveyed tunnel mouths and change their radiological conditions. In addition, such secondary transport can increase radioactivity concentrations in the bottom layer of the atmosphere.

2.2. Factors specific to the vertical method of building supplementary protection

When vertical method is used to enhance the existing protection, radiological conditions caused by common factors can be aggravated by potential contamination of the work site during drilling operations. For example, contaminants can be brought from the tunnels together with drill cuttings, as well as together with dust (in the form of aerosols) emitted through the boreholes either while drilling additional boreholes or while building supplementary protection (when concrete is injected into the tunnels). Secondary transport of contaminants (air-borne dust, movement of the equipment and personnel) can also cause changes in the radiological conditions within the work sites.

These factors can change radiological conditions at the work sites and bring about increased radioactivity concentrations in the bottom layer of the atmosphere.

2.3. Factors specific to the horizontal method of building supplementary protection

When the horizontal method is implemented for providing supplementary protection, the radiological conditions of the tunnel near-portal areas can be affected by potential entry of contaminants into the environment together with the drilled out materials/rocks. Though the radiation level of drilled out materials is continuously monitored, still there is a risk that radioactive contaminants can get into the near-portal areas because of the specific character of operations (movements of personnel, transportation vehicles and other equipment). Tunnel ventilation can

also cause emission of radioactive aerosols into the tunnel mouth areas. Radioactive aerosol emissions can result in increased radioactivity concentrations in the bottom layer of the atmosphere and increased specific activity of the soil in the event of their fallout.

Radiological conditions of the near-portal areas can also be affected by the secondary transport of contaminants by personnel and equipment either from the tunnels or from piles of materials/rocks removed from the tunnels.

3. RADIATION SURVEY TECHNIQUES

Initial data were studied and estimates were made while planning for the field surveys and laboratory tests. Based on the preliminary work, it was inferred that ^{90}Sr, ^{137}Cs, ^{151}Sm, ^{241}Am, and Pu isotopes are most likely to be found in the tunnel near-portal areas and around them.

A uniformed grid was used for the field surveys and sampling. EDR's and topsoil contamination with α and β particles were determined at fixed points. Using Search mode, EDR's were also determined in some intermediate points. Additional soil samples were taken in highly contaminated areas.

Depending on topography, surveyed areas varied in the range from 8,000 to 44,000 m^2. A GPS receiver was used to locate grid point coordinates. Fixed-point EDR measurements were taken in the 10m x 5m grid points of the tunnel mouth areas. A 20 m x 20m grid was used to measure α- and β-particle flux density.

Where the vertical method was to be used, additional EDR measurement points were chosen by applying the Search mode over the segments approximately 50 m wide and 250 m long, which were located along the tunnel central lines, with the zero end being at the bore cut-in point.

Multipoint sampling included the following:

- sampling point of soils within the proposed work site;
- sampling point of soils within the proposed field camp area;
- sampling point of soils over the uniform grid covering the entire survey area (the number of sampling points makes approximately 5-8% of the total number of points, where radiation parameters would be measured);
- composite soil samples taken from piles/stacks and tunnel backfills;
- sampling point of soils in the areas exhibiting high radiation parameters; and
- water vapour and air samples taken at the proposed bore cut-in points.

Gamma-ray, beta-ray and alpha spectrometry was used to conduct laboratory tests.

Final survey design was similar to the preliminary survey.

4. RADIOLOGICAL CONDITIONS OF THE TUNNEL NEAR-PORTAL AREAS BEFORE THE BEGINNING OF WORKS (2006-2009)

4.1. Radiological conditions as per radiometric measurements and radiation dosimetry

The preliminary survey data were used to build schematic maps of radiation distribution over the areas in question (Figure 9 – Figure 26).

The diagram below shows maximal and minimal EDR values measured during the preliminary survey of the tunnels (Figure 7).

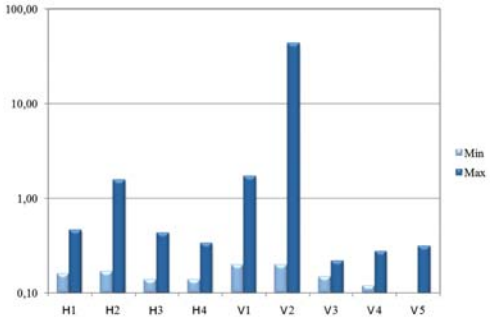


Figure 7. Maximal and minimal EDR values obtained during the preliminary survey of the tunnels, *μSv/hour*

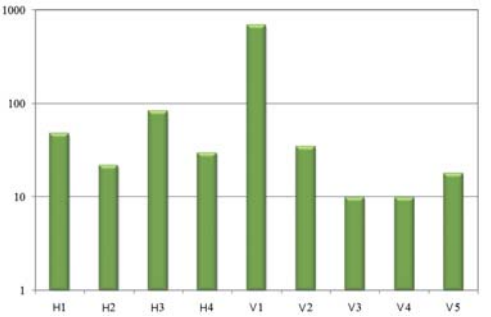
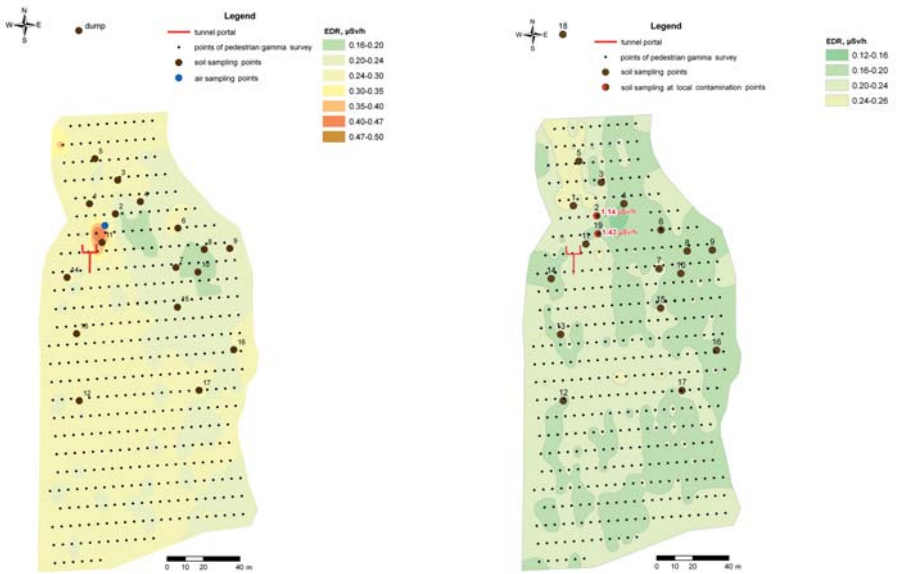


Figure 8. Maximum flux densities of β-particles obtained during the preliminary survey of the tunnels, *particles/(min*cm²)*

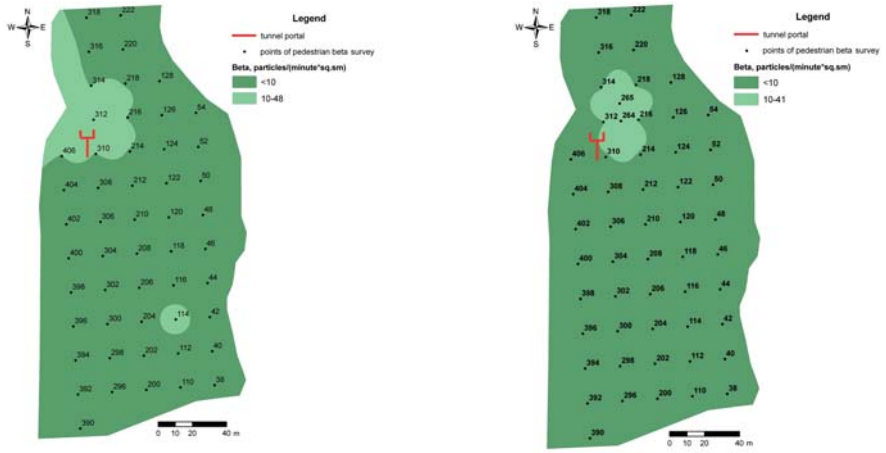
As per the preliminary survey, minimum β-particle flux densities were below 10 particles/(min*cm²) in all the tunnel mouth areas. Maximum β-particle flux densities measured during the preliminary survey are given in the diagram below (Fig. 8).

Flux densities of α-particles, registered during the preliminary survey, were below the detection limits (0.2 – 10 particles/(min*cm²)) of the used measuring instruments for nearly all investigated areas with the exception of two tunnel mouth areas, namely: H4 (140 particles/(min*cm²)) and V2 (100 particles/(min*cm²)).



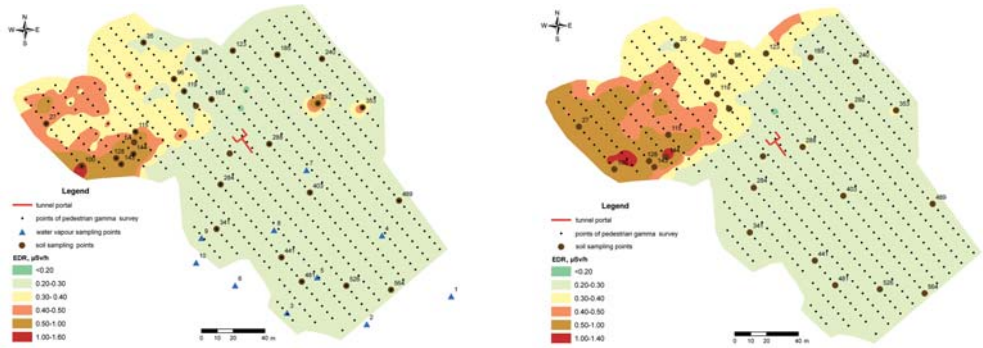
a) b)

Figure 9. EDR distributions over the H1 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



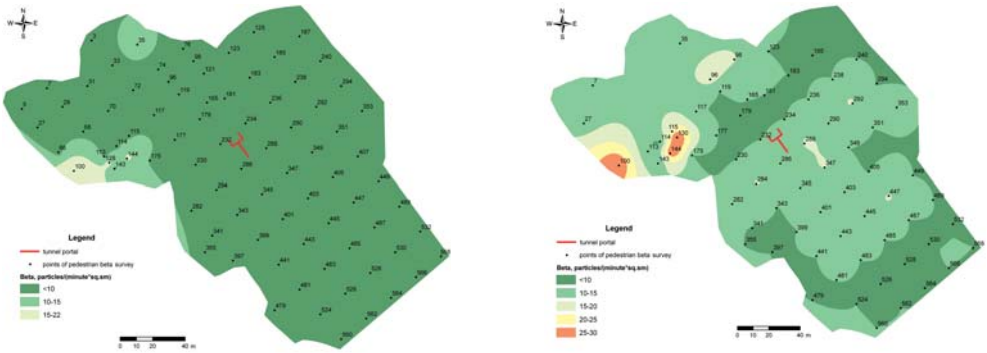
a) b)

Figure 10. Distributions of β-particles flux density over the H1 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



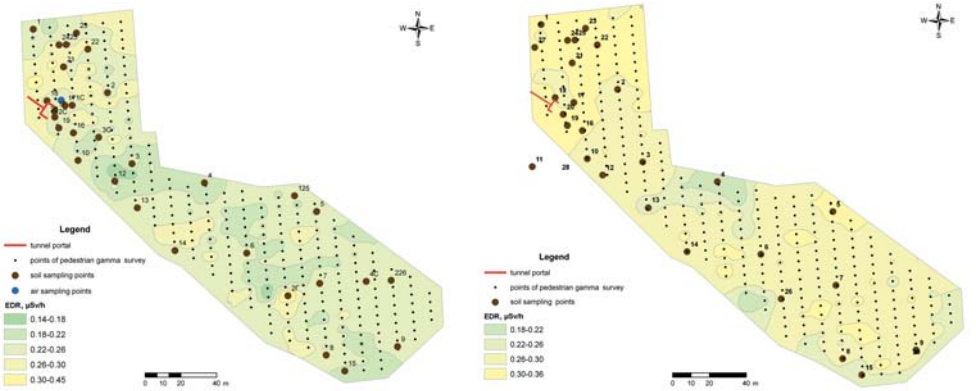
a) b)

Figure 11. EDR distributions over the H2 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



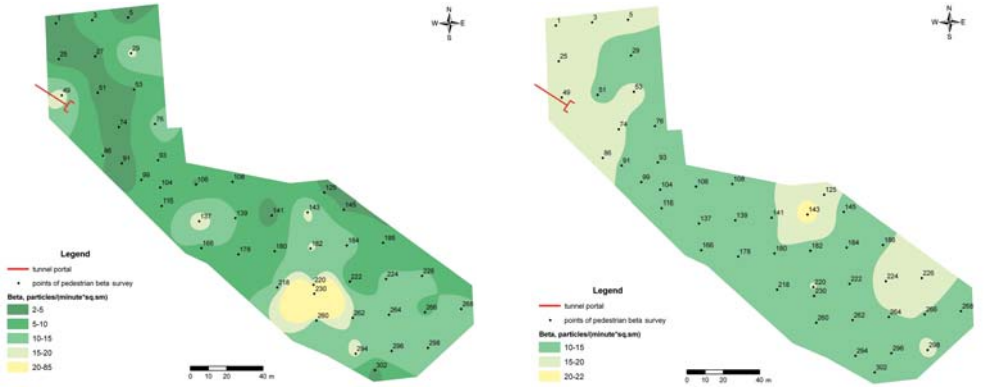
a) b)

Figure 12. Distributions of β-particles flux density over the H2 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



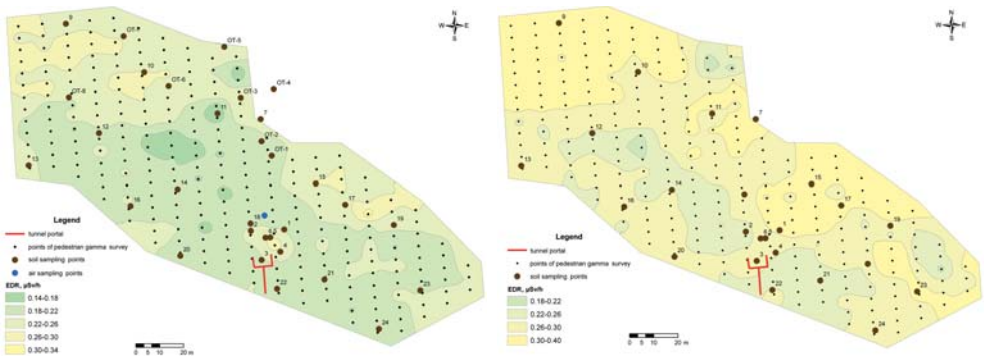
a) b)

Figure 13. EDR distributions over the H3 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



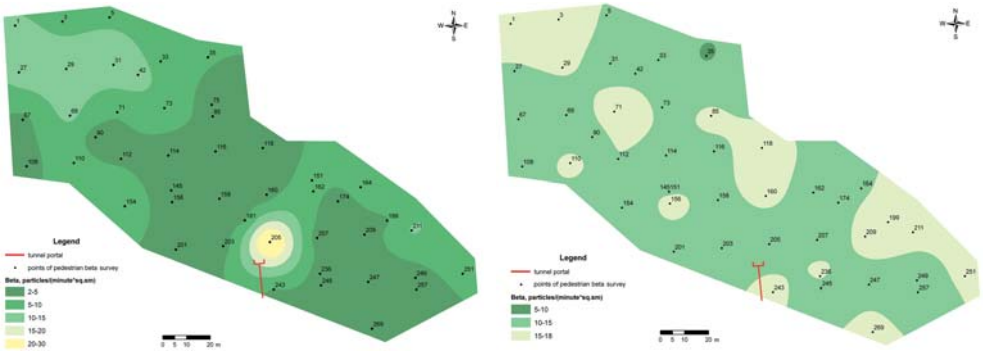
a) b)

Figure 14. Distributions of β-particles flux density over the H3 tunnel mouth area as registered during the preliminary survey (a)



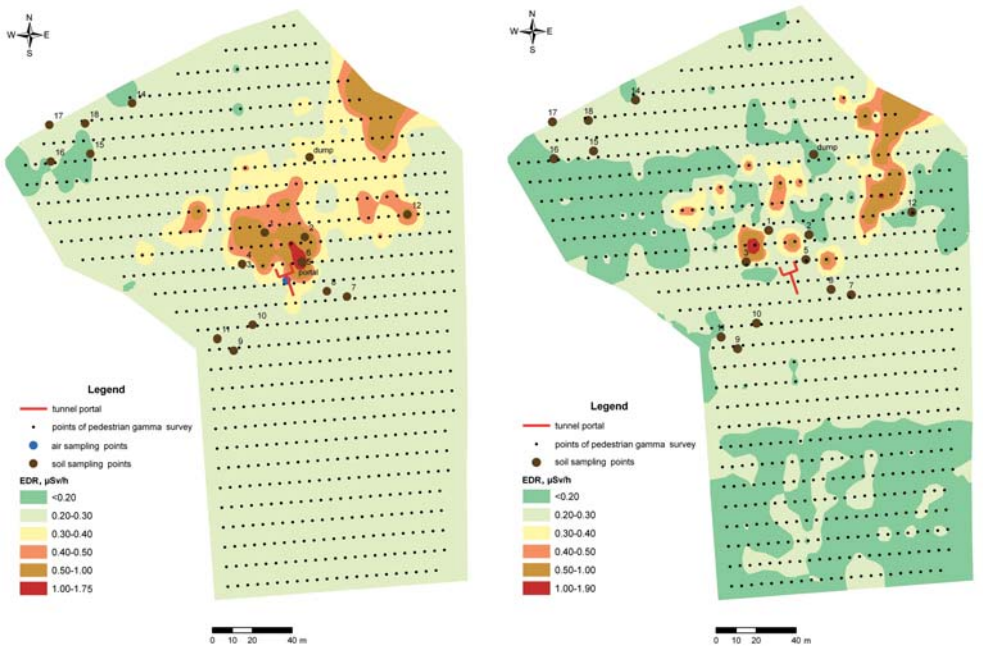
a) b)

Figure 15. EDR distributions over the H4 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



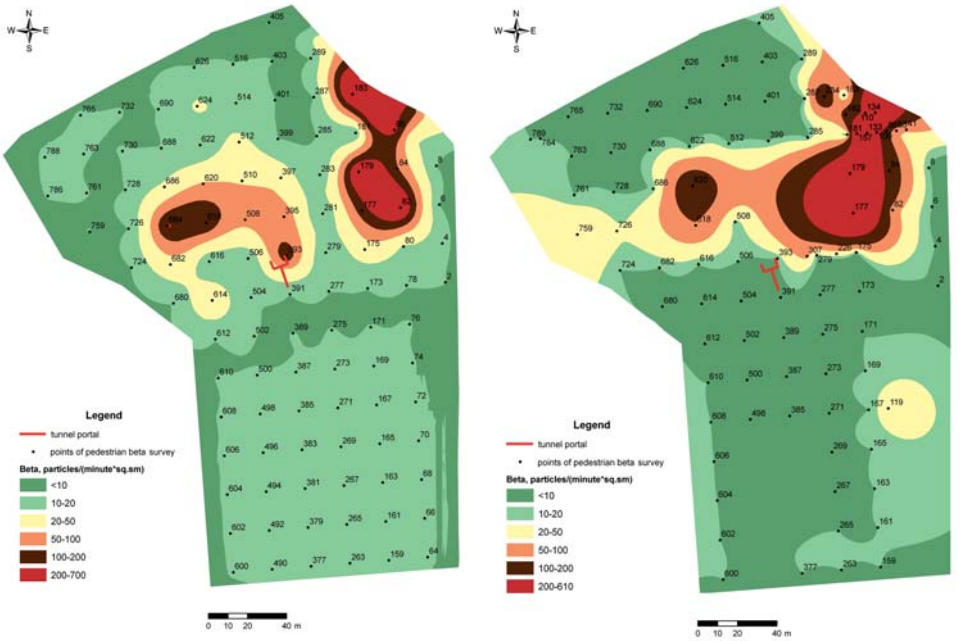
a) b)

Figure 16. Distributions of β-particles flux density over the H4 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



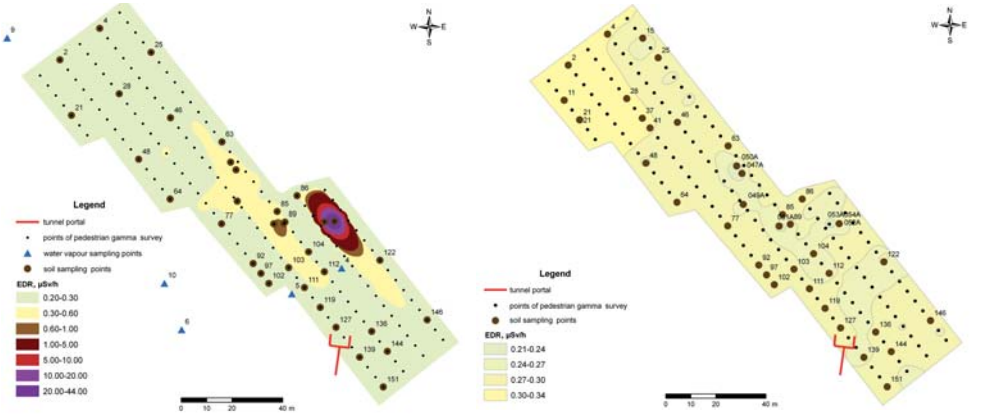
a) b)

Figure 17. EDR distributions over the V1 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



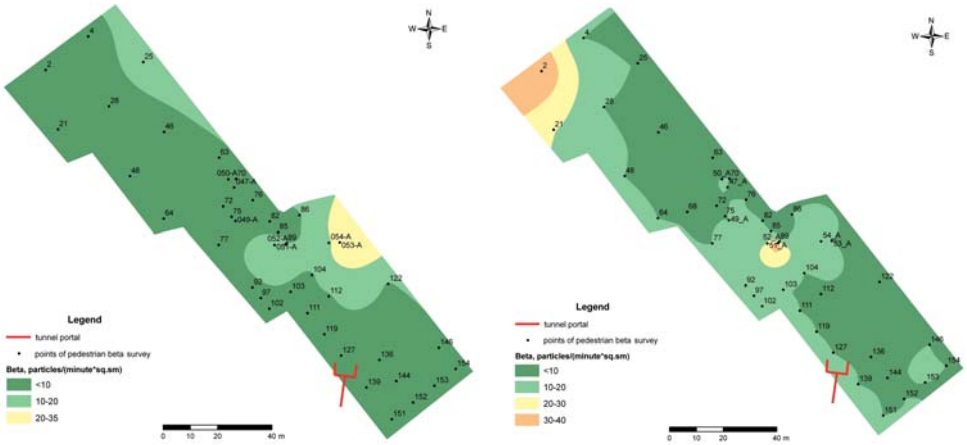
a) b)

Figure 18. Distributions of β-particles flux density over the V1 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



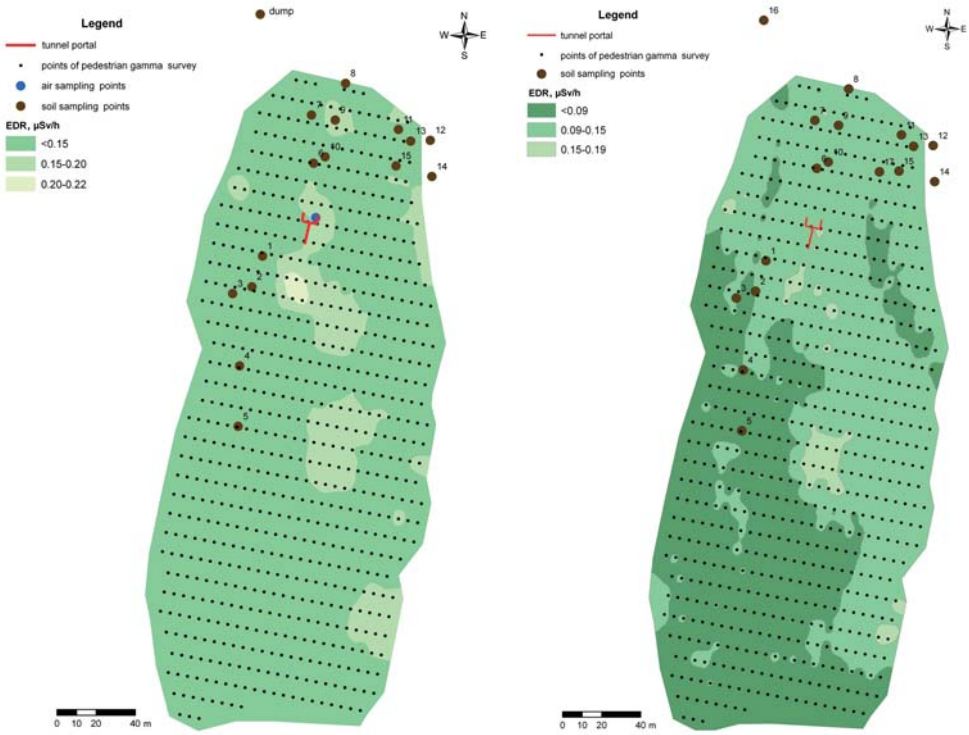
a) b)

Figure 19. EDR distributions over the V2 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



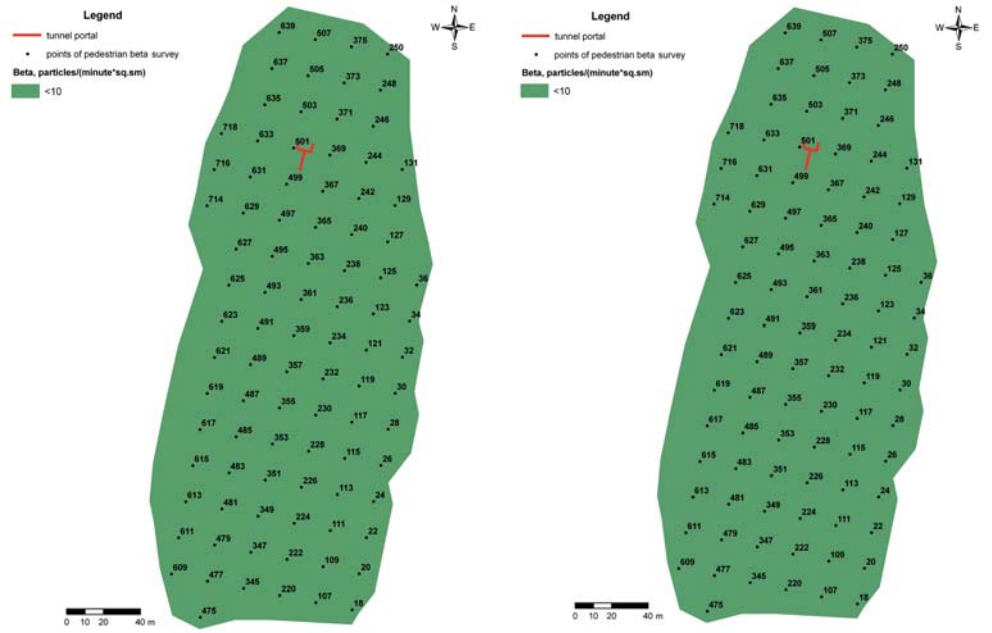
a) b)

Figure 20. Distributions of β-particles flux density over the V2 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



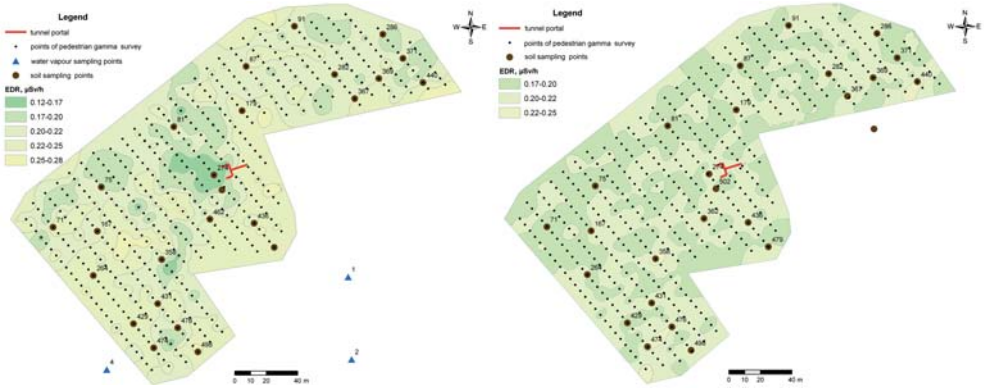
a) b)

Figure 21. EDR distributions over the V3 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



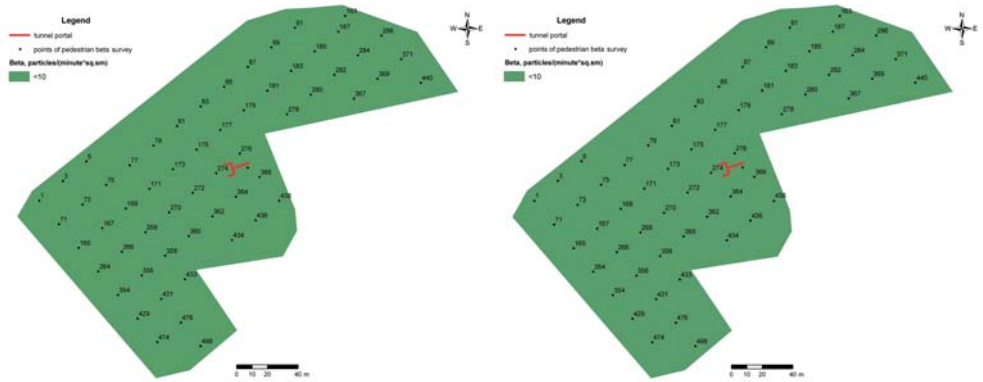
a) b)

Figure 22. Distributions of β-particles flux density over the V3 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



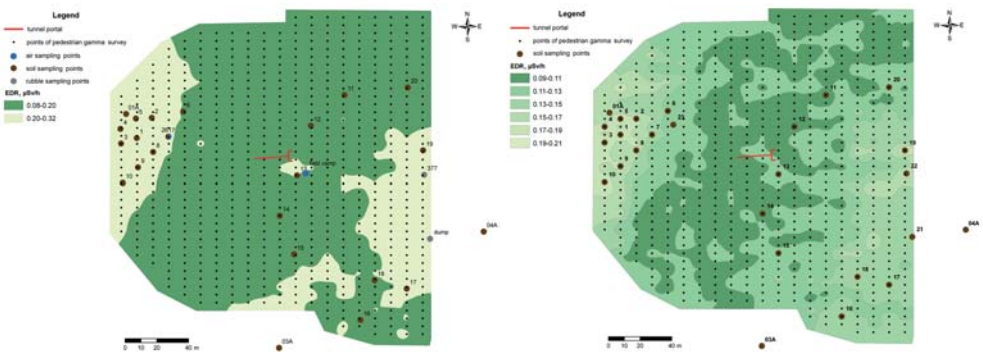
a) b)

Figure 23. EDR distributions over the V4 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



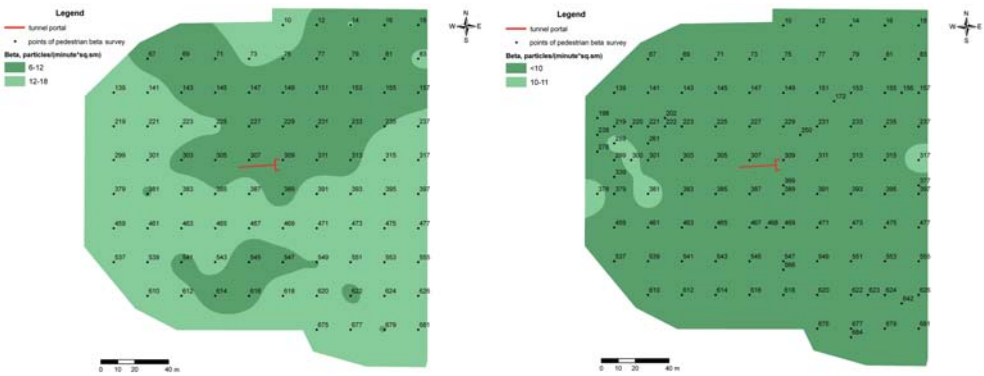
a) b)

Figure 24. Distributions of β-particles flux density over the V4 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



a) b)

Figure 25. EDR distributions over the V5 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)



a) b)

Figure 26. Distributions of β-particles flux density over the V5 tunnel mouth area as registered during the preliminary survey (a) and final survey (b)

Comparison of the 1999 tunnel survey results with the results obtained before the beginning of works showed that the radiation hazard was highest in the V2 tunnel mouth area in both cases. A part of this area exhibited EDR values as high as 44 μSv/hour and α-particle flux densities up to 100 particles/(min*cm^2), which are higher than allowable rates for Category A personnel permanently present in contaminated areas (Figure 19a) [4]. Radiation parameters exceeding NRB-99 levels (Radiation Safety Norms regulatory doc) were also measured in the V4 area (with α-particle flux density of 140 particles/(min*cm^2)) [4]. It should be noted that radiation parameters in the V1 mouth area were within the allowable levels, but a highly contaminated 330 m^2 spot was found in this mouth area, which had quite high β-particle flux densities (100 – 700 particles/(min*cm^2)). This contamination was caused by radioactive contaminants washed out from the tunnel by water.

As compared to the 1999 data, the radiological conditions changed in the H1, H3 and V2 near-portal areas and significantly deteriorated in the H2, H4 and V2 mouth areas. It can be explained by the fact that these mouths were broken in more than once and contaminated soils and metal structures were moved to the day surface. For example, an unauthorized manhole leading to the H4 tunnel (Figure 27) was found during the preliminary survey and metal structure pieces were found outside the surveyed area (with EDR values of 23 μSv/hour and α-particle flux density of 140 particles/(min*cm^2)).

Figure 27. Manhole to the V4 tunnel

4.2. Topsoil contamination with radionuclides

To tentatively estimate the level of radionuclide contamination of the surveyed area, spectrometric and radiochemical analyses were carried out for soil samples collected during the preliminary survey of 9 tunnel near-portal areas located in Degelen massif.

Naturally occurring radionuclide concentrations in the investigated tunnel near-portal areas correspond to the natural background of the region (predominant rocks are granites). As to artificial radionuclides, the following specific activity values were found as background counts: 30 Bq/kg for ^{137}Cs, 20 Bq/kg for ^{90}Sr, 0.2 Bq/kg for ^{241}Am, and 1 Bq/kg for $^{239+240}$Pu.

Table 2 and the diagram in Figure 28 show highest specific activities of ^{137}Cs, ^{90}Sr, ^{241}Am and $^{239+240}$Pu measured during the preliminary survey of tunnel mouths.

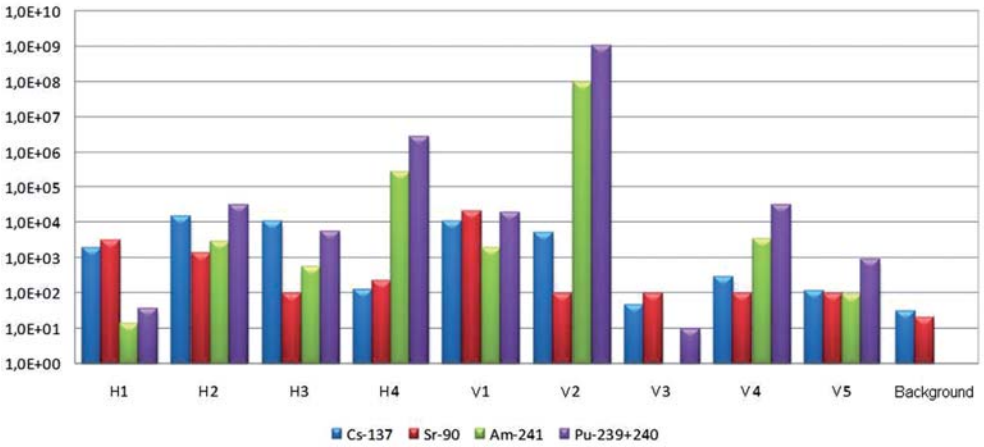


Figure 28. Maximum specific activities of radionuclides measured during the tunnel mouths preliminary survey, *Bq/kg*

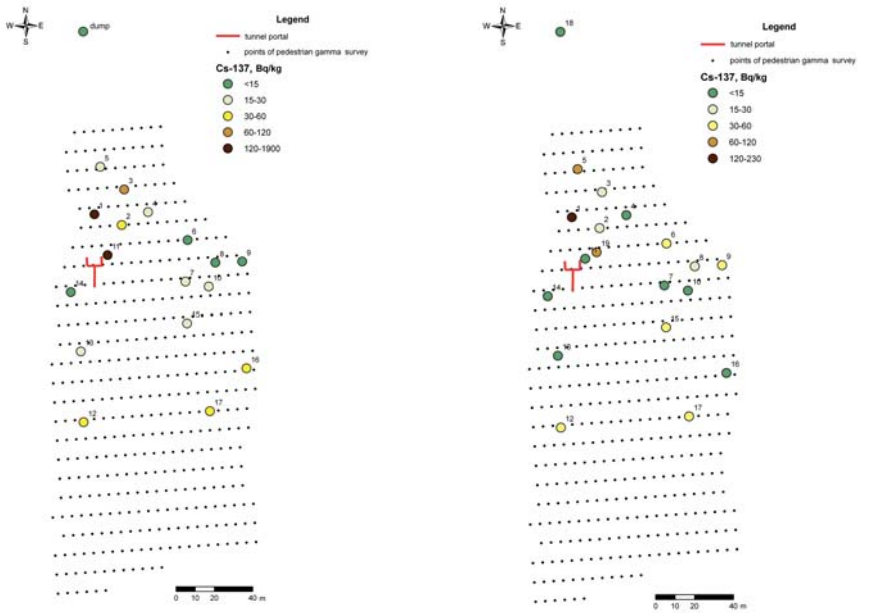
For several points, concentrations of $^{239+240}$Pu were calculated based on the laboratory test data. As proven for H2, V2 and V4 tunnel mouths, the ratio of ^{241}Am to $^{239+240}$Pu concentrations was 1:11 in soil samples collected from these mouth areas. In all other cases, the average ratio of ^{241}Am to $^{239+240}$Pu was taken equal to 1:10, which was determined based on the STS soil laboratory test data and was the most common for the STS as a whole.

Maximum measured ^{90}Sr concentrations for the surveyed near-portal areas did not exceed $1 \cdot 10^5$ Bq/kg, which is the MSSA for this radionuclide.

Based on the comparison of the maximum observed specific activities of ^{137}Cs, ^{241}Am and $^{239+240}$Pu with their standard levels (MSSA, NRB-99), the tunnel mouths were grouped as follows:

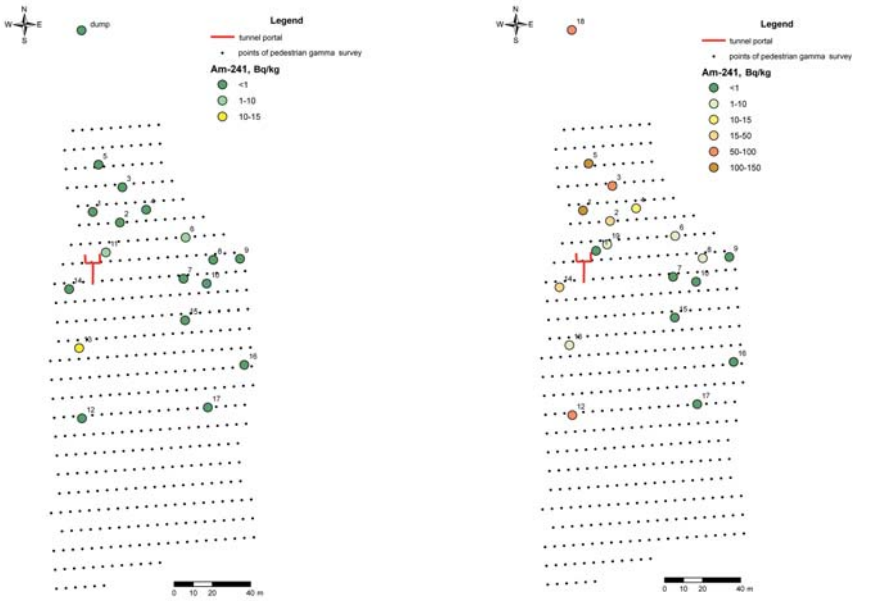
1. tunnel mouth areas, where soil concentrations of radionuclides in question do not exceed the standard levels (H1, V3 and V5 tunnel mouths);
2. tunnel mouth areas, where soil concentrations of ^{137}Cs do not exceed the standard MSSA level of $1 \cdot 10^4$ Bq/kg, while concentrations of ^{241}Am and $^{239+240}$Pu transuranium radionuclides exceed the MSSA level of $1 \cdot 10^3$ Bq/kg (H4, V2 and V4 tunnel mouths);
3. tunnel mouth areas, where soil concentrations of ^{137}Cs exceed the standard MSSA level of $1 \cdot 10^4$ Bq/kg and concentrations of ^{241}Am and $^{239+240}$Pu transuranium radionuclides also exceed the MSSA level of $1 \cdot 10^3$ Bq/kg (H2, V3 and V1).

Tunnels belonging to the first group are most radiologically safe. Soil concentrations of ^{137}Cs, ^{90}Sr, ^{241}Am and $^{239+240}$Pu in the H1, V3 and V5 were below the standard limits (MSSA, NRB-99), i.e. below $1 \cdot 10^4$, $1 \cdot 10^5$, $1 \cdot 10^3$ and $1 \cdot 10^3$ Bq/kg, respectively (Figure 29 – Figure 34). During the preliminary stage, maximum concentrations observed in these near-portal areas were as follows: $1.9 \cdot 10^3$ Bq/kg for ^{137}Cs, $3.1 \cdot 10^3$ Bq/kg for ^{90}Sr (H1), 90 Bq/kg for ^{241}Am, and 900 Bq/kg for $^{239+240}$Pu (V5 tunnel mouth).



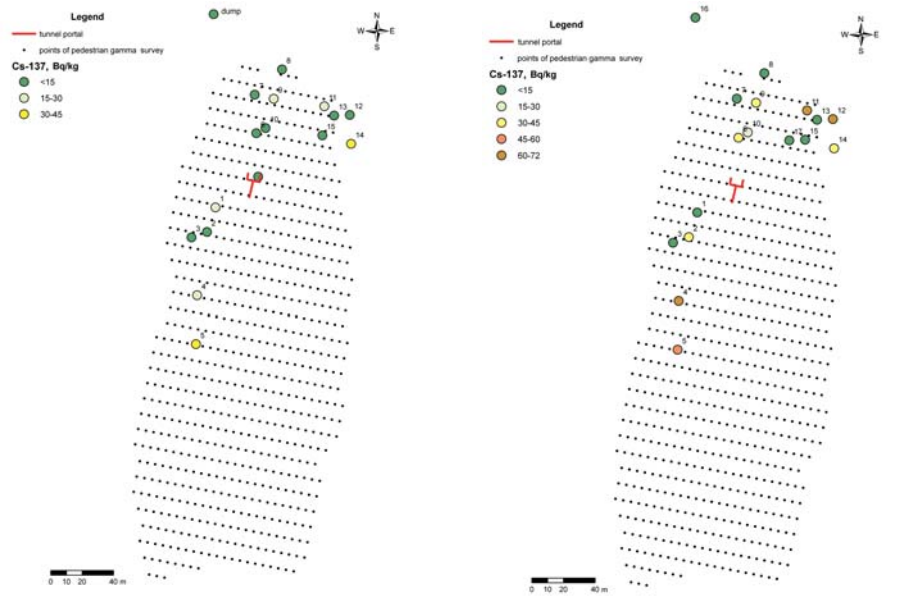
a) b)

Figure 29. Distribution of ^{137}Cs over the H1 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)



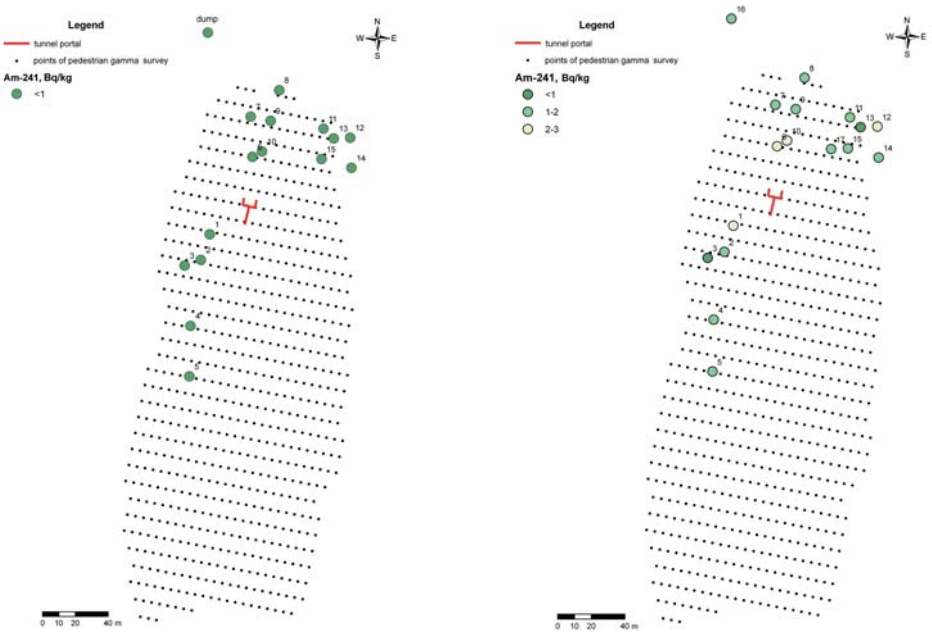
a) b)

Figure 30. Distribution of ^{241}Am over the H1 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)



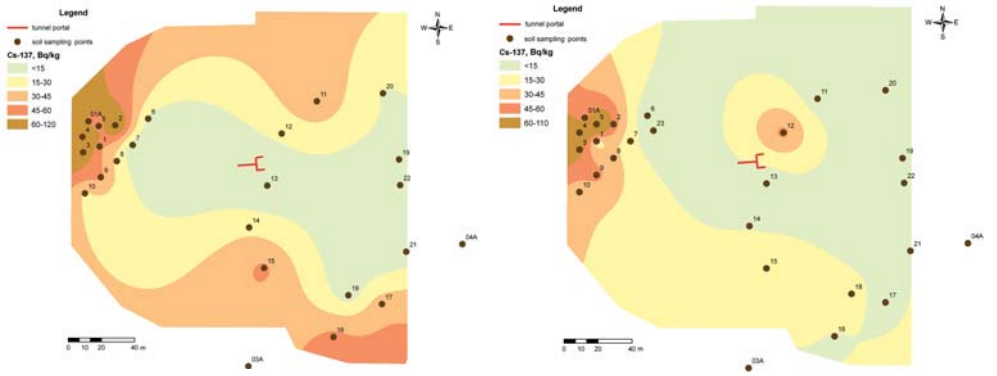
a) b)

Figure 31. Distribution of ^{137}Cs over the V3 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)



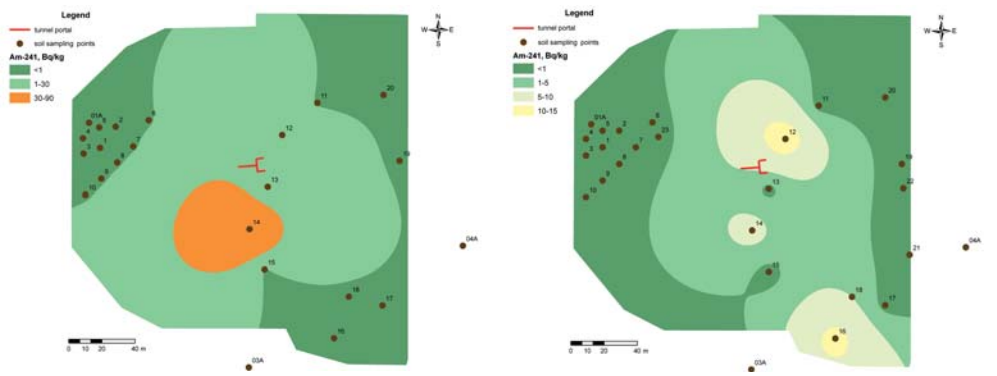
a) b)

Figure 32. Distribution of ^{241}Am over the V3 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)



a) b)

Figure 33. Distribution of ^{137}Cs over the V5 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)



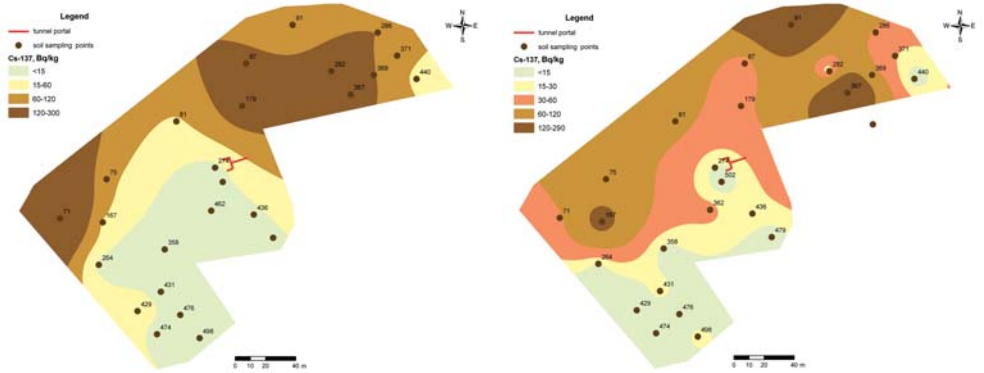
a) b)

Figure 34. Distribution of ^{241}Am over the V5 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)

The second group includes V4, H4 and V2 tunnel mouth areas, which are characterized by elevated concentrations of ^{241}Am and $^{239+240}$Pu in their soils. Radionuclides ^{241}Am, ^{137}Cs and ^{239,240}Pu are ubiquitous in the V4 tunnel mouth area (Figure 35, Figure 36). Maximum concentration of ^{137}Cs in this area was 300 Bq/kg, which is below the allowable limit. Artificial radionuclides ^{241}Am and $^{239+240}$Pu were found in virtually all investigated points, with maximum values being as follows: $3.3 \cdot 10^3$ Bq/kg for ^{241}Am and $3.3 \cdot 10^4$ Bq/kg for $^{239+240}$Pu (the area of piles and backfills).

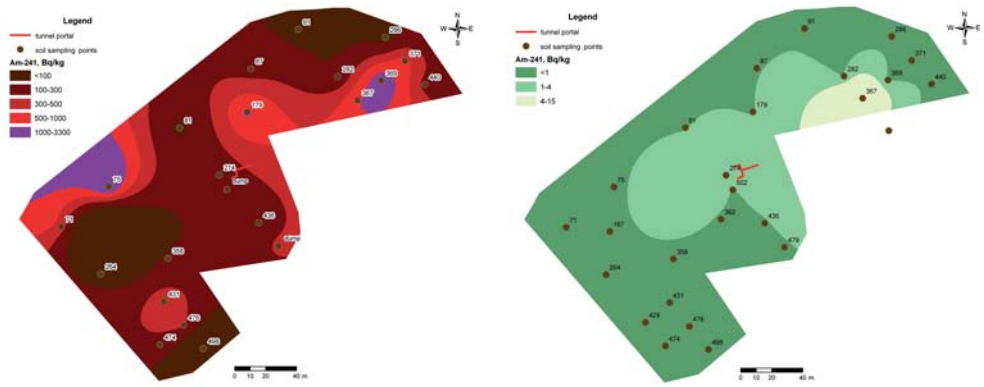
The maximum concentration of ^{137}Cs observed in the H4 mouth area was 120 Bq/kg and that of ^{241}Am was 250 Bq/kg. The only exclusion here is a localized contamination spot caused by unauthorized activities, such as burning off cable insulations, which had taken place before the resealing operations (Figure 37 and Figure 38). The maximum concentration of ^{241}Am was $2.8 \cdot 10^5$ Bq/kg. Given that the average ^{241}Am to $^{239+240}$Pu ratio is 1:10, the anticipated concentration of ^{239,240}Pu can attain $2.8 \cdot 10^6$ Bq/kg. Based on the ^{241}Am and ^{239,240}Pu concentrations observed in the soil samples collected from these points, the soils can be clas-

sified as the medium active waste (from 10^1 to 10^5 kBq/kg) [4, 5]. Quantity of radioactive waste located within the work site of this mouth area can vary from several cubic meters to dosens or hundreds of cubic meters. This localized contamination spot is small (< 2 m^2). However, since it is located in the backfill area of the H4 tunnel mouth area, it is quite likely that big quantities of contaminated soils can be found in the subsurface.



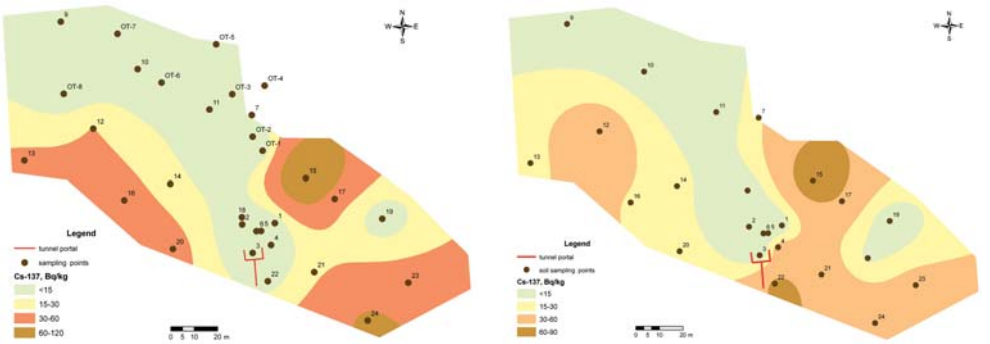
a) b)

Figure 35. Distribution of ^{137}Cs over the V4 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)



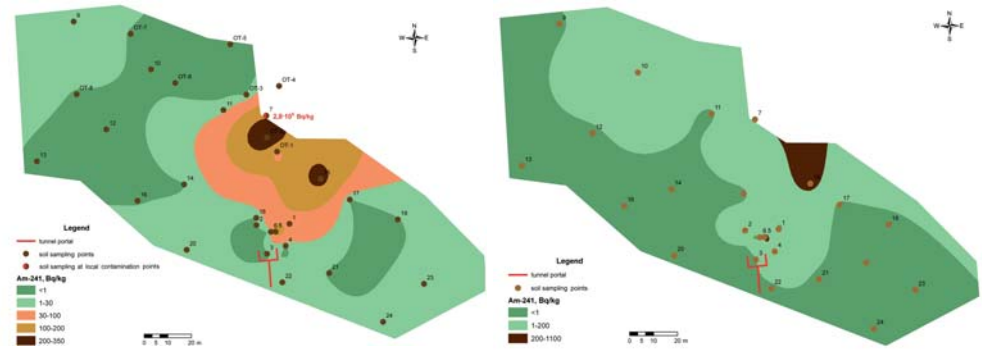
a) b)

Figure 36. Distribution of ^{241}Am over the V4 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)



a) b)

Figure 37. Distribution of ^{137}Cs over the H4 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)

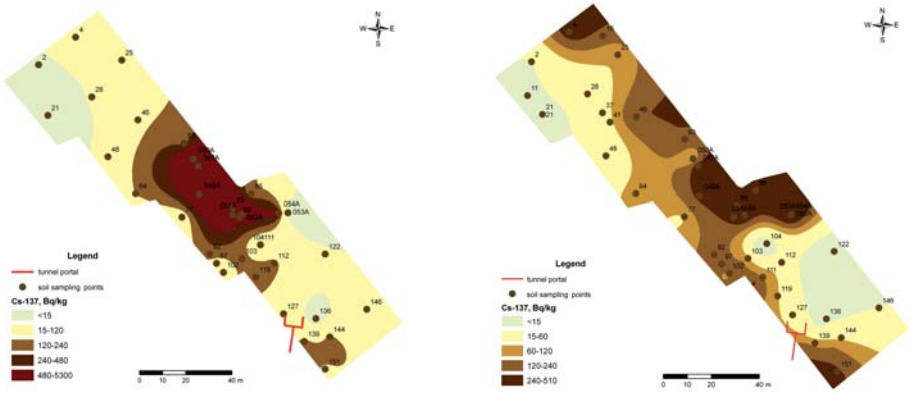


a) b)

Figure 38. Distribution of ^{241}Am over the H4 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)

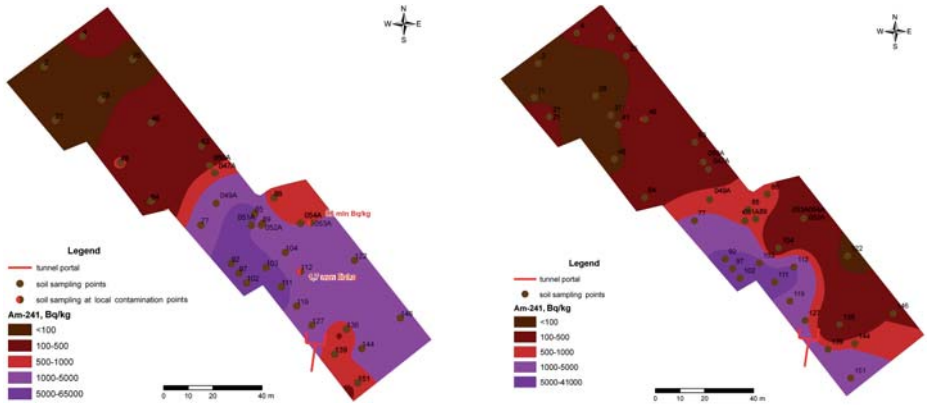
Preliminary investigation data obtained for the V2 tunnel mouth evidence that the entire mouth area is contaminated with the artificial ^{241}Am radionuclide, specific activity of which varies in the range from <3 up to $9.6 \cdot 10^7$ Bq/kg (Figure 39 and Figure 40). Specific activity of $^{239+240}$Pu in virtually all measurement points of the V2 mouth area is in excess of the standard level (MSSA for $^{239+240}$Pu is $1 \cdot 10^3$ Bq/kg). During the resealing operations, special attention was paid to the points designated as 053A and 112. ^{241}Am concentration in these points was $9.6 \cdot 10^7$ and $1.7 \cdot 10^6$ Bq/kg, respectively, while specific activity of ^{239,240}Pu can be as high as $1 \cdot 10^9$ and $2 \cdot 10^8$ Bq/kg, respectively.

^{137}Cs concentrations over the V2 mouth area vary in the wide range of activities and reach $5.3 \cdot 10^3$ Bq/kg.



a) b)

Figure 39. Distribution of ^{137}Cs over the V2 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)



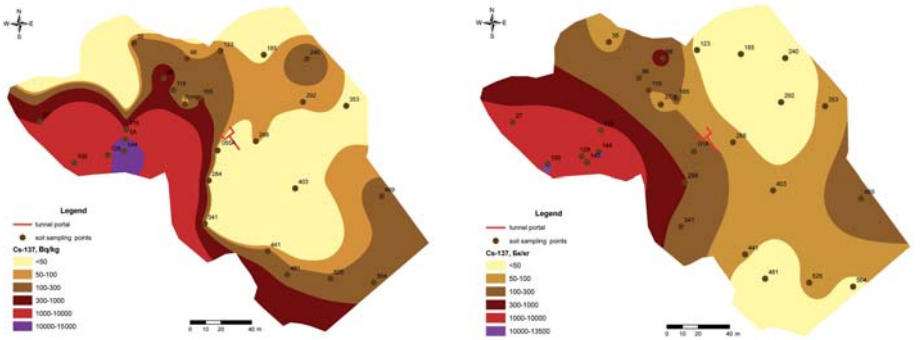
a) b)

Figure 40. Distribution of ^{241}Am over the V2 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)

The third group includes H2, H3 and V1 near-portal areas and is characterized by elevated (higher than MSSA) concentrations of ^{137}Cs and transuranium radionuclides (^{241}Am and $^{239+240}$Pu). Maximum observed specific activities were as follows: $1.5 \cdot 10^4$ Bq/kg for ^{137}Cs, $3.0 \cdot 10^3$ Bq/kg for ^{241}Am, and $3.3 \cdot 10^4$ Bq/kg for $^{239+240}$Pu (H2 tunnel). While studying the H2 tunnel mouth, the following contaminated areas were found: piles of soil/crushed rocks and some spots close to the mouth. A localized contamination was found at the V3 tunnel mouth, which most likely was caused by burning insulations of cables stolen from nearby tunnels. Distribution of ^{137}Cs and ^{241}Am over the H2 and H3 tunnel near-portal areas is given in the figures below (Figure 41 – Figure 43).

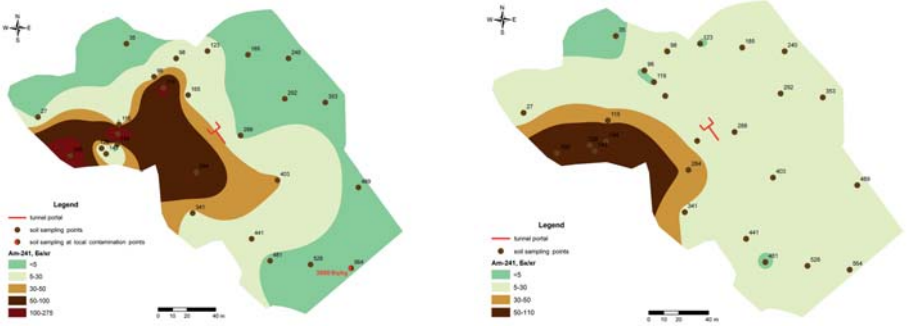
Three spots (points 3, 4 and 5) contaminated with ^{137}Cs and ^{241}Am were found in the V1 tunnel mouth area. Soils collected from Points 4 and 5 (located in the dry brook close to the tunnel mouth) can be classified into the category of limited-use materials (from 300 Bq/kg to MSSA). The soil collected from Point 3 is classified as radioactive waste. ^{137}Cs specific

activity in this point was $1.1 \cdot 10^4$ Bq/kg, while that of ^{241}Am was $1.9 \cdot 10^3$ Bq/kg. Anticipated content of Pu isotopes can be around $2.0 \cdot 10^4$ Bq/kg, which is a 20-fold increase over the minimal significant specific activity for this radionuclide (Figure 44 and Figure 45).



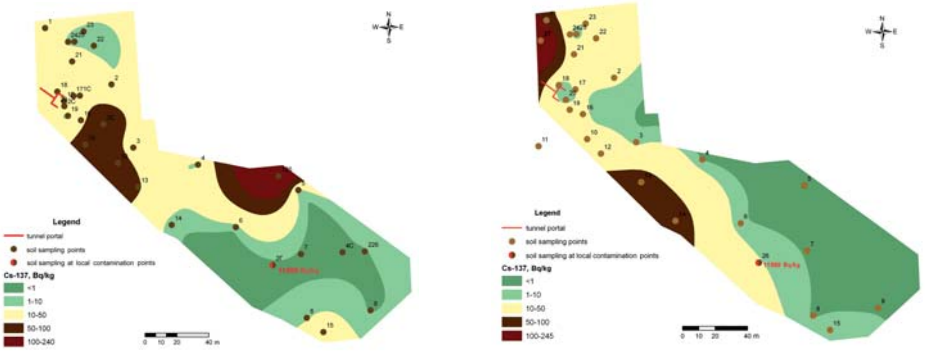
a) b)

Figure 41. Distribution of ^{137}Cs over the H2 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)



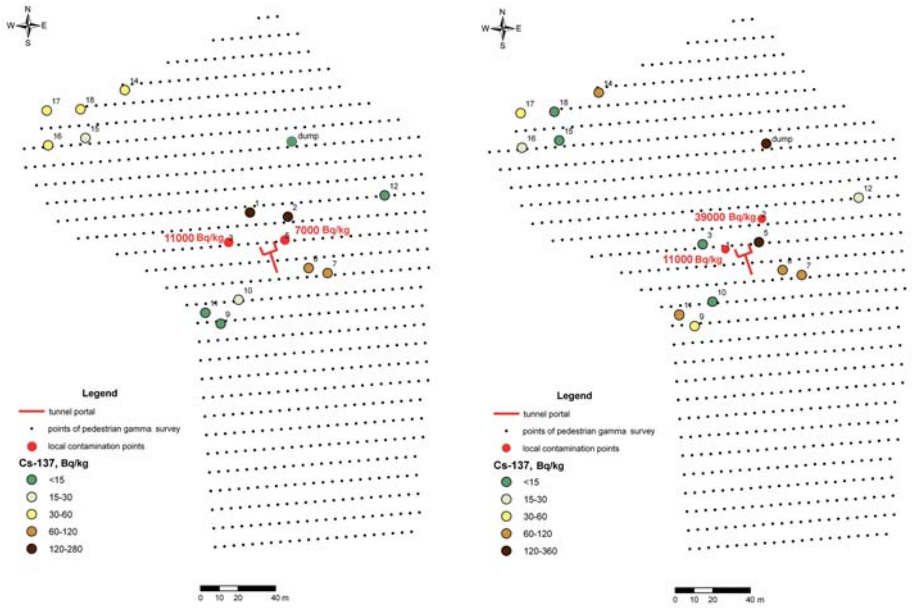
a) b)

Figure 42. Distribution of ^{241}Am over the H2 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)



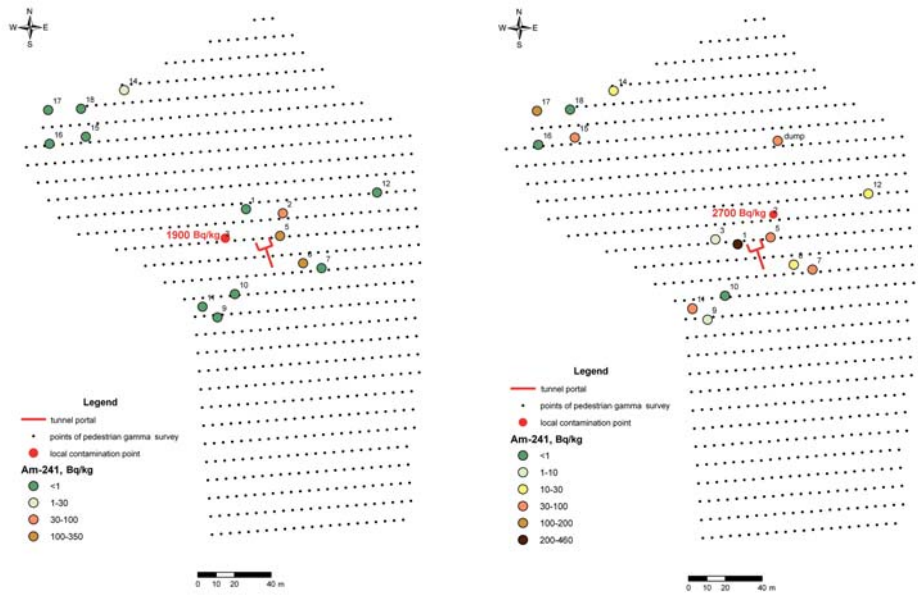
a) b)

Figure 43. Distribution of ^{137}Cs over the H3 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)



a) b)

Figure 44. Distribution of ^{137}Cs over the V1 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)



a) b)

Figure 45. Distribution of ^{241}Am over the V1 tunnel mouth area obtained during the preliminary survey (a) and final survey (b)

4.3. Bottom layer of the atmosphere contaminated with radionuclides

Such artificial radionuclides as ^{3}H, ^{90}Sr, ^{137}Cs, ^{241}Am and $^{239+240}Pu$ are the main radioactive materials characterizing the contamination in the bottom layer of the atmosphere. Direct methods (as per [6 and 7]) of measurement were used to determine volumetric activity of artificial radionuclides of the sampled aerosols and water vapours. Table 2 shows maximum volumetric activities of artificial radionuclides registered during the preliminary survey.

The survey data showed that ambient air concentrations of artificial radionuclides as measured in all the survey points were lower than the allowable volumetric activity stipulated by NRB-99 (Radiation Safety Standards) [4].

^{3}H concentrations in Degelen site air vary in the wide range from 0.2 to 240 Bq/m^3, which is one order of magnitude lower than the allowable volumetric activity for the Community Air category. However, under certain conditions, tritium can contribute quite heavily (up to 10%) into the radiation dose.

4.4. Preliminary survey findings

The preliminary survey indicated that the observed radiation hazard was the highest in the V2 tunnel mouth area. Radiation parameters in this tunnel mouth area exceeded the allowable values for *Category A* personnel in case they permanently stay on-site [4]. Comparison with the 1999 survey data showed that radiological conditions deteriorated in the H2, H4 and V2 tunnel near-portal areas as a result of break-in attempts, which resulted in removal of metal structures and soils from the tunnels onto the day surface.

During the preliminary survey, the following maximum specific activities were observed: $1.5 \cdot 10^4$ Bq/kg for ^{137}Cs in the H2 mouth area; $2.2 \cdot 10^4$ Bq/kg for ^{90}Sr in the V1 mouth area; $9.6 \cdot 10^7$ Bq/kg for ^{241}Am and $1.0 \cdot 10^9$ Bq/kg for $^{239+240}Pu$ in the V2 mouth area. Maximum observed specific activities for these radionuclides (with the exception of ^{90}Sr) exceed the standard levels stipulated by MSSA and NRB-99 [4].

^{137}Cs specific activities exceeded MSSA in three tunnel mouth areas, while ^{241}Am and $^{239+240}Pu$ specific activities exceeded MSSA in six mouth areas. Contaminant were found in the pile and backfill materials, and also in localized spots formed in the course of unauthorized activities.

Artificial radionuclide concentrations in the ambient air of all the surveyed areas were below the allowable volumetric activity levels stipulated by NRB-99 [4].

Prior to works, the areas with elevated measurements were decontaminated by covering them with fresh soil in order to avoid excessive exposure of the personnel.

5. RADIOLOGICAL CONDITIONS AT THE TUNNEL NEAR-PORTAL AREAS AFTER COMPLETION OF THE WORKS

5.1. Radiological conditions as per radiometric and dosimeter measurements

Schematic maps of radiation distribution over the surveyed areas were built based on the final survey results (Figure 9 – Figure 26).

Data on maximum and minimum measured EDR values for the period of the final survey are represented in the diagram below (Figure 46).

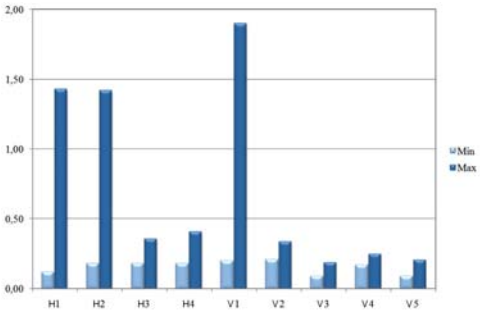


Figure 46. Maximum and minimum measured EDR values at the tunnel near-portal areas after completion of works, *μSv/h*

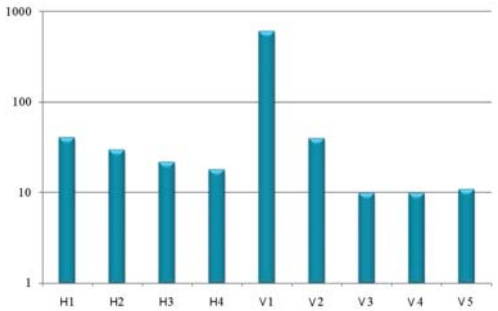


Figure 47. Maximum values of β-particle flux density for the preliminary survey of the tunnels, *particles/(min*cm²)*

Minimum values of β-particle flux density, recorded during the preliminary survey in all the tunnel mouth areas, did not exceed 10 particles/(min·sm^2). Maximum values of β-particle flux density, recorded during the preliminary survey, are represented in Figure 47.

Values of α-particle flux density, recorded during the final survey in all surveyed areas, did not exceed the detection limit of the used measuring devices (0.2 – 10 particles/(min*cm^2)).

According to the final survey data, highest radiation parameters were recorded in the following tunnel mouth areas: V1 (EDR – 1.9 μSv/h, β-particles flux density – 610 particles/(min*cm^2) (Fig. 17, b)); H1 and H2 (1.4 μSv/h) (Figure 9b and Figure 11b). The survey conducted upon completion of the supplementary protection works showed that radiation parameters did not exceed the allowable levels for the Category A personnel in all the tunnel near-portal areas [4].

5.2 Topsoil contamination with radionuclides

An assessment of residual contamination of the tunnel near-portal areas with artificial radionuclides was carried out at the final stage of resealing operations. Based on the measurements, schematic maps of ^{137}Cs and ^{241}Am distribution over the surveyed areas were built (Figure 29 - Figure 45).

Maximum specific activities of ^{137}Cs, ^{90}Sr, ^{241}Am and $^{239+240}$Pu obtained during the final survey of near-portal areas are given in the Table 2 and Figure 48.

As shown in the diagram, the highest concentration of ^{137}Cs radionuclide was recorded in the V1 tunnel mouth area ($3.9 \cdot 10^4$ Bq/kg), while that of ^{241}Am and plutonium isotope, in the V2 tunnel near-portal areas ($4.1 \cdot 10^4$ and $4.5 \cdot 10^5$ Bq/kg, respectively).

Maximum concentration of ^{90}Sr registered in the surveyed tunnel areas did not exceed MSSA for this radionuclide ($1 \cdot 10^5$ Bq/kg).

Having compared the maximum observed specific activities of ^{137}Cs, ^{241}Am and $^{239+240}$Pu radionuclides with the standard levels (MSSA, Radiation Safety Standards NRB-99), the tunnel near-portal areas can be divided into two groups depending on the nature of their contamination:

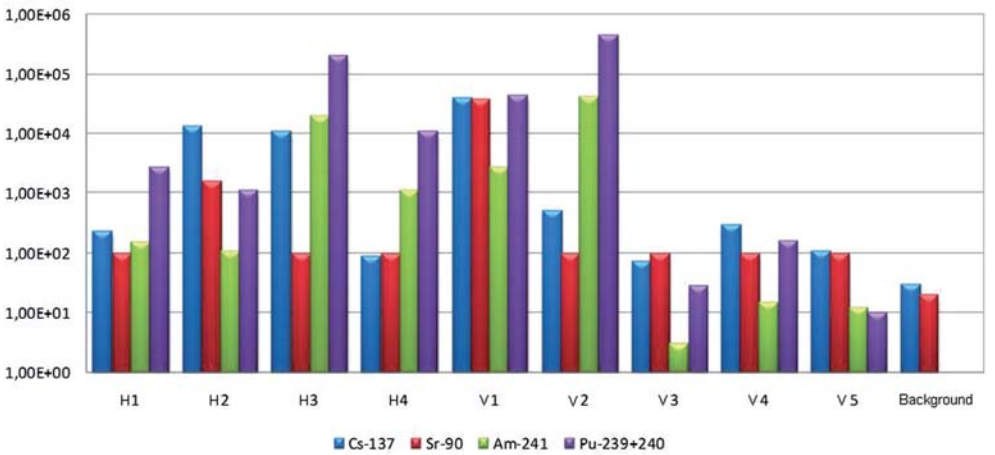


Figure 48. Maximum specific activities of radionuclides obtained during the final survey period, *Bq/kg*.

1. tunnels, where radionuclides concentrations in soil do not exceed the standard level for all radionuclides in question (V3, V4, and V5 tunnels);
2. tunnels, where maximum specific activities of ^{241}Am or $^{239+240}$Pu transuranium radionuclides exceed MSSA equal to $1 \cdot 10^3$ Bq/kg (H1-H4, V1, and V2 tunnels).

Radiation hazard is zero for the first group facilities. During the final stage, maximum concentrations in the tunnel near-portal areas were as follows: $2.9 \cdot 10^2$ Bq/kg for ^{137}Cs, 15 Bq/ kg for ^{241}Am, 165 Bq/kg for $^{239+240}$Pu, and <100 Bq/kg for ^{90}Sr (tunnel V4).

The second group of tunnels was characterized by elevated concentrations of ^{241}Am and/or $^{239+240}$Pu transuranium radionuclides in soils, which exceeded MSSA levels. Maximum observed specific activities were $4.1 \cdot 10^4$ Bq/kg for ^{241}Am and $4.5 \cdot 10^5$ Bq/kg for $^{239+240}$Pu (tunnel V2). These values exceed the standard level of $1 \cdot 10^3$ Bq/kg for such radionuclides (MSSA, Radiation Safety Standards 99), with the exceedances being as follows: 41 times for ^{241}Am and 450 times for $^{239+240}$Pu.

Concentrations of ^{137}Cs, ^{90}Sr and ^{241}Am in soils of the surveyed H1 tunnel near-portal areas do not exceed the standard levels (MSSA, Radiation Safety Standards NRB-99), equal to $1 \cdot 10^4$, $1 \cdot 10^5$ and $1 \cdot 10^3$ Bq/kg, respectively. Estimated data on Plutonium and radiochemical analysis of the collected samples showed that there was a risk that the $^{239+240}$Pu specific activity could be above the standard level (MSSA - $1 \cdot 10^3$ Bq/kg) in three points of the surveyed area (points 1, 5, and 18).

^{241}Am and ^{137}Cs were found in nearly all surveyed points of the H2 tunnel area. Maximum specific activity of ^{241}Am did not exceed 1/10 MSSA for this radionuclide (100 Bq/ kg), while maximum activity of ^{137}Cs was $1.4 \cdot 10^4$ Bq/kg. Concentration of $^{239+240}$Pu in the soil sampled from three points (100, 143, and 144) was within the limits of MSSA, with the observed maximum being $1.1 \cdot 10^3$ Bq/kg.

Laboratory tests of soil sampled from the H3 tunnel mouth area showed insignificant presence of the ^{137}Cs radionuclide (maximum equal to 75 Bq/kg) and absence of ^{241}Am (<1 Bq/kg). The following areas of localized contamination were the exception:

- area #1: ^{241}Am – $2.0 \cdot 10^4$ Bq/kg, ^{137}Cs – $1.2 \cdot 10^3$ Bq/kg, ^{152}Eu – 32 Bq/kg, ^{60}Co – 5 Bq/kg, ^{90}Sr – $7.7 \cdot 10^2$ Bq/kg;
- area #2 (contamination was found during the preliminary survey): ^{241}Am – $6.0 \cdot 10^2$ Bq/kg, ^{137}Cs – $1.1 \cdot 10^4$ Bq/kg, ^{152}Eu – $2.5 \cdot 10^2$ Bq/kg, ^{154}Eu – 19 Bq/kg, ^{60}Co – 45 Bq/kg, and ^{90}Sr – $4 \cdot 10^2$ Bq/kg.

Expected concentration of $^{239+240}$Pu in the first area is about $2.0 \cdot 10^5$ Bq/kg. Thus, soil of the area #1 can be classified as the medium-activity radioactive waste [5]. Expected content of $^{239+240}$Pu in the area #2 is $6.0 \cdot 10^3$ Bq/kg, which is classified as the low-activity radioactive waste.

Specific activities of ^{90}Sr and ^{137}Cs in soils sampled from the H4 tunnel mouth area did not exceed the MSSA standard levels. However, presence of ^{241}Am was recorded in 45% of points in this tunnel, mostly in the backfill and pile areas. The maximum measured value was $1.1 \cdot 10^3$ Bq/kg (backfill), which was 1.1 times higher than MSSA for this radionuclide. Presence of man-caused contamination was also registered in a localized dumped area, which was found during the preliminary survey ($2.7 \cdot 10^2$ Bq/kg for ^{241}Am). This contamination could be caused by dumping soil from a man-disturbed hillside onto contaminated areas, while the dumped soil could contain transuranium radionuclides in deeper subsurface layers. Field gamma-ray spectrometry was conducted during the period of remedial actions, but no deeper man-caused contamination was detected, which was found only after performing laboratory analyses of crushed rocks sampled during the final survey.

Maximum specific activity of ^{90}Sr measured at the V1 tunnel work site did not exceed the standard level equal to $1 \cdot 10^5$ Bq/kg. As to ^{137}Cs, the level of $1 \cdot 10^4$ Bq/kg was exceeded in the points 2 and 3 ($3.9 \cdot 10^4$ Bq/kg and $1.1 \cdot 10^4$ Bq/kg, respectively). This excess is explained by redistribution of contaminated soil from the work site and migration of radionuclide contamination from the tunnel.

Upon completion of works in the surveyed area, ^{241}Am concentrations did not exceed the standard level of $1 \cdot 10^3$ Bq/kg stipulated by NRB-99. The only exception was Point 2 ($2.7 \cdot 10^3$ Bq/kg), located within the work site.

Estimated data on Plutonium and radiochemical analysis of the samples indicate that $^{239+240}$Pu concentrations in the points 2, 3 and 5 can be 40 times higher than the standard MSSA level of $1 \cdot 10^3$ Bq/kg ($4.4 \cdot 10^4$ Bq/kg for Point 2).

In the course of the engineering works at the V1 tunnel, radionuclide contamination was most likely moved out of the tunnel and got into the area close to the tunnel mouth. However, containment actions were taken, which prevented contamination from spreading into the surrounding areas.

The V2 tunnel mouth area remained the most radiation-hazardous after completion of the final works.

According to the laboratory tests, about 83% of all analyzed soil samples can be classified as radioactive wastes based on the fact that $^{239+240}$Pu and ^{241}Am concentrations are in excess of MSSA ($1 \cdot 10^3$Bq/kg) for these radionuclides. ^{241}Am specific activity varies in the range from <3 to $4.1 \cdot 10^4$ Bq/kg, with maximum exceeding being 43-fold MSSA. Expected concentration of $^{239+240}$Pu in almost all points can exceed the standard MSSA level of $1 \cdot 10^3$ Bq/kg for ^{239,240}Pu. Thus, maximum possible specific activity of $^{239+240}$Pu can reach $4.5 \cdot 10^5$ Bq/kg.

In addition, a survey of field camp area was carried out, and 2 spots of localized radioactive contamination were found. Both spots were not detected during the preliminary survey, as they are quite small (≈1 m^2) and are situated outside of the preliminary radioecological survey coverage and outside of the operations site. The contaminated spots are located ≈30 meters away from the accommodation blocks; these contaminations are crushed stones dumped on the rocky surface. The Max point represents a radiation hazard. In this point, concentration of ^{137}Cs is $1.6 \cdot 10^5$ Bq/kg and that of ^{90}Sr is $2.9 \cdot 10^5$ Bq/kg; these values exceed MSSA (10^4 and 10^5 Bq/kg, respectively). Since no works were carried out in this area, contamination was not connected with activities carried out at the V2 facility.

Though the laboratory tests indicated that ^{241}Am and $^{239+240}Pu$ specific activities went down upon completion of decontamination works in the V2 tunnel area, residual concentrations of these radionuclides significantly exceed the allowable levels (MSSA).

5.3. Bottom layer of the atmosphere contaminated with radionuclides

The preliminary survey did not detect any significant volumetric activities of radionuclides in the bottom layer of the atmosphere, despite the fact that radionuclide concentrations in soil were quite high. Therefore, there was no need to study the bottom layer of the atmosphere during the final survey.

5.4. Findings of the final survey of tunnel near-portal areas

The final survey indicated that the highest radiation parameters were registered in the V1, H1 and H2 tunnel mouth areas. Upon completion of resealing/supplementary protection operations, radiation parameters in all the tunnel near-portal areas were within the allowable limits for Category A personnel [4].

Maximum specific activities of radionuclides were registered during the final stage of resealing operations within several localized contamination spots located in the near-portal areas of the following tunnels: V1 ($3.9 \cdot 10^4$ Bq/kg for ^{137}Cs and $3{,}8 \cdot 10^4$ Bq/kg for ^{90}Sr) and V2 ($4.1 \cdot 10^4$ Bq/kg for ^{241}Am and $4.5 \cdot 10^5$ Bq/kg for $^{239+240}Pu$). Maximum observed specific activities of ^{137}Cs, ^{241}Am and $^{239+240}Pu$ exceed the standard levels (MSSA, NRB-99), equal to $1 \cdot 10^4$, $1 \cdot 10^3$ and $1 \cdot 10^3$ Bq/kg, respectively, while the highest concentration of ^{90}Sr does not exceed the MSSA of $1 \cdot 10^5$ Bq/kg.

^{137}Cs exceeded the MSSA in 3 tunnel mouth areas, while ^{241}Am and $^{239+240}Pu$ transuranium radionuclides, in 6 mouth areas. Similar to the preliminary survey, contaminations were found not only in the piles/stacks and backfills, but also in localized spots cause by unauthorized activities undertaken near the tunnel mouths.

6. IMPACT ASSESSMENT OF PERFORMED WORKS ON RADIOLOGICAL CONDITIONS OF TUNNEL MOUTH AREAS

6.1. Change in radiological conditions estimated per radiometric and dosimeter measurements

The EDR maximum values observed during the preliminary and final surveys of the tunnels are represented in the diagram below (Figure 49).

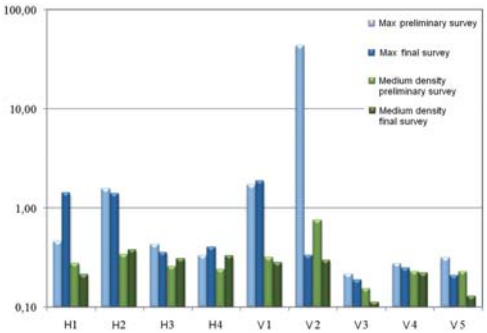


Figure 49. Maximum values and average density of EDR for the period of preliminary and final surveys of the tunnels, *μSv/h*

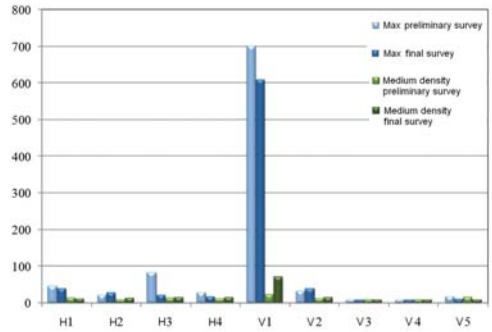


Figure 50. Maximum values and average density of β-particle flux density for the period of preliminary and final survey of the tunnels, *particles/(min*cm²)*

As the diagram shows, maximum values of EDR were recorded during the preliminary survey of the localized contaminations of tunnels H2, V1, and V2. Maximum value of EDR was recorded at the V2 tunnel (44 μSv/h). The radiation safety norms NRB-99 stipulate that in the event any full-time works are to be performed under the conditions similar to the described above, then the effective dose for personnel can be exceeded ~4 times only from external sources of radiation [4]. Maximum values of EDR during the final survey were recorded in the localized contamination parts of the tunnels H1, H2, and V1.

It is worth noting that upon completion of works at the tunnels V2 and V5, maximum EDR values decreased. Therefore, it can be inferred the performed works led to decrease in the levels of radioactive contamination at the near-portal areas of these tunnels. During the final survey of the H1 and V1 tunnels, EDR values were higher than those of the preliminary survey. Therefore, one can assume that the performed works had a negative impact on the radiological conditions of these mouth tunnel areas.

However, maximum EDR values do not fully characterize the radiological conditions at the tunnel mouths, as they do not take into account the total area of contamination. Calculation of the average density of contamination was made in order to assess the impact of the contaminated area.

Acreages of each mouth area were estimated in order to calculate average density of contamination. Based on the radiation parameter distributions, the key isolines were determined for each tunnel depending on the range of EDR values. In order to calculate the average density of contamination, the upper values of EDR range were taken, and areas for each isoline were calculated. Contribution of various EDR values was estimated based on the ratio of the area characterized by the surveyed site-specific values to the total area of the site. Results of the average contamination density calculations for the near-portal areas based on the preliminary and final survey data are given in the table and diagram below (Table 2, Figure 49).

Having compared the changes in maximum EDR values before and after resealing, as well as calculation results for the average contamination density, we subdivide the tunnels into the following groups:

1. tunnels, where changes in EDR values and average densities are within the accuracy limits (H2, H3, H4, V3, and V4); therefore, the performed works did not influence the radiological conditions of the mouth areas;
2. tunnels, where maximum EDR values and contamination density decreased (V2 and V5) as a result of decontamination activities undertaken prior to the works; and
3. tunnels, where an increase in maximum EDR values was accompanied by insignificant change in the average density (within its accuracy limits) or tunnels H1 and V1. Contamination here has a localized character and could be caused by contaminated soil moved out from the tunnel (H1) or by nearby objects having increased radiation hazard (V1), i.e. piles, brook area, water inflow from the tunnel.

Data on maximum and minimum values of β-particle flux density measured during the preliminary and final surveys of tunnels are given in Figure 50.

Maximum values of β-particle flux density were recorded during the preliminary and final surveys of the localized contamination part of the V1 tunnel (preliminary survey showed 700 particles/(min*cm^2), whereas the final survey gave 610 particles/(min*cm^2)). Tentatively, we can say that the performed works brought about a decrease in radiation contamination of the H3, H4, V1 and V5 tunnel mouth areas. Since measured values of β-particle flux density did not exceed the detection limit of the used measuring devices both during the preliminary and final surveys of the V3 and V4 tunnel areas, it can be inferred that the performed works did not influence the radiological conditions.

In addition, based on the β-particle flux density measurements taken during the preliminary and final surveys, average contamination densities were calculated for the near-portal areas (Table 2, Figure 50).

Average contamination densities calculated for the near-portal areas from the measurements of β-particle flux density indicate that the radiological conditions have deteriorated on the V1 tunnel after the works completion.

Having compared changes in the maximum values of β-particle flux density before and after the resealing works, as well as calculation results for average contamination densities, we subdivided the tunnels into the following groups:

1. tunnels, where changes in β-particle flux density values and average densities were within the accuracy limits (H1, H2, V2, V3, and V4); therefore, the performed works did not influence the radiological conditions of the mouth areas;
2. tunnels, where maximum β-particle flux densities decreased and average contamination densities changed insignificantly, within the accuracy limits (tunnels H3, H4 and V5). These changes could have resulted from decontamination activities undertaken prior to beginning of works. Insignificant changes in the average density indicate that the maximum values of β-particle flux density, recorded during the preliminary survey, were related to small-size localized contaminations, which insignificantly contributed into the overall average density;
3. tunnel V1, which is characterized by an increased average contamination density with insignificant changes in β-particle flux density (within the accuracy limits). Here, contamination was caused by redistribution/transport of contaminated soils

from highly hazardous objects, and such soils were spread throughout the entire area in the course of the works;

Based the analysis, the following conclusions were made:

1. H1: contamination of the area was caused by the tunnel resealing operations/ works; this localized contamination arose by moving soils with increased content of γ-emitting radionuclides.
2. H2, V3, and V4: resealing operations did not influence the radiological conditions of the tunnel area.
3. H3, H4, V2, and V5: decontamination activities, undertaken over the near-portal areas prior to the tunnel resealing operations, improved the conditions in the tunnel mouth areas.
4. V1: contamination of the area resulted from the tunnel resealing operations; it had a localized character and was caused by nearby piles with increased concentrations of γ-emitting radionuclides; redistribution was also recorded of the then-existing β-emitting radionuclides over the mouth area.

6.2. Changes in radionuclide contamination of topsoil

To assess changes in radioecological conditions of the investigated areas, it is worthwhile to conduct a comparative analysis of changes in maximum specific activities of artificial radionuclides, recorded during the preliminary and final stages of the work (Figure 51).

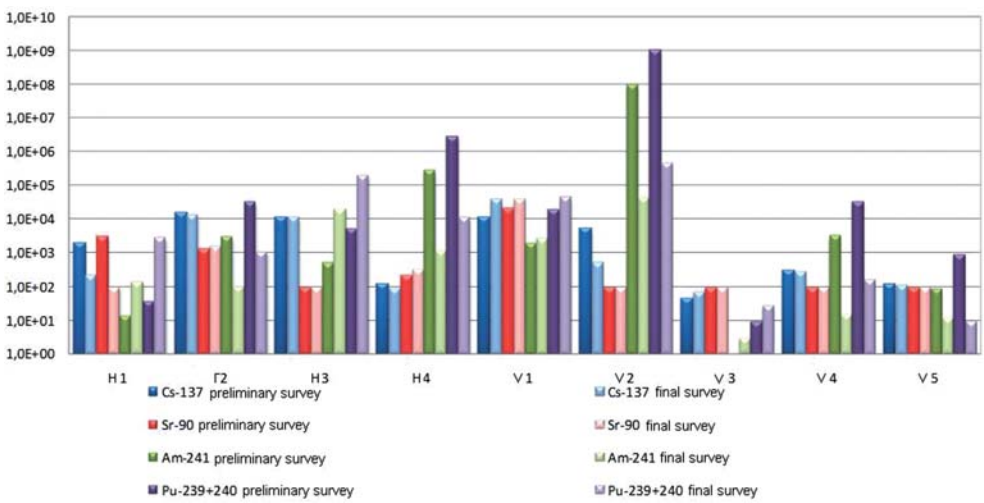


Figure 51. Comparison of maximum specific activities of radionuclides obtained during the preliminary and final surveys of the mouth areas, *Bq/kg*

Decreased maximum specific activities of radionuclides were observed in the H2, H4, V2, V4 and V5 tunnel mouth areas, which evidenced that the radioecological conditions these areas had improved. Three tunnels (H3, V1 and V3) exhibited elevated maximum values of radionuclide activities, with maximums of the preliminary and final surveys over the V3 tunnel mouth area being within the accuracy limits of the radionuclide measurement

method. A decrease in maximum specific activities of ^{137}Cs and ^{90}Sr, observed for the H1 tunnel, was accompanied by an increase in concentrations of ^{241}Am and $^{239+240}$Pu transuranic radionuclides.

An increase in maximum specific activities of radionuclides, observed in the tunnel mouth areas, can be explained by redistribution/movement of artificial radionuclides over the site, which happened after the preliminary survey. This increase could also be caused by the engineering works performed in the tunnel, while contamination of the surrounding territory could be a result of the secondary transport.

Based on the laboratory tests of each investigated tunnel, average specific activities were calculated for ^{137}Cs, ^{90}Sr, ^{241}Am, and ^{239,240}Pu radionuclides. Average specific activities were calculated for each radionuclide found in each investigated tunnel mouth area with the account of all sampling points in each respective mouth area. Number of points investigated in each mouth area varied in the range from 17 to 42, depending on the landscape, level of contamination, and type of the works performed. Table 2 gives the comparative analysis results for average concentrations of artificial radionuclides in soils sampled during the preliminary and final surveys of the tunnels.

The final survey data were used to calculate average specific activities of radionuclides in soil. The calculations show that radiological conditions have improved and average specific activities of artificial radionuclides have significantly decreased throughout the entire surveyed site. In many cases, specific activities of these radionuclides decreased upon completing works aimed at mitigating radiation hazards of the contaminated radioactive areas (H2, H4, V2, V4, and V5).

However, the H1, H3, V1 and V3 tunnel near-portal areas exhibited higher specific activities during the final survey as compared with those obtained during the preliminary survey. These higher average values can be explained by redistribution of radionuclides and by engineering works carried out on the tunnels.

Areal contamination levels have been also calculated, since it is impossible to fully assess radiological conditions of the tunnel near-portal areas using just maximum and average values. For H2, V2 and V4 tunnels, the soil sampling patterns covered the entire area of each mouth. For this reason, schematic maps of ^{137}Cs and ^{241}Am radionuclide distributions were built only for these tunnels and areal contamination levels were calculated using isolines, i.e. directly based on the gamma-ray spectrometry data. Areal contamination levels of the H2, V2 and V4 tunnels are given in Table 2.

For the H2 tunnel, total acreage of the spots containing ^{137}Cs in concentrations more than 10^4 Bq/kg decreased from 76 m^2 to 55 m^2, but values of the average areal contamination with ^{137}Cs in this tunnel mouth area are the highest of the three tunnels mentioned in the paragraph above. These spots are in the piles located 100 m away from the tunnel mouth.

The V2 tunnel mouth area was found to be the most heavily contaminated with ^{241}Am, as inferred from the analysis of maximum and average specific activities of this radionuclide and based on calculations of areal contamination levels. Works carried out to build supplementary protection for this tunnel did not exacerbate the situation at the tunnel mouth area, while the decontamination measures helped to significantly reduce the quantity of day-surface radionuclides. There was a 2-fold decrease (from $3.5 \cdot 10^3$ m^2 to $1.5 \cdot 10^3$ m^2) in the total area contaminated with ^{241}Am in concentrations more than 10^3 Bq/kg.

It was not possible to map soil contaminations with ^{137}Cs, ^{241}Am, ^{90}Sr and $^{239+240}Pu$ for the rest of the tunnels because of the numbers of sampling points were insufficient.

This comparative analysis based on calculating areal contamination levels confirms the trend of maximum and average specific activities of radionuclides. In some cases, it is more a more conservative approach that helps us to avoid any underestimation of the risks for personnel and communities.

The preliminary and final survey data have been compared, changes in maximum and average specific activities of artificial radionuclides have been analyzed, and values of average areal contamination have been calculated. Based on changes in radiological conditions in the mouth areas, tunnels can be subdivided into the following groups:

1. tunnels, where changes in maximum and average specific activities of radionuclides, as well as average densities were within the accuracy limits (H1, H2, V2, V4, and V5); therefore, the performed works did not influence the radiological conditions of the mouth areas;
2. tunnels, where, based on all used evaluation criteria, ^{137}Cs and ^{90}Sr concentrations went down, while concentrations of transuranium radionuclides went up (tunnels H1and H2). These changes could have resulted from decontamination activities undertaken prior to beginning of works; and
3. tunnels, which are characterized by elevated maximum and average specific activities of ^{137}Cs, ^{241}Am, ^{90}Sr, and $^{239+240}Pu$ radionuclides (tunnels V1, V3). The site contamination was brought about by contaminated soils moved out of the tunnels or redistributed over the site in the course of the works, which caused a 2-fold increase in radionuclide concentrations.

Table 2.

Summary table of radiation parameters obtained for degelen tunnel near-portal areas during the 2006-2009 preliminary and final surveys

Parameter		H1	H2	H3	H4	V1	V2	V3	V4	V5
EDR, μSv/h	Preliminary surveying	0.47±0.09	1.60±0.32	0.44±0.09	0.34±0.07	1.75±0.31	44.00±8.80	0.22±0.04	0.28±0.06	0.32±0.06
	Final surveying	1.43±0.29	1.42±0.28	0.36±0.07	0.40±0.08	1.90±0.38	0.34±0.07	0.19±0.04	0.25±0.05	0.21±0.04
Average contamination density based on EDR measurements, μSv/h	Preliminary surveying	0.28±0.08	0.34±0.10	0.26±0.08	0.24±0.07	0.32±0.10	0.76±0.23	0.16±0.05	0.23±0.07	0.23±0.07
	Final surveying	0.21±0.06	0.38±0.11	0.31±0.09	0.33±0.10	0.28±0.08	0.30±0.09	0.11±0.03	0.22±0.07	0.13±0.04
Maximal beta-flux density, particles/(min*cm^2)	Preliminary surveying	48±10	22±4	85±17	30±6	700±140	35±7	10±2	10±2	18±4
	Final surveying	41±8	30±6	22±4	18±4	610±122	40±8	10±2	10±2	11±2
Average density of contamination based on beta-flux density measurements, particles/(min*cm^2)	Preliminary surveying	14±4	10±3	15±5	11±3	22±7	13±4	10±3	10±3	16±5
	Final surveying	11±3	15±5	17±5	16±5	71±21	16±5	10±3	10±3	10±3
Maximal alpha-flux density, particles/(min*cm^2)	Preliminary surveying	<0.2	<0.2	<0.2	140±30	<0.2	100±20	<0.2	<0.2	<0.2
	Final surveying	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
Maximal volume activity in air at preliminary surveying, Bq/m^3	^{137}Cs	$4\cdot10^{-4}\pm 1\cdot10^{-4}$	$< 8\cdot10^{-2}$	$2\cdot10^{-3}\pm 2\cdot10^{-4}$	$<3\cdot10^{-3}$	$5\cdot10^{-4}\pm 9\cdot10^{-5}$	$< 1\cdot10^{-1}$	$5\cdot10^{-4}\pm 2\cdot10^{-4}$	$< 6\cdot10^{-2}$	$1.7\cdot10^{-4}\pm 5.5\cdot10^{-5}$
	^{241}Am	$<9\cdot10^{-5}$	$< 4\cdot10^{-2}$	$< 2\cdot10^{-2}$	$< 2\cdot10^{-3}$	$<9\cdot10^{-5}$	$< 6\cdot10^{-2}$	$<2\cdot10^{-4}$	$< 4\cdot10^{-2}$	$< 9.25\cdot10^{-5}$

Parameter			H1	H2	H3	H4	V1	V2	V3	V4	V5
Maximal specific activity, Bq/kg	^{137}Cs	Preliminary surveying	$1.9 \cdot 10^{3} \pm 0.4 \cdot 10^{3}$	$1.5 \cdot 10^{4} \pm 0.4 \cdot 10^{3}$	$1.1 \cdot 10^{4} \pm 0.2 \cdot 10^{4}$	$1.2 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$	$1.1 \cdot 10^{4} \pm 0.2 \cdot 10^{4}$	$5.3 \cdot 10^{3} \pm 1.1 \cdot 10^{3}$	$4.5 \cdot 10^{1} \pm 0.9 \cdot 10^{1}$	$3.0 \cdot 10^{2} \pm 0.6 \cdot 10^{2}$	$1.2 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$
		Final surveying	$2.3 \cdot 10^{2} \pm 0.5 \cdot 10^{2}$	$1.4 \cdot 10^{4} \pm 0.4 \cdot 10^{4}$	$1.1 \cdot 10^{4} \pm 0.2 \cdot 10^{4}$	$9.0 \cdot 10^{1} \pm 1.8 \cdot 10^{1}$	$3.9 \cdot 10^{4} \pm 0.8 \cdot 10^{4}$	$5.1 \cdot 10^{2} \pm 1.8 \cdot 10^{2}$	$7.2 \cdot 10^{1} \pm 1.4 \cdot 10^{1}$	$2.9 \cdot 10^{2} \pm 0.6 \cdot 10^{2}$	$1.1 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$
	^{90}Sr	Preliminary surveying	$3.1 \cdot 10^{3} \pm 0.6 \cdot 10^{3}$	$1.4 \cdot 10^{3} \pm 0.4 \cdot 10^{3}$	$1.5 \cdot 10^{4} \pm 0.3 \cdot 10^{4}$	$2.3 \cdot 10^{2} \pm 0.5 \cdot 10^{2}$	$2.2 \cdot 10^{4} \pm 0.4 \cdot 10^{4}$	$<1.0 \cdot 10^{2}$	$<1.0 \cdot 10^{2}$	$<1.0 \cdot 10^{2}$	$<1.0 \cdot 10^{2}$
		Final surveying	$<1.0 \cdot 10^{2}$	$1.6 \cdot 10^{3} \pm 0.4 \cdot 10^{3}$	$4.1 \cdot 10^{3} \pm 0.8 \cdot 10^{3}$	$3.2 \cdot 10^{2} \pm 0.5 \cdot 10^{2}$	$3.8 \cdot 10^{4} \pm 0.8 \cdot 10^{4}$	$<1.0 \cdot 10^{2}$	$<1.0 \cdot 10^{2}$	$<1.0 \cdot 10^{2}$	$<1.0 \cdot 10^{2}$
	^{241}Am	Preliminary surveying	$1.4 \cdot 10^{1} \pm 0.3 \cdot 10^{1}$	$3.0 \cdot 10^{3} \pm 0.6 \cdot 10^{3}$	$5.4 \cdot 10^{2} \pm 1.1 \cdot 10^{2}$	$2.8 \cdot 10^{5} \pm 0.6 \cdot 10^{5}$	$1.9 \cdot 10^{3} \pm 0.4 \cdot 10^{3}$	$9.6 \cdot 10^{7} \pm 1.9 \cdot 10^{7}$	<1.0	$3.3 \cdot 10^{3} \pm 0.6 \cdot 10^{3}$	$9.0 \cdot 10^{1} \pm 1.4 \cdot 10^{0}$
		Final surveying	$1.5 \cdot 10^{2} \pm 0.3 \cdot 10^{2}$	$1.1 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$	$2.0 \cdot 10^{4} \pm 0.4 \cdot 10^{4}$	$1.1 \cdot 10^{3} \pm 0.2 \cdot 10^{3}$	$2.7 \cdot 10^{3} \pm 0.5 \cdot 10^{3}$	$4.1 \cdot 10^{4} \pm 0.8 \cdot 10^{4}$	$3.0 \cdot 10^{0} \pm 0.6 \cdot 10^{0}$	$1.5 \cdot 10^{1} \pm 0.3 \cdot 10^{1}$	$1.2 \cdot 10^{1} \pm 0.2 \cdot 10^{1}$
	$^{239+240}$Pu	Preliminary surveying	$3.8 \cdot 10^{1} \pm 0.8 \cdot 10^{1}$	$3.3 \cdot 10^{4} \pm 0.7 \cdot 10^{4}$	$5.4 \cdot 10^{3} \pm 1.1 \cdot 10^{3}$	$2.8 \cdot 10^{6} \pm 0.6 \cdot 10^{6}$	$2.0 \cdot 10^{4} \pm 0.4 \cdot 10^{4}$	$1.1 \cdot 10^{9} \pm 0.2 \cdot 10^{9}$	$1.1 \cdot 10^{1} \pm 0.2 \cdot 10^{1}$	$3.3 \cdot 10^{4} \pm 0.7 \cdot 10^{4}$	$9.0 \cdot 10^{2} \pm 0.2 \cdot 10^{1}$
		Final surveying	$2.8 \cdot 10^{3} \pm 0.6 \cdot 10^{3}$	$1.1 \cdot 10^{3} \pm 0.2 \cdot 10^{3}$	$2.0 \cdot 10^{5} \pm 0.4 \cdot 10^{5}$	$1.1 \cdot 10^{4} \pm 0.2 \cdot 10^{4}$	$4.4 \cdot 10^{4} \pm 0.9 \cdot 10^{4}$	$4.5 \cdot 10^{5} \pm 0.9 \cdot 10^{5}$	$2.8 \cdot 10^{1} \pm 0.6 \cdot 10^{1}$	$1.7 \cdot 10^{2} \pm 0.3 \cdot 10^{2}$	$1.0 \cdot 10^{1} \pm 0.2 \cdot 10^{1}$
Average specific activity, Bq/kg	^{137}Cs	Preliminary surveying	$2.2 \cdot 10^{2} \pm 0.4 \cdot 10^{2}$	$1.9 \cdot 10^{3} \pm 0.4 \cdot 10^{3}$	$4.9 \cdot 10^{2} \pm 1.0 \cdot 10^{2}$	$2.2 \cdot 10^{1} \pm 0.4 \cdot 10^{1}$	$1.6 \cdot 10^{3} \pm 0.2 \cdot 10^{3}$	$6.0 \cdot 10^{2} \pm 1.2 \cdot 10^{2}$	$1.3 \cdot 10^{1} \pm 0.3 \cdot 10^{1}$	$7.9 \cdot 10^{1} \pm 1.6 \cdot 10^{1}$	$2.5 \cdot 10^{1} \pm 0.5 \cdot 10^{1}$
		Final surveying	$3.9 \cdot 10^{1} \pm 0.8 \cdot 10^{1}$	$1.5 \cdot 10^{3} \pm 0.3 \cdot 10^{3}$	$4.4 \cdot 10^{2} \pm 0.9 \cdot 10^{2}$	$2.2 \cdot 10^{1} \pm 0.4 \cdot 10^{1}$	$3.2 \cdot 10^{3} \pm 0.6 \cdot 10^{3}$	$1.5 \cdot 10^{2} \pm 0.3 \cdot 10^{2}$	$2.9 \cdot 10^{1} \pm 0.6 \cdot 10^{1}$	$5.7 \cdot 10^{1} \pm 1.2 \cdot 10^{1}$	$2.3 \cdot 10^{1} \pm 0.5 \cdot 10^{1}$
	^{90}Sr	Preliminary surveying	$9.2 \cdot 10^{2} \pm 1.8 \cdot 10^{2}$	$2.1 \cdot 10^{2} \pm 0.4 \cdot 10^{2}$	$6.3 \cdot 10^{2} \pm 1.3 \cdot 10^{2}$	$1.8 \cdot 10^{2} \pm 0.3 \cdot 10^{2}$	$3.7 \cdot 10^{3} \pm 0.7 \cdot 10^{3}$	$1.0 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$	$1.0 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$	$1.0 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$	$1.0 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$
		Final surveying	$1.0 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$	$2.2 \cdot 10^{2} \pm 0.4 \cdot 10^{2}$	$2.6 \cdot 10^{2} \pm 0.5 \cdot 10^{2}$	$1.1 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$	$6.6 \cdot 10^{3} \pm 1.3 \cdot 10^{3}$	$1.0 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$	$1.0 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$	$1.0 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$	$1.0 \cdot 10^{2} \pm 0.2 \cdot 10^{2}$
	^{241}Am	Preliminary surveying	$9.3 \cdot 10^{0} \pm 1.9 \cdot 10^{0}$	$1.5 \cdot 10^{2} \pm 0.3 \cdot 10^{2}$	$2.2 \cdot 10^{1} \pm 0.5 \cdot 10^{1}$	$1.2 \cdot 10^{4} \pm 0.2 \cdot 10^{4}$	$2.0 \cdot 10^{2} \pm 0.4 \cdot 10^{2}$	$2.7 \cdot 10^{6} \pm 0.5 \cdot 10^{6}$	$1.0 \cdot 10^{0} \pm 0.2 \cdot 10^{0}$	$4.5 \cdot 10^{2} \pm 0.9 \cdot 10^{2}$	$3.0 \cdot 10^{0} \pm 0.2 \cdot 10^{0}$
		Final surveying	$3.4 \cdot 10^{1} \pm 0.7 \cdot 10^{1}$	$2.5 \cdot 10^{1} \pm 0.5 \cdot 10^{1}$	$2.2 \cdot 10^{1} \pm 0.5 \cdot 10^{1}$	$1.4 \cdot 10^{2} \pm 0.3 \cdot 10^{2}$	$2.3 \cdot 10^{2} \pm 0.5 \cdot 10^{2}$	$3.8 \cdot 10^{3} \pm 0.8 \cdot 10^{3}$	$1.0 \cdot 10^{0} \pm 0.2 \cdot 10^{0}$	$1.0 \cdot 10^{0} \pm 0.2 \cdot 10^{0}$	$1.0 \cdot 10^{0} \pm 0.2 \cdot 10^{0}$
	$^{239+240}$Pu	Preliminary surveying	$1.5 \cdot 10^{1} \pm 0.3 \cdot 10^{1}$	$1.6 \cdot 10^{3} \pm 0.3 \cdot 10^{3}$	$2.2 \cdot 10^{2} \pm 0.5 \cdot 10^{2}$	$1.2 \cdot 10^{5} \pm 0.2 \cdot 10^{5}$	$2.0 \cdot 10^{3} \pm 0.4 \cdot 10^{3}$	$3.0 \cdot 10^{7} \pm 0.6 \cdot 10^{7}$	$1.0 \cdot 10^{1} \pm 0.2 \cdot 10^{1}$	$4.4 \cdot 10^{3} \pm 0.9 \cdot 10^{3}$	$3.2 \cdot 10^{1} \pm 1.8 \cdot 10^{0}$
		Final surveying	$4.2 \cdot 10^{2} \pm 0.8 \cdot 10^{2}$	$2.5 \cdot 10^{2} \pm 0.5 \cdot 10^{2}$	$7.3 \cdot 10^{3} \pm 1.5 \cdot 10^{3}$	$1.4 \cdot 10^{3} \pm 0.3 \cdot 10^{3}$	$3.4 \cdot 10^{3} \pm 0.7 \cdot 10^{3}$	$4.2 \cdot 10^{4} \pm 0.9 \cdot 10^{4}$	$1.3 \cdot 10^{1} \pm 0.3 \cdot 10^{1}$	$8.7 \cdot 10^{0} \pm 1.7 \cdot 10^{0}$	$9.0 \cdot 10^{0} \pm 1.8 \cdot 10^{0}$

Parameter			H1	H2	H3	H4	V1	V2	V3	V4	V5
Average areal contamination, Bq/m^2	^{137}Cs	Preliminary surveying		$2.4 \cdot 10^4 \pm 0.5 \cdot 10^4$				$2.3 \cdot 10^3 \pm 0.5 \cdot 10^3$		$2.0 \cdot 10^3 \pm 0.4 \cdot 10^3$	
		Final surveying		$2.0 \cdot 10^4 \pm 0.4 \cdot 10^4$				$3.1 \cdot 10^3 \pm 0.6 \cdot 10^3$		$1.4 \cdot 10^3 \pm 0.3 \cdot 10^3$	
	^{241}Am	Preliminary surveying		$7.8 \cdot 10^2 \pm 1.6 \cdot 10^2$				$1.2 \cdot 10^5 \pm 0.2 \cdot 10^5$		$8.5 \cdot 10^3 \pm 1.7 \cdot 10^3$	
		Final surveying		$4.9 \cdot 10^2 \pm 1.0 \cdot 10^2$				$5.0 \cdot 10^4 \pm 1.0 \cdot 10^4$		$4.0 \cdot 10^1 \pm 0.8 \cdot 10^1$	

CONCLUSION

The radioecological survey data were analyzed, with the data obtained for Degelen tunnel near-portal areas before and after the works undertaken to build supplementary protection of engineering facilities; and an assessment was made of the effect of such works on radiological conditions of the tunnel mouth areas.

Based on the changes in the field and laboratory data obtained for the surveyed tunnel mouth areas, the tunnels can be subdivided into the following groups characterized by area-specific radiological conditions:

1. H2, H4, V2, V4, and V5: resealing operations did not affect the radiological conditions or the conditions improved due to decontamination measures undertaken within the highly contaminated spots found during the preliminary surveying;
2. H1, H3, and V3: insignificant transuranium radionuclide contamination levels were detected in the mouth areas, with these levels being below MSSA for these radionuclides; these contaminations by moving out or redistributing radionuclides in the course of the works on the tunnels (except the tunnel H3, where a localized increase – from $5.4 \cdot 10^2$ to $2.0 \cdot 10^4$ Bq/kg – in transuranium radionuclide concentrations was observed, which did not cause any changes in the average activities of the area). V3 contamination values were within the accuracy limits; and
3. V1: contamination of the area resulted from the tunnel resealing operations; it had a localized character and was caused by nearby piles with increased concentrations of radionuclides.

In general, the approved action plan was implemented and in many cases, it brought about improved radioecological conditions on the tunnel mouth areas. In some cases, contamination of the areas was caused by the presence of objects with high concentrations of artificial radionuclides. For example, several spots with very high radiometric parameters were detected in the V1 tunnel area; these spots were located in the dry brook near the tunnel mouth. The acreage of all the detected spots made 12% of the total surveyed acreage. In addition, some of these spots were located along the access ways; and clayey soils were moved around the site by motor vehicles.

Similar radiological monitoring plans will be used in the future. However, when designing similar operations in the future, it will be required to compile more site-specific monitoring plans for any tunnel operations as radioecological conditions can be negatively impacted by such operations (when using the horizontal method or when objects with high radiological hazards are located nearby).

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ДЕГЕЛЕҢ ТАУЛЫ МАССИВІНДЕГІ ШТОЛЬНЯЛАРДЫҢ ИНЖЕНЕРЛІК ҚҰРЫЛЫСТАРЫНА ҚОСЫМША ҚОРҒАУ ҚҰРУ БОЙЫНША ЖҮРГІЗІЛГЕН ЖҰМЫСТАРДЫҢ ПОРТАЛАЛДЫ ТЕЛІМДЕРДІҢ РАДИАЦИЯЛЫҚ ЖАҒДАЙЫНА ӘСЕРІ

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Ядролық қаруды тарату қауіпін азайту мақсатындағы ядролық сынақтардың инфрақұрылымын жою бойынша жұмыстар аясында Семей сынақ полигоны Дегелең таулы массивінің 9 штольнясын қайтадан консервілеу жүргізілді. Қоршаған орта нысандарының ахуалын бағалау, сондай-ақ аталған штольнялардың порталалды телімдеріндегі жүргізілген жұмыстардың кері әсерінің болу мүмкіндігін бағалау аталған радиоэкологиялық зерттеулер мақсаты болып табылады. Бақылаудағы әр штольняның порталалды телімдеріндегі радиоэкологиялық жағдайдың нәтижелері келтірілген.

Түйін сөздер: Семей сынақ полигоны, Дегелең таулы массиві, штольнялар, инженерлік құрылыстарға қосымша қорғау, радиоэкологиялық зерттеу, радиациялық жағдай, эквиваленттік доза қуаты, α-, β-бөлшектер ағынының тығыздығы, радионуклид, тиесілі белсенділік, алаңдық ластану

ВЛИЯНИЕ ПРОВЕДЕННЫХ РАБОТ ПО СОЗДАНИЮ ДОПОЛНИТЕЛЬНОЙ ЗАЩИТЫ ИНЖЕНЕРНЫХ СООРУЖЕНИЙ ШТОЛЕН ГОРНОГО МАССИВА ДЕГЕЛЕН НА РАДИАЦИОННУЮ ОБСТАНОВКУ ПРИПОРТАЛЬНЫХ УЧАСТКОВ

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В рамках работ по ликвидации инфраструктуры ядерных испытаний в целях уменьшения угрозы распространения ядерного оружия было проведено повторное консервирование 9 штолен горного массива Дегелен Семипалатинского испытательного полигона. Целью данных радиоэкологических исследований являлась оценка состояния объектов окружающей среды, а также возможного негативного влияния проведенных работ на припортальных участках данных штолен. Представлены результаты изменения радиоэкологической обстановки на припортальных участках каждой контролируемой штольни.

Ключевые слова: Семипалатинский испытательный полигон, горный массив Дегелен, штольни, дополнительная защита инженерных сооружений, радиоэкологическое обследование, радиационная обстановка, мощность эквивалентной дозы, плотность потока α-, β-частиц, радионуклид, удельная активность, площадное загрязнение.

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STUDY OF ACTIVITY RATIO OF ^{241}PU AND ^{241}AM IN SOIL OF MAJOR TEST GROUNDS OF STS

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A method for determining ^{241}Pu in soil of major test grounds at Semipalatinsk test site has been developed. Activities of ^{241}Am and ^{241}Pu in soil samples taken from the major testing grounds of STS have been determined. The ratio of ^{241}Pu to ^{241}Am was calculated.

In most cases at the local grounds the ratio of ^{241}Pu/^{241}Am lies in the fairly narrow range, which makes it possible to assess the activity of ^{241}Pu by the results of gamma-spectrometric analysis of ^{241}Am. The accuracy of assessing ^{241}Pu activity with this method is about 50% which is sufficient in most of the cases.

Key words: nuclear tests, monitoring, radioecology, contamination, radionuclides, test site.

INTRODUCTION

Radioecological situation at the Semipalatinsk test site has been well studied and, seemingly, in length of time, should only get better due to the decay of the dose-forming radionuclides. However, activity of one of the main dose-forming radionuclides - ^{241}Am - has been increasing so far. ^{241}Am is a ^{241}Pu decay daughter with its concentration in the weapons-grade Pu being by an order more than the ^{239}Pu activity, which is used to be conventionally determined in the environment with a relatively simple and popular alpha-spectrometric method. Having a relatively short half-life (14 years), ^{241}Pu decays rapidly in ^{241}Am ($T_{1/2}$ = 432 years). After a lapse of approximately 60 years the ^{241}Am activity at STS will reach its maximum, which, considering its half-life will not change in the near future. During the same period the activity of the elements: ^{90}Sr and ^{137}Cs will reduce by four times. Thus, in length of time the ^{241}Am input in the STS radiological situation will only increase. In order to provide a qualitative change forecasting that will take place in radiological situation as time goes one should know how ^{241}Pu is distributed across the STS territory at the moment.

It is rather difficult to determine ^{241}Pu. Over the whole period of the test site existence, no more than several dosens of determinations were made. Few leading radiochemical laboratories around the world are able to determine ^{241}Pu. A detailed study of ^{241}Pu distribution across the STS territory, taking into account its area, requires several thousand samples to be analyzed. All the more, taking into consideration the analysis cost, such studies look unreasonably expensive.

It is much cheaper to assess the ^{241}Pu concentration by the ^{241}Am concentration being the ^{241}Pu decay daughter. However, it is possible if the ^{241}Pu/^{241}Am concentration ratio is known and constant at least within the STS local sites.

Consequently, within the framework of the task two problems were addressed to:

- developing a technique for determining ^{241}Pu;
- determining the ^{241}Pu/^{241}Am soil concentration ratio at the STS main test sites.

1. LITERATURE REVIEW

^{241}Pu as a beta emitter is notable for a very low radiation energy (E_{max} = 20.8 keV), which in its turn limits to some extent its activity measurement.

All ionizing devices using gas as a working substance, including proportional counters, ionization chambers can be used for registering a soft beta radiation. Similar devices possess a 100% detection efficiency and maximum sensitivity only if the measuring substance is added to the working gas. However, it requires special methods for preparing the counting samples and also their chamber walls to be cleaned after each measurement [1].

The liquid scintillation (LS) detection methods are also based on dissolution of a measured material in a cocktail. Within the low energies areas typical for ^{241}Pu and ^{3}H, the detection efficiency decrease due to scintillation quench is seen. The above effects are corrected by constructing a curve of quench. The semiconductor detectors for registering soft beta rays have not found application so far.

The paper [2] describes how to determine ^{241}Pu in the environmental samples by applying LS. This method of determining is based on the co-precipitation of isotopes of plutonium with cerium fluoride, followed by alpha spectrometry for determining a chemical yield. Following the alpha-spectrometry, filter dissolves in acetone and transfers to the steel disk, being placed in the liquid scintillation cocktail. Measurement of ^{241}Pu is conducted using the LS spectrometer. The detection window was chosen within 0.5 keV-7.0 keV. The detection efficiency in the selected window was assessed using ^{241}Pu known activity labels.

Determination of the $^{239+240}$ Pu and ^{241}Pu isotopes activity is conducted in the *Isotope and Electron Microscopy Laboratories*, *Agricultural University of Norway* [3]. The technique of determination is based on the co-precipitation of plutonium with iron hydroxide and calcium oxalate, followed by the anion-exchange chromatography. For determining the chemical yield the Pu tracer should be added to the sample at the beginning of radiochemical preparation. The final organic solution is measured by the liquid scintillation spectrometer. The chemical yield for the sea water sample (100 l) ranges from 25% to 50%, while for ashing soil (40 g) – 30-60%. The detection efficiency for $^{239+240}$ Pu amounts to almost 100%, while for ^{241}Pu – 48.8%. The detection limit for ^{241}Pu is 2.2 mBq.

The paper [4] gives no description of the ^{241}Pu determination methods as such, however, it reports that tritium was used as a beta-emitter when calibrating liquid scintillation spectrometer by its detection efficiency. It should be mentioned that in the paper describing the method for determining the absolute plutonium alpha-activity in the agent samples [5], ^{241}Pu was considered an interfering radionuclide, and its detection zone was also determined by tritium.

While determining the radionuclide composition of hot particles as a result of the accident at the *Chernobyl Nuclear Power Plant*, the ^{241}Pu alpha line with a quantum yield of 2.04×10^{-3}% [6] was used. It is safe to say that this method does not provide the best detection limit.

Summarizing the above said, the paper [7] gives a brief description and also evaluates five methods for determining ^{241}Pu in the agent samples. Two of them should be mentioned. In the first technique the ^{241}Pu detection bases on increment of ^{241}Am activity. The *counting sample* should be stored within 2 years. In the second - a solution after the acid leaching of the sample is divided into two parts in the ratio 1:3. A smaller part should be subject to a standard sample preparation procedure for alpha-spectrometric analysis, a larger one - for

liquid scintillation spectrometry (co-precipitation of plutonium isotopes with iron hydroxide and calcium oxalate, and solvent extraction).

Thus, omitting the radiochemistry peculiarities, it may be concluded that two methods for determining ^{241}Pu in environmental samples should be considered as the most acceptable:

- Determining ^{241}Pu by the ^{241}Am activity increment in the ^{238}Pu detection area;
- Determining ^{241}Pu by the LS-beta-spectrometry.

2. DEVELOPMENT OF A METHOD FOR ^{241}Pu DETERMINATION

2.1. The recommended methodology framework for determining ^{241}Pu

For effective application of the LS-analysis method it is necessary to make the Pu isotopes free from other beta-emitting radionuclides present in the sample. Methods of radiochemical separation of plutonium isotopes are well known. However, when applying them, losses are inevitable considering the Pu multivalency, which makes it impossible to quantify plutonium without a tracer. Also, when using the LS method, it is necessary to take into account correction for a sample "quench", which can be achieved by using a standard ^{241}Pu solution.

The recommended method is based on a liquid scintillation analysis of the samples with known activity of alpha decaying plutonium isotopes: $^{239+240}$ Pu and ^{238}Pu. As a tracer, those plutonium isotopes which exist already in the sample, are expected to be used. For this purpose, two weights from the same carefully prepared samples are taken. The first weight intended for determining ^{241}Pu by the liquid scintillation analysis method is subject to standard procedure of radiochemical separation of plutonium isotopes by using the ion-exchange chromatography method. However, the plutonium isotopes are sorbing on the DOWEX anion exchanger with further description with hydroxylamine hydrochloride. Then, the derived solution is evapourated and dissolved in a small quantity of hydrochloric acid and mixed with the UltimaGold scintillator. The $^{239+240}$Pu and ^{238}Pu activity has been determined using the standard alpha spectrometry method from the second weight. Based on the resulted LS-spectra, the count rate of ^{241}Pu and the total alpha-activity of the sample - the $^{239+240+238}$ Pu activity (the LS method possesses a 100% alpha particles detection efficiency) are being determined. Chemical yield shows the ratio between activities of the alpha-emitters resulted from the liquid scintillation spectrometer and the known $^{239+240}$ Pu and ^{238}Pu. A tritium quench curve is used for recording a "quench" effect when determining ^{241}Pu.

2.2. Selecting detection areas

Figure 1 below shows typical amplitude spectrum of the liquid scintillation solution containing the Pu isotopes. The soft area within the range of 0 – 15 keV shows the ^{241}Pu beta-particles spectrum, however, if it extends 30 keV one can see a very diffuse resulting alpha-counting line. The highest level of beta spectrum energy and a resulting alpha-counting peak point are depending on quench of the spectrometer sample. The higher the sample quench the more maximal beta spectrum energy, and the resulting alpha-counting peak will shift to a low-energy part of the spectrum. As far as the resulting alpha-counting peak is concerned, no loss of count rate will take place.

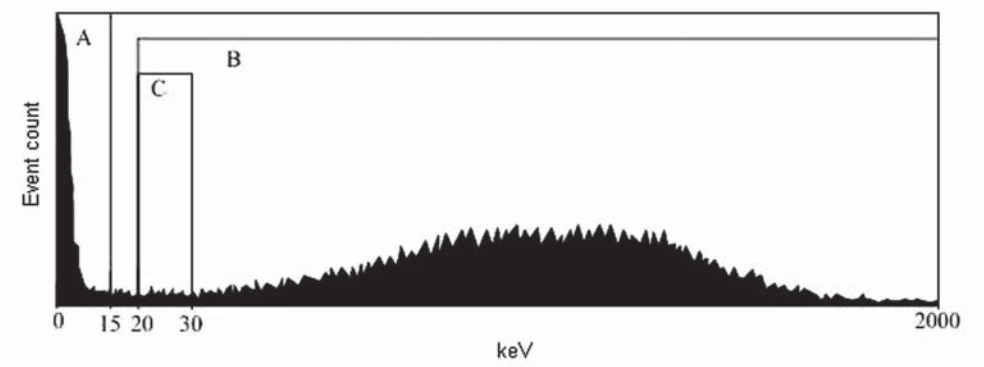

Figure 1. Amplitude spectrum of the liquid scintillation solution containing plutonium isotopes

The ^{241}Pu detection area on the LS spectrometer within the range of 0 - 15 keV was chosen, because it is a minimum area within which the growth rate comes up to 100% when measuring the unsuppressed sample of ^{3}H.

The range of a sample purity test from 20 to 30 keV was chosen so that the resulting alpha-counting peak does not contribute in this area even under strong sample quench, and no ^{241}Pu contribution to this area is seen even in the course of measuring the unsuppressed sample.

The resulting alpha-counting detection area within the range of 20 - 2000 keV was chosen. The reason for this is that the resulting alpha-counting peak fully falls within this area even when the sample is subject to intensive quench.

Thus, for recording the count rates in the concerned areas, three spectrum areas should be distinguished: area A – the ^{241}Pu (0 ÷ 15 keV) detection area, B - the resulting alpha-counting detection area (20.0-2000.0 keV) and C – the sample purity test area (20.0-30.0 keV).

The above detection areas include not only the useful signal, but also a substrate background. To determine the backgrounds in the A, B and C areas, measurements of background pattern are made in the specified detection areas using the UltimaGold scintillator.

2.3. Determining the ^{241}Pu detection efficiency

In the course of LS-measurement, chemical and physical characteristics of the sample can impact the process of scintillation, leading to changing light output because the sample is mixing with the scintillator. This phenomenon is called quench. Its basic forms are as follows: chemical quench, when emission of light is affected by chemical composition of the sample, and colour quench, when transmission of light from the scintillator atoms to the photomultiplier varies in direct ratio to absorption in the scintillation cocktail. The latter also varies depending on the sample volume, while chemical quench does not depend on its volume. In addition, the optical quench occurs when using a dirty counting tube. Thus, the spectrometer sample quench emerges in two cases: degradation of the detection efficiency and constriction of a spectrum towards lower energies. Dependence of the detection efficiency on the quench factor is used to call a quench curve.

2.3.1. Plotting a quench curve

When calculating the ^{241}Pu activity in the agent sample the a quenching effect should be taken into account. Given that the beta particle energies are close in magnitude (^{241}Pu – 20,8 keV, ^{3}H – 18.6 keV), the efficiency of its detection using the LS-method differs slightly. Therefore, to provide a quality control of measurements and to record the "quench" effect when determining ^{241}Pu, one can use the tritium quench curve prepared in advance. When making detection efficiency calibration of the spectrometer, the standard quench solutions for tritium produced by Hewlett-Packard, with a known concentration of tritium and various quenchs were used. When the samples are subject to measurement, the count rate in a selected detection window and transformed spectral index of the sample was recorded for determining the sample quench magnitude. Bearing in mind the activity of tritium contained in the sample, detection efficiency for samples with various quenchs were calculated. The transformed spectral index of the sample is calculated automatically when the sample is exposed to radiation with soft X-rays from the ^{133}Ba source (attached to a liquid scintillation spectrometer). The detection efficiency calibration should be resulted in a quench curve: dependence of detection efficiency on spectral index of the sample (Figure 2).

The resulting curve shows the dependence of detection efficiency on the magnitude of the sample quench, which is well approximated by a *second-degree polynomial* as follows:

$$\varepsilon = -0.05 + 0.00131 \times t - 6{,}75 \times 10^{-7} \times t^2$$

The detection efficiency is considered a function, while the *Transformed Spectral Index* of the *External Standard* (tSIE) - an argument.

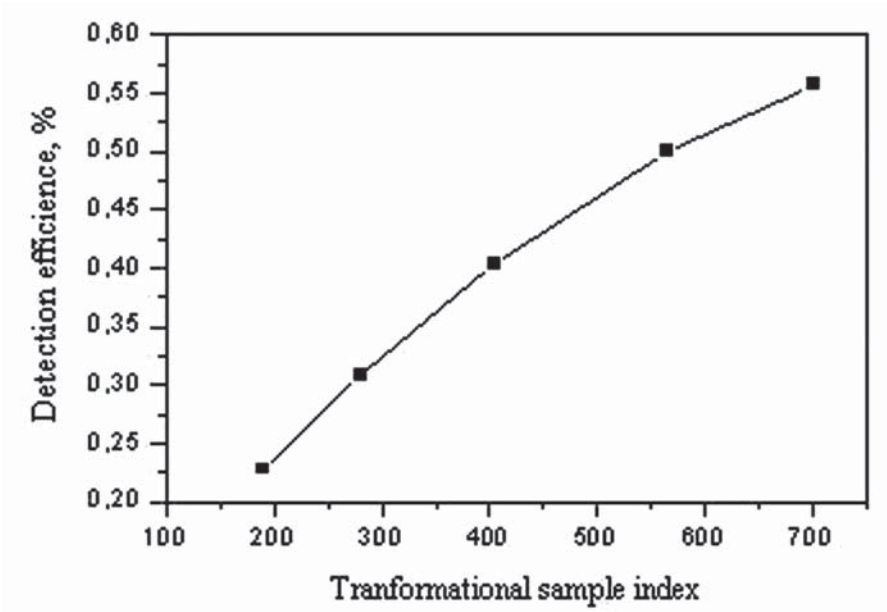

Figure 2. Quench curve of the sample for ^{3}H

2.4. Assessing the impact of interfering factors

2.4.1. Controlling the sample purity

It is quite difficult to identify and separate radionuclides contained in the sample under study with the help of the LS spectrometry. The most routine method appears to be as follows: the liquid scintillation spectrum of the sample should be measured first, then, for the appropriate quench level from the library of the base individual radionuclides spectra, using interpolation and extrapolation method, the normalized model spectra of individual radionuclides should be determined. Next, the difference between the sample spectrum and sum of the individual radionuclides model spectra, multiplied by the factors determining the activity of the individual radionuclides should be minimized using the least-squares method. However, spectrometers, using these methods do not allow identifying radionuclides having low activity if high activity radionuclides are existed in the sample.

In this case, all natural and artificial beta-emitting radionuclides existing in the sample can impact the quality of the ^{241}Pu measurements. List of beta-emitting radionuclides which are most likely existed in the sample, and their maximum beta-particles energy are shown in Table 1.

Table 1.

Beta-emitting radionuclides which are most likely presented in the soil samples, and their maximum beta-particles energy

Nuclide	Max Eβ, MeV	Nuclide	Max Eβ, MeV
^{3}H	0.0186	^{214}Bi	3.272
^{241}Pu	0.0208	^{210}Pb	0.063
^{99}Tc	0.293	^{210}Bi	1.170
^{90}Sr	0.535	^{231}Th	0.305
^{137}Cs	1.170	^{227}Ac	0.046
^{151}Sm	0.076	^{211}Pb	1.372
^{60}Co	0.318	^{207}Tl	1.423
^{152}Eu	1.474	^{228}Ra	0.039
^{154}Eu	1.843	^{228}Ac	2.069
^{234}Th	0.199	^{212}Pb	0.573
^{234m}Pa	1.205	^{212}Bi	2.254
^{214}Pb	1.024	^{40}K	1.311

Radiochemical procedure for separation of plutonium isotopes assures a reasonably good removal of the following radionuclides: ^{228}Ra, ^{40}K, ^{151}Sm, ^{3}H, ^{137}Cs, ^{90}Sr, ^{60}Co, ^{207}Tl, as well as isotopes of both lead and bismuth, however, it gives no guarantee of a constant high level removal of radionuclides such as ^{227}Ac, ^{99}Tc, and isotopes of uranium and thorium. Since the procedure of identifying and separating the spectra from various radionuclides by the

liquid scintillation spectrometry is rather complicated, it is necessary to provide c*ontinuous monitoring* of *purity* of the spectrometric samples.

Since the maximum beta-particles energy of other radionuclides possibly presenting in the sample is at least twice the maximum energy of the ^{241}Pu beta-particles (^{228}Ra from ^{232}Th series has a maximum energy of 39 keV, see Table 1), their presence in the spectrometric sample have to lead to an increased count rate in the area located behind the ^{241}Pu detection area. For this reason, monitoring of the sample purity for beta-emitting radionuclides is recommended to be carried out by using the count rate recording method within the range from 20 to 30 keV, and by comparing its results with the count rate registered in the same area while conducting measurement of a clean scintillator. If the count rate in the area above the background fluctuations exceeds more than 10%, the sample should be considered "dirty" and sent back for retreatment.

If the sample contains a soft beta-emitter like ^{227}Ac, the above recommended monitoring method does not provide a 100% probability of culling of "dirty" sample, because under a strong enough quench of a spectrometric sample, the spectrum is compressed towards a soft-energy side, thus, it is likely that the given radionuclide will not give additional count rate within the range of 20-30 keV. Thus, it is necessary when conducting the gamma-spectrometric analysis (prior to the radiochemical separation of plutonium isotopes) to pay attention to the ^{235}U concentration in the test sample. The ^{235}U concentration in the soil averages about 2 Bq/kg, its daughter ^{227}Ac shows the same concentration. When working with 10 g of the weight, under conditions of potentially poor radiochemical ^{227}Ac purification, its maximum activity in the spectrometric sample would be 0.02 Bq. Such activity will lead to no more than 20% overestimation of the ^{241}Pu detection results, even if analyzing concentrations close to detection limit by the recommended method.

2.4.2. Determining the impact of the reagents used for the radiochemical purification of the sample on the background count rate

In the course of isolation of the Pu isotopes by column extraction chromatography, the wash-out of extractant (DOWEX) and hydroxylamine hydrochloride and their ingress into the spectrometric sample may occur, and it may cause changes in the count rates. In order to find out how it impacts the background count rate the following experiment was conducted. 3 counting samples of the following composition were prepared:

- 20 ml of the UltimaGold scintillator;
- 1 ml of 0.5 M of hydroxylamine hydrochloride + 19 ml of the UltimaGold scintillator;
- 0.5 g of DOWEX + 20 ml of the UltimaGold scintillator.

Measurements were performed within 2 hours using a liquid scintillation spectrometer TRI-CARB 2900. The count rate was recorded within the range of 0-15, 20-30, 20-2000 keV. The measurements were repeated 24 hours later. The measurement results are presented in Table 2. According to the results, contribution of the used reagents to the background count rate is negligible and does not exceed the statistical error in the background count rate by the 2σ criterion. Contribution to the background count rate from the reagents used does not change as time goes.

Table 2.

Measurements the counting samples with various reagents

Measurement series	Window range, keV	Sample No					$2\sigma_B$, count/min
		1	2		3		
		S_B, count/min	S_O, count/min	$\lvert S_O - S_B \rvert$, count/min	S_O, count/min	$\lvert S_O - S_B \rvert$, count/min	
1	0 - 15	14.8	15.1	0.3	14.9	0.1	0.7
	20 - 30	3.6	3.8	0.2	3.7	0.1	0.3
	20 - 2000	37.2	36.8	0.4	37.1	0.1	1.1
2	0 - 15	15.1	14.8	0.3	14.9	0.2	0.7
	20 - 30	3.7	3.7	0	3.8	0.1	0.4
	20 - 2000	37.5	37.4	0.1	37.9	0.4	1.1

Thus, it may be concluded that the reagents used for the radiochemical purification of the sample and possibly occurring in the counting probe, have no impact on the background count rate.

2.4.3. Verification of the utensils and used reagents cleanliness

For verification purposes there were prepared five spectrometric samples, containing only reagents used according to the radiochemical separation method, and which were subject to all the procedures under the recommended method. Their measurements were conducted applying the liquid scintillation spectrometer TRI-CARB 2900 over 2 hours. The count rate was recorded within 0-15, 20-30, 20-2000 keV. The measurement results are presented in Table 3.

Table 3.

Measurement of blank samples

Range, keV	Count rate S, count/min					Average count rate S_{av}, count/min	RMSE, count/min	Relative RMSE
	Sample No							
	1	2	3	4	5			
0 - 15	14.7	14.5	15.1	14.9	15.2	14.9	0.3	0.02
20 - 30	3.6	3.7	3.9	3.8	3.5	3.7	0.2	0.04
20 - 2000	37.5	37.4	36.9	38.1	38.5	37.7	0.62	0.02

The performed experiment did not reveal the effect of used in the formulation of methods of dishes, on the background count rate in the spectral band of interest.

2.5. Verification the total alpha-activity accuracy by additive method

Method of additives was chosen for verification of the total alpha-counting accuracy. Further, according to a previously described method, 5 soil samples were prepared, the preliminary measurement of total alpha-counting was conducted, 2.16 Bq of ^{242}Pu were added to each sample and measurements were repeated. The results of verification of the total alpha-activity accuracy are shown in Table 4.

Table 4.

Verification of the total alpha-activity accuracy

##	Total alpha-activity, Bq 1st measurement	Added, Bq	Total, Bq	Total alpha-activity, Bq 2nd measurement	Deviation, %
1	4.03	2.16	6.19	6.30	-1.77
2	5.07	2.16	7.23	7.03	2.77
3	2.91	2.16	5.07	5.16	-1.87
4	2.22	2.16	4.38	4.36	0.36
5	4.90	2.16	7.06	7.15	-1.37

Based on the data presented in Table 4, it may be concluded that the total alpha-counting determination was conducted correctly.

3. DESCRIPTION OF THE TECHNIQUE FOR ^{241}Pu DETERMINATION

The recommended analysis outline provides for a careful homogenization of the sample and is applicable only to the samples containing the sum of $^{239+240}$ Pu and ^{238}Pu and being no less than 0.2 Bq per a sample.

3.1. Preliminary sample preparation

The samples under study (no less than 1 kg) should be dried to air-dry weight, of them a <2 mm fraction should be extracted and by using a quartering technique the weight of about 200 g should be taken away from the isolated fraction. The weight should be grinded in a mill. For testing the sample homogeneity after grinding, 2 parallel subsamples of approximately 100 g each taken from each sample should be prepared by quartering method to determine ^{241}Am by gamma-spectrometry method. If measured concentration of ^{241}Am in the subsamples differs for more than 10%, the subsamples should be combined and grinded again to provide further homogenization. Then, again, 2 parallel subsamples of approximately 100 g each should be prepared by quartering method to determine ^{241}Am by gamma-spectrometry method. If the results of determining ^{241}Am shows sufficient precision, the samples have to be combined, and the weight of 50-75 g should be withdrawn by quartering and again grinded with a mill. From the grinded sample using the quartering method, two samples of 10 g should be taken for further determination of $^{239+240}$ Pu, ^{238}Pu and ^{241}Pu.

3.2. Determining $^{239+240}$ Pu and ^{238}Pu

Determination of $^{239+240}$ Pu and ^{238}Pu is carried out using one of the subsamples in accordance with the standard procedure [1], including the following: ashing, complete acid digestion, and Pu isotopes isolation by column extraction chromatography followed by elution and obtaining the alpha-spectrometric sources through electrodeposition on a stainless steel substrate. Measurement and calculation of activity should be carried out by the standard alpha-spectrometry methods. ^{242}Pu should be used as a tracer.

The sample purity test for the alpha-emitting radionuclides should be performed as follows: the own alpha-camera background is deducted from the measured alpha-spectrum, then the peak areas in the detection window of $^{239+240}$ Pu, ^{238}Pu, and the integral counting within the entire detection range are recorded on the spectrum. If the integral counting exceeds the sum of peak areas of $^{239+240}$ Pu and ^{238}Pu by 10%, the sample are qualifying as "dirty", and both weighed samples are sending back for preparation.

3.3. Determination of ^{241}Pu

The plutonium isotopes are isolated from the second weight by the same method, with no tracer being added. Following desorption, about 80 ml of the eluate are collected in a 100 ml glass jar. All eluate is transferred to a 20 ml glass vessel successively by 10 ml and evapourated to dryness. Then 0.2 ml of 0.5 M hydrochloric acid and 20 ml scintillator UltimaGold are added to this glass vessel.

Measurements are carried out using the liquid scintillation spectrometer TRI-CARB 2900 (Hewlett Packard). To register the count rate of ^{241}Pu, the area within the range from 0 to 15 keV is shown. Area for recording the total alpha-counting within the range from 20 to 2000 keV is shown. The sample purity test ranges from 20 to 30 keV. The measurement lasts for 2 hours Function of calculation of the transformed spectral index of the sample (tSIE) must be included into the measurements.

The LS spectrometer is prepared for operation in accordance with its operating guide. The spectrometer is subject to self-normalization automatically once a day. Prior to measuring, the background count rate is determined by conducting measurements of the sample containing 20 ml of scintillator within 2 hours.

$$A(Pu^{241}) = \frac{\frac{(N_A - N_{A,F})}{\varepsilon} \cdot A(Pu^{238+239+240})}{(N_B - N_{B,F})}, \text{ where} \qquad (1)$$

Further, the ^{241}Pu absolute activity is calculated by the following formula:

A ($^{Pu238\ 239,240}$) – the sum of absolute $^{239+240}$Pu, ^{238}Pu activities deduced based on the alpha-spectrometric data.

(NA-NA, F) – difference between the count rate of the sample and the background count rate within the range from 0 to 15 keV, expressed in terms of pulses per second.

(NB-NB, F) – difference between the count rate of the sample and the background count rate within the range from 20 to 2000 keV expressed in terms of pulses per second.

ε – the ^{241}Pu detection efficiency calculated by formula:

$$\varepsilon = -0.05 + 0.00131 \times t - 6{,}75 \times 10^{-7} \times t^2 \text{ , where} \qquad (2)$$

t – the transformed spectral index of the sample (tSIE), which is calculated automatically by the program of the LS-spectra processing installed on a spectrometer TRI-CARB 2900.

4. EVALUATION OF METROLOGICAL CHARACTERISTICS OF THE ^{241}PU DETERMINATION TECHNIQUE

Evaluation of metrological characteristics of the methodology encloses calculation of the reproducibility of the parallel samples, the ^{241}Pu detection limit and the uncertainty of spectrometric measurements.

4.1. Calculating the technique detection limit

To calculate the technique detection limit, three background measurements in the ^{241}Pu detection area (0-15 keV) were conducted within 2 hours, with its average value being 14.7 count per minute. The ^{241}Pu detection limit based on the developed technique, taking into account the average 50% chemical yield and the 30% average ^{241}Pu detection efficiency by criterion 3δ is equal to 20 Bq/kg.

If it is necessary to lower the ^{241}Pu detection limit, then the measurement time could be increased. If the measurement time takes 4 hours, the ^{241}Pu detection limit is 13 Bq/kg, if measurement takes 8 hours - 9 Bq/kg.

4.2. Determining the reproducibility of measurements

To calculate the reproducibility of measurements, three samples of known specific activity of ^{241}Pu were used. Each sample was divided into five equal parts, and each part was analyzed independently in accordance with the developed technique.

Based on the LS-analysis results, the specific activity of each series of parallel samples, the average specific activity, the standard deviation of the specific activity and the specific value of standard deviation were calculated. It takes 2 hours to measure each sample. The resulted values are shown in Table 5.

Table 5.

Verification of the reproducibility of the ^{241}Pu determination technique

Sample No	Activity, Bq/kg					Average, Bq/kg	MSRE, Bq/kg	Relative value, %
	Sample No							
	1	2	3	4	5			
1	-	55	64	51	37	52	11	21
2	420	535	598	480	366	479	91	19
3	8059	7812	7238	7411	7456	7595	332	4.4

4.3. Calculation of the measurement uncertainty

Calculation of the spectrometer measurements uncertainty was conducted based on spectrometer measurements of the parallel samples. Mathematical calculation was carried out based on "Guide to the Expression of Uncertainty in Measurement" [11]. It is important to note that the sampling uncertainty should not be considered at this stage. The averaged values of the relative spectrometric measurements uncertainty with a confidence coefficient of 99% for each range of specific activity are presented below:

- Up to 100 Bq/kg - more than 50%;
- 100-1000 Bq/kg - 20%;
- 1000-10000 Bq/kg -15%.

High measurement uncertainty of the lowest activity range may occur due to the closeness of the activity values (52 Bq/kg) to the detection limit.

5. DETERMINING THE ^{241}Pu AND ^{241}Am IN SOIL OF MAIN STS TEST SITES

5.1. Sampling

For the purpose of studying the topsoil samples (0-5 cm deep) were taken from the most typical test sites of the former STS:

- tunnels within Degelen mountain massif where tests were conducted;
- excavation explosion area ("Atomic" lake);
- "*Opytnoye Pole*" and territory within the circle of influence;
- site testing of radiological warfare agents (RWA);

A total of 19 samples was prepared for the study purposes. The sampling areas are presented in Table 6.

Table 6.

The areas of sampling for the purpose of investigation with an indication of a number of samples and supposed type of contamination

#	Number of samples	Area of sampling	Type of contamination
1	9	"*Opytnoye Pole*" and its impact area	Radioactive contamination as a result of ground and aerial bombings, hydronuclear tests and also as a result of fallout of radioactive agents during the passage of radioactive cloud
2	3	The "RWA" site	Radioactive contamination as a result of testing of radiological warfare agents
3	2	Tunnel 177	Radioactive contamination as a result of removal of radionuclides from water flowing from the tunnels
4	2	Tunnel 143	
5	3	"Atomic" lake	Radioactive contamination resulting from the thermonuclear explosion

5.2. Preliminary sample preparation and analysis procedure

The investigated samples of 0.8-1.2 kg were dried to air-dry weight, of them a <2 mm fraction were extracted and by using a quartering technique a sample of 600 g was taken away. The sample was grinded in the attrition mill (*PULVERISETTE 9 Fritsch)* within 20 minutes. Then, from each sample three parallel samples of ~ 200 g were prepared for determining ^{241}Am. If the convergence of the results on parallel samples is lower than 10%, the samples were combined, and additional homogenization by grinding (except when the concentration of ^{241}Am approaches the detection limit) was carried out. After determination it

was completed, the samples were combined, by using quartering method the weight of 100-150 g was taken and additional grinding took place within 20 minutes, of them by quartering two samples of 10 g were taken for further determination of $^{239+240}$Pu, ^{238}Pu and ^{241}Pu.

5.3. Determining the ^{241}Am and ^{241}Pu concentrations

Measurements were conducted by the instrumental gamma spectrometry using the "Canberra" germanium detector BE 2020 with a carbon-composite entrance window and 122.0 keV - 650 eV energy resolution along the line. The measurement time for minimizing the statistical error varied from 3 hours to 3 days depending on the samples activity. Accuracy of the ^{241}Am determination was monitored by measurements of the IAEA standards (IAEA-134, IAEA-135) from periodically. Comparison of the results with the certified concentration values showed absence of a systematic error applied by the analytical technique. Also, the analysis accuracy is verified by the data of interlaboratory comparison conducted in 2009. Results of the international interlaboratory comparison are presented in Table 7.

Table 7.

Results of the International interlaboratory comparison

Sample No	Measured activity of ^{241}Am, Bq/kg	Verified activity of ^{241}Am, Bq/kg	Deviation, %
1	48.9 ± 1.0	51.900 ± 0.620	-5.78
2	1.6 ± 0.3	1.579 ± 0.060	1.35
3	50.2 ± 1.0	51.900 ± 0.620	-3.28

The ^{241}Pu determination was conducted in compliance with the developed technique.

6. RESULTS AND DISCUSSION

6.1. "*Opytnoye Pole*" and its impact area

Table 8 presents the results of determining ^{241}Pu and ^{241}Am, and their concentration ratio in the soil samples taken at the "*Opytnoye Pole*" site. Figure 3 shows the sampling points.

Table 8.

Results of determining ^{241}Pu and ^{241}Am, and their ratio in the soils sampled at the "*Opytnoye Pole*" site and in the territory within its impact area

Sample	^{241}Am, Bq/kg	^{241}Pu, Bq/kg	^{241}Pu/^{241}Am	
OP-01	480 ± 40	1680 ± 330	3.5 ± 1.0	Average 2.4 ± 0.7
OP-07	124 ± 12	200 ± 40	1.6 ± 0.5	
OP-08	4730 ± 420	9100 ± 1800	2.0 ± 0.5	
OP-09	107 ± 8	540 ± 110	5.0 ± 1.5	

Sample	^{241}Am, Bq/kg	^{241}Pu, Bq/kg	^{241}Pu/^{241}Am	
OP-10	44 ± 4	170 ± 30	3.9 ± 1.0	Average 2.4 ± 0.7
OP-11	710 ± 140	920 ±184	1.3 ± 0.3	
OP-12	60 ± 5	180 ± 40	3.0 ± 0.7	
OP-13	570 ± 40	1400 ± 280	2.5 ± 0.6	
OP-14	320 ± 30	620 ± 120	2.0 ± 0.5	

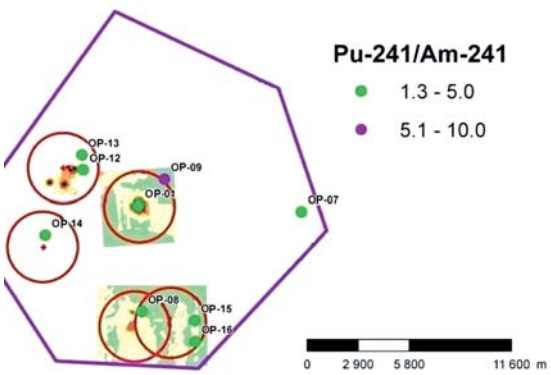

Figure 3. Sampling points and the resulted ratio revealed at the "*Opytnoye Pole*" site and in the territory within its impact area.

In 90% of cases the ^{241}Pu/^{241}Am ratio registered immediately at the test sites shows a narrow range within 1.3 – 3.9 with its average value being 2.4. To assess the reliability of the data, theoretical calculation of the eventual ^{241}Pu/^{241}Am ratio was conducted.

Due to the fact that activities of ^{241}Pu and its daughter ^{241}Am radionuclide are following the radioactive decay law, thus assuming that at time "zero" the ^{241}Am activity is also equal to zero, eventually it is possible to calculate the change in their activity ratio. The curve describing the change in ^{241}Pu/^{241}Am activity is presented in Figure 4.

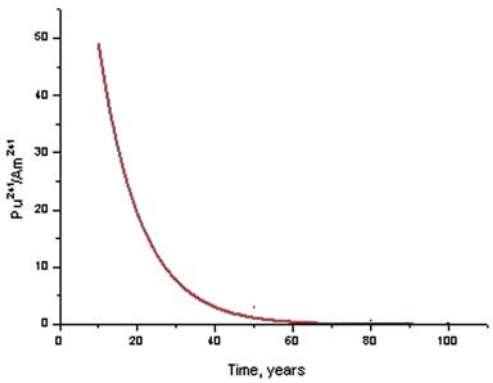

Figure 4. Changes in the ^{241}Pu/^{241}Am ratio with time

Tests were conducted at "*Opytnoye Pole*" within the period from August 29, 1949 to January 24, 1962. Assuming that the ^{241}Am activity of an exploding charge at the moment of test is the lowest (for further calculations - zero), with due account for the radioactive decay law, theoretically calculated eventual ^{241}Pu/^{241}Am ratio may be seen within the range from 2.4 to 4.7.

Some differences between the experimental data and theoretical calculations may be accounted for various migration properties of plutonium and americium and their redistribution in the soil.

6.2. Tunnels of Degelen mountain massif

Table 9 below presents results of determining ^{241}Pu and ^{241}Am, and their ratio in the soil samples taken at the "*Degelen*" site.

The range of the resulted ^{241}Pu/^{241}Am ratio for each of the two individual tunnel is within a rather narrow range. The test conducted in the 177 tunnel took place on March 30, 1983; currently, the theoretically calculated ^{241}Pu/^{241}Am ratio is equal to 16. 2, while according to the analysis results it averages to 18.8 ± 4.7. The test conducted in the 143 tunnel took place on October 30, 1976, currently, the theoretically calculated ^{241}Pu/^{241}Am ratio is equal to 10.5, while according to the analysis results it averages to 3.8 ± 1.0.

According to [12], such difference between the analysis results and theoretical calculation made in the 143 tunnel can be accounted for by the following:

- the purpose of the test taken place in the 177 tunnel is "Development or improvement of nuclear weapons (DNW)";
- the purpose of the test taken place in the 143 tunnel is "Investigation of the NW damage effects and their impact on military and civilian facilities (IDE).

Table 9.

Determined ^{241}Pu and ^{241}Am, and their ratio found in the STS soils contaminated as a result of radionuclide migration with water flowing from the tunnels

Sample	^{241}Am, Bq/kg	^{241}Pu, Bq/kg	^{241}Pu/^{241}Am	
tunnel 177/1	940 ± 65	17600 ± 3500	18.7 ± 4.7	Average 18.8 ± 4.7
tunnel 177/2	810 ± 60	15300 ± 3100	18.9 ± 4.7	
tunnel 143/1	410 ± 36	2100 ± 400	5.0 ± 1.3	Average 3.8 ± 1.0
tunnel 143/2	746 ± 65	2000 ± 400	2.6 ± 0.6	

The above enables to assume quite reliably that for the test taken in the tunnel 177 a newly created "fresh" charge was used, while in test taken place in the tunnel 143– a series-production "proved" charge manufactured long ago. The reverse theoretical calculation showed that the best time of this charge creation - 1958-1959 years, that is the above charge remained stored about 18 years. This is entirely complying with the nuclear charge shelf life, which according to some data is about 30 years [10].

Also, the difference between the test data and theoretical calculations may be accounted for by various physical and chemical properties of plutonium and americium, their various migration properties and, as a consequence, by their redistribution in the soil.

6.3. "Atomic" lake

Table 10 shows the results of determining ^{241}Pu and ^{241}Am and their ratio in the soil samples, picked on the "*Atomic" lake* site.

Table 10.

Determined ^{241}Pu and ^{241}Am and their ratio in the STS soils contaminated as a result of excavated explosion

Sample	^{241}Am, Bq/kg	^{241}Pu, Bq/kg	^{241}Pu/^{241}Am	
AL-01	344 ± 28	1430 ± 290	4.2 ± 1.0	Average 3.7 ± 1.0
AL-02	1090 ± 83	3090 ± 620	2.8 ± 0.7	
AL-03	533 ± 48	2160 ± 430	4.1 ± 1.0	

Relatively narrow range of the ^{241}Pu/^{241}Am ratio has the registered limits from 2.8 to 4.2 with its average - 3.7.

The test took place on January 15, 1965. Currently, the theoretically calculated ^{241}Pu/^{241}Am ratio is equal to 5.3, while the resulted average is 3.7 ± 1.0.

6.4 The "RWA" site

Table 11 presents the results of determining ^{241}Pu and ^{241}Am and their ratio in soil samples taken at the "RWA" site.

Table 11.

Determined ^{241}Pu and ^{241}Am, and their ratio in the STS soils contaminated as a result of testing of radiological warfare agents

Sample	^{241}Am, Bq/kg	^{241}Pu, Bq/kg	^{241}Pu/^{241}Am	
RWA-01	20 ± 1,6	29.8 ± 6.0	1.5 ± 0.4	Average 1.3 ± 0.4
RWA-02	26 ± 1,8	45.2 ± 9.0	1.7 ± 0.4	
RWA-03	690 ± 57	450 ± 90	0.6 ± 0.2	

The range of the ^{241}Pu/^{241}Am ratio has registered limits from 0.6 to 1.5 with the average equal to 1.3.

Tests at the "RWA" site took place within the period from 1953 to 1957, and currently, the range of the theoretically calculated ^{241}Pu/^{241}Am ratio varies from 2.4 to 4.7. However, in this case it is difficult to calculate theoretically the ^{241}Pu/^{241}Am ratio, because this area was contaminated as a result of spraying mixtures of radiological warfare agents made of the spent reactor fuel after radiochemical separation of plutonium isotopes, and thus containing a fair amount of ^{241}Am which accumulated in the reactor. This fact leads to a significant overstatement of the theoretically calculated range of the ratio.

CONCLUSIONS

The technique for determination of ^{241}Pu in soils of the main STS test sites has been developed. The detection limit of the proposed method is 20 Bq/kg.

In most cases, the ^{241}Pu/^{241}Am ratio observed at the local sites is seen in a fairly narrow range, which enables to provide an efficient and relatively cheap assessment of the ^{241}Pu activity based on the ^{241}Am gamma-spectrometric results. The accuracy of the ^{241}Pu activity assessment using this method will be about 50%, and it is sufficient for most cases. However, this assessment method is not applicable to all STS's test sites. Both sites, "RWA" and "Degelen" are being an exception because contamination at the "RWA" site is having a local spots nature with significantly different isotopic ratios, and on the "Degelen" site a significant radionuclides redistribution impact along the streambeds is registered.

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ССП НЕГІЗГІ СЫНАҚ АЛАҢДАРЫНЫҢ ТОПЫРАҒЫНДАҒЫ ^{241}Pu ЖӘНЕ ^{241}Am БЕЛСЕНДІЛІКТЕРІНІҢ АРАҚАТЫНАСЫН ЗЕРТТЕУ

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Семей сынақ полигонының негізгі сынақ алаңдарының топырағындағы ^{241}Pu анықтау әдісі әзірленді. ССП негізгі сынақ алаңдарының топырағынан алынған сынамалардан ^{241}Am және ^{241}Pu белсенділігін анықтаудың нәтижелері ұсынылды. ^{241}Am деген ^{241}Pu белсенділігінің арақатынасы есептелді.

Көптеген жағдайларда жергілікті алаңдарда ^{241}Pu/^{241}Am арақатынасы өте жақын аралықта жатыр, бұл өз кезегінде ^{241}Am гамма-спектрометриялық талдамасының нәтижелері бойынша ^{241}Pu белсенділігіне бағалау жасауға мүмкіндік береді. Бұндай әдіспен ^{241}Pu белсенділігін бағалау дәлдігі шамамен 50 %-ды құрайды, бұл көп жағдайда жеткілікті болып табылады.

Түйінді сөздер: ядролық сынақтар, мониторинг, радиоэкология, радиоактивті ластану, радионуклидтер, полигон.

ИССЛЕДОВАНИЕ ОТНОШЕНИЙ АКТИВНОСТИ ^{241}Pu И ^{241}Am В ПОЧВЕ ОСНОВНЫХ ИСПЫТАТЕЛЬНЫХ ПЛОЩАДОК СИП

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Разработана методика определения ^{241}Pu в почвах основных испытательных площадок Семипалатинского испытательного полигона. Представлены результаты определения активностей ^{241}Am и ^{241}Pu в пробах почвы, отобранных на основных испытательных площадках СИП. Рассчитаны отношения активностей ^{241}Pu к ^{241}Am.

В большинстве случаев на локальных площадках отношение ^{241}Pu/^{241}Am лежит в достаточно узком интервале, что делает возможным проведение оценки активности ^{241}Pu по результатам гамма-спектрометрического анализа ^{241}Am. Точность оценки активности ^{241}Pu таким методом будет составлять около 50 %, что в большинстве случаев вполне достаточно.

Ключевые слова: ядерные испытания, мониторинг, радиоэкология, радиоактивное загрязнение, радионуклиды, полигон.

CHAPTER: SAFEGUARDING OF WORK PERFORMANCE AT STS

УДК 577.4:615.849:504.064:539.16:622.258.4

INDIVIDUAL DOSE FORMATION AT WORKS ON SUPPLEMENTARY PROTECTION OF ENGINEERING STRUCTURES IN TUNNELS OF "DEGELEN" TEST SITE

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The article reports on calculated individual doses gained by personnel involved in creation of supplementary protection for engineering structures in tunnels of "Degelen" test site. The main factors that contribute to individual doses gained during these works have been determined; we have calculated the dose loads on personnel from external sources of ionizing radiation and due to inhalation of radionuclides. Recommendations for optimization of radiation safety assurance at construction of additional engineering protection have been developed.

Key words: test site, "Degelen", radionuclides, protection of engineering structures, tunnel, radiological situation, radiological parameters, dose formation, external irradiation and exposure, internal irradiation.

INTRODUCTION

Since 2005, in the territory of STS works have been being carried out to create additional protection for engineering structures in the tunnels of "Degelen" site. These works were performed based on a trilateral agreement between Republic of Kazakhstan, Russian Federation and the United States.

Carrying out works on the construction of additional protection for the facilities could lead to increased radiation doses to personnel and to environmental contamination with radioactive substances.

When working IRSE NNC professionals maintained a radiation safety operation system, which includes a preliminary radiological survey, ensuring radiation safety when erecting the protection and a final examination after completion of works. The main objective of radiation safety is to prevent overexposure of personnel involved in the works and to prevent negative impact of the works on the environment.

Generalized performance criterion for ensuring radiation safety of the personnel is occupational exposure dose [1]. Radiological exposure data after processing and generalization become an initial material to assess the overall performance of the radiation safety assurance system and to provide basis for development of practical recommendations on improvement of working conditions of the staff at realization of radiation-hazardous works. The purpose of an overall assessment of individual doses and control is to avoid exceeding the dose limits of personnel and obtaining information to optimize the activities on RS.

1. METHODS TO CREATE ADDITIONAL ENGINEERING PROTECTION

Tunnel is a horizontal opening in the rock; its cross-section is ~ 9 - 7 m^2 and length is up to 2 km (Figure 1).

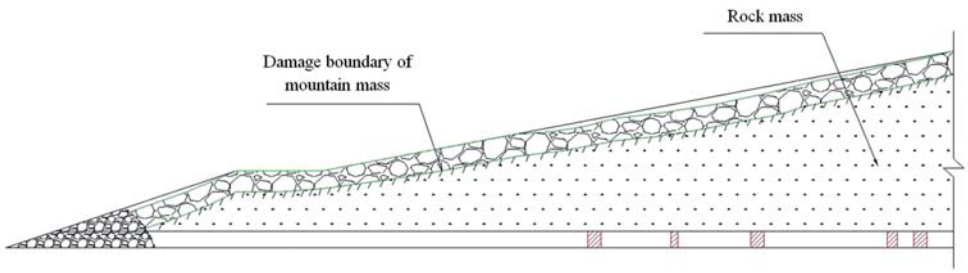


Figure 1. Tunnel sketch

To reduce the threat of proliferation of nuclear activities wastes (NAW) we are creating the additional protection on the facilities, based on filling the cavities of tunnels with cementing material that eliminates the possibility of unauthorized extraction of NAW.

Creation of additional protection, according to the design assignment is done by two methods:

- Creation of additional engineering protection through vertical wells drilled from the surface of a rock - the vertical method;
- Creation of additional engineering protection by opening the portal of the structures and the subsequent working the tunnels - horizontal method.

Creation of additional engineering protection for the tunnels, depending on the chosen method of work includes several major steps:

Vertical method

- setting-up;
- drilling the prospecting hole to the cavity of the tunnel;
- creation of additional engineering protection through vertical holes.

Works at tunnels when constructing additional engineering protection with vertical method can take from 1.5 to 3 months.

Horizontal method

- setting-up;
- opening the tunnel portal and cleanup the cavity of the tunnel from collapses and blockages;
- removal of concrete "plugs";
- creation of additional engineering protection;
- closure of the facility.

Works at tunnels when constructing additional engineering protection with horizontal method can take from 3.5 to 5 months:

Depending on the method for creating a supplementary protection, the tunnels have been assigned as:

- horizontal method of creating additional protection – H1, H2, H3, H4;
- vertical method of creating additional protection – V1, V2, V3, V4, V5.

2. RADIOLOGICAL CONDITIONS AT THE SITE

Continuous radiation monitoring was established during the works. Radiation safety assurance system during the works included a set of daily measurements of radiation parameters at the work site and in the field camp: EDR, flux density of β-particles and volumetric activity of radionuclides in the air. Control of radiation parameters at the work site and in the field camp was conducted on a daily basis. Maximum and minimum values of radiation parameters, registered during the period of works for each tunnel are presented in Table 1 and Figures 2-4.

Table 1.

Data on the radiological situation in working venues

Tunnel code	EDR, μSv/h		flux density of β-particles, particles/(min*cm²)		EEVA ^{222}Rn, Bq/m³		EEVA ^{220}Rn, Bq/m³		Volumetric activity in the air, Bq/m³					
									^{241}Am		^{137}Cs		^{3}H	
	min	max	min	max	min	max	min	max	min	max	min	max	min	max
H1	0,14	50	<10	$4\cdot10^{4}$	<4	$1\cdot10^{4}$	<8	$4\cdot10^{3}$	$<6\cdot10^{-7}$	$8\cdot10^{-2}$	$<2\cdot10^{-5}$	$1\cdot10^{-2}$	$<5\cdot10^{-2}$	$4\cdot10^{1}$
H2	0,19	2,7	<10	$3\cdot10^{2}$	<4	$4\cdot10^{3}$	<8	$4\cdot10^{3}$	$<8\cdot10^{-6}$	$<5\cdot10^{-3}$	$<6\cdot10^{-5}$	$<8\cdot10^{-3}$	$<3\cdot10^{-1}$	$2\cdot10^{1}$
H3	0,12	3,98	<10	$2\cdot10^{2}$	<4	$4\cdot10^{4}$	<8	$5\cdot10^{2}$	$2\cdot10^{-5}$	$6\cdot10^{-3}$	$<9\cdot10^{-6}$	$4\cdot10^{-1}$	$<1\cdot10^{-1}$	$7\cdot10^{1}$
H4	0,16	16	<10	$2\cdot10^{4}$	<4	$3\cdot10^{2}$	<8	$2\cdot10^{1}$	$<2\cdot10^{-5}$	$<3\cdot10^{-2}$	$<3\cdot10^{-6}$	$<3\cdot10^{-1}$	$<1\cdot10^{-2}$	$8\cdot10^{0}$
V1	0,12	0,30	<10	$1\cdot10^{1}$	<4	$1\cdot10^{3}$	<8	-	$7\cdot10^{-5}$	$<7\cdot10^{-4}$	$<8\cdot10^{-5}$	$<9\cdot10^{-3}$	$<1\cdot10^{-1}$	$3\cdot10^{1}$
V2	0,15	0,30	<10	$2\cdot10^{1}$	<4	$1\cdot10^{2}$	<8	-	$<2\cdot10^{-3}$	$<5\cdot10^{-3}$	$<8\cdot10^{-3}$	$<1\cdot10^{-2}$	$<5\cdot10^{-1}$	$5\cdot10^{1}$
V3	0,10	0,18	<10	$1\cdot10^{1}$	<4	$1\cdot10^{2}$	<8	$4\cdot10^{3}$	$<4\cdot10^{-5}$	$<5\cdot10^{-4}$	$<2\cdot10^{-4}$	$<6\cdot10^{-3}$	$<5\cdot10^{-2}$	$8\cdot10^{-1}$
V4	0,17	0,34	<10	$6\cdot10^{4}$	<4	$2\cdot10^{2}$	<8	-	$<3\cdot10^{-5}$	$<2\cdot10^{-3}$	$<5\cdot10^{-5}$	$<8\cdot10^{-3}$	$<1\cdot10^{-1}$	$4\cdot10^{0}$
V 5	0,18	0,20	<10	$4\cdot10^{4}$	<4	$4\cdot10^{1}$	<8	-	$<1\cdot10^{-7}$	$3\cdot10^{-3}$	$<3\cdot10^{-7}$	$<5\cdot10^{-3}$	$<7\cdot10^{-3}$	$7\cdot10^{-2}$

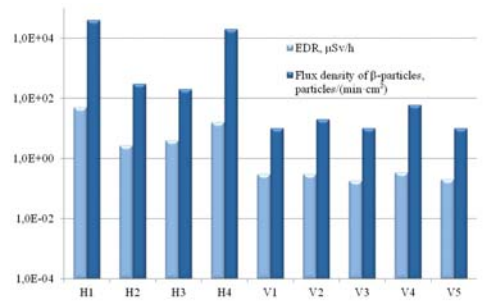


Figure 2. Maximal values of radiation parameters for the period of works

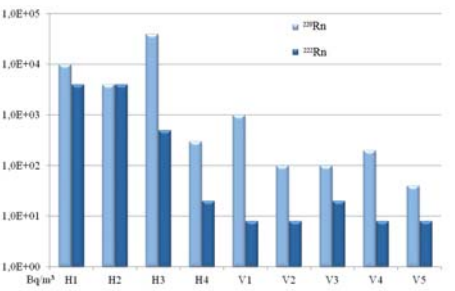


Figure 3. Maximal values of volumetric activity of radon decay products in the air for the period of works

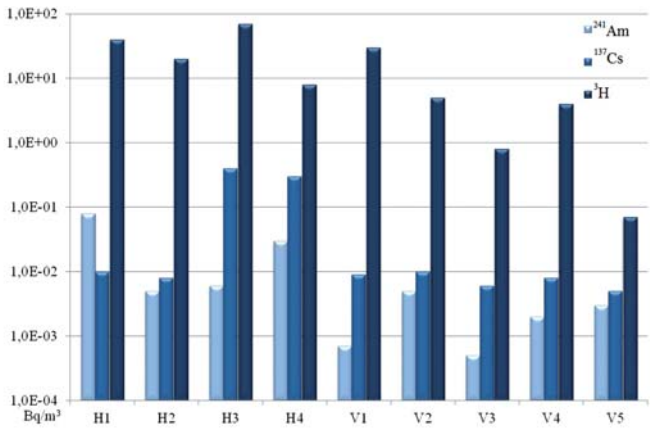


Figure 4. Maximal values of ^{241}Am, ^{137}Cs, ^{3}H volumetric activity in the air for the period of works

Measured radiological parameters in places of studies show that the tunnels where horizontal method was employed demonstrated higher readings of monitored radiological parameters.

3. MAIN DOSE FORMING FACTORS

Effective dose during the works at the tunnels consists of external exposure dose and internal dose from radionuclides intake by people at contaminated areas.

According to initial data we can assume that currently possible sources of radioactive contamination in the tunnels of "Degelen" site are fission products of ^{239}Pu, the remnants of fissile materials and activation products [2].

Computational data of probable specific activity allow us to suppose that possible release of radionuclides from the tunnel to the surface the area of tunnel and adjacent areas may result in contamination with significant concentrations of the following main radionuclides:

- fission products – ^{90}Sr, ^{90}Y, ^{121}Sn-m, ^{137}Ba-m, ^{151}Sm, ^{155}Eu, ^{99}Tc, ^{137}Cs;
- remnants of fissile material – ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am;
- activation products – ^{60}Co, ^{152}Eu.

3.1. Factors influencing the formation of doses from external sources of ionizing radiation (SIR)

Formation of doses from external SIR involves [3]:

- natural radionuclides in soils and rocks;
- natural decay products of natural radon in the atmosphere;
- artificial radionuclides in soils and rocks.

Gamma- and beta-emitting radionuclides have major influence on the dose formation from external radiation sources. Under the conditions of work at "Degelen" tunnels all of the listed radionuclides will contribute to the formation of the dose from external exposure, but the main effect (given the energy emitted by the gamma and beta rays, and expected residual activity, taking into account the half-life) would come from ^{90}Sr, ^{90}Y, ^{137}Cs, ^{137m}Ba.

3.2. Factors contributing to dose formation at internal exposure

Internal exposure, particularly from the alpha- and beta-emitting radionuclides, places much greater biological hazard than the external one. It is formed due to radionuclide intake into body and their subsequent retention in organs and tissues during some time determined by ability of this radionuclide to retain in the body and to be excreted out of the body [4]. Thus, for example, the half-period of ^{137}Cs is about 70 days, ^{241}Am - about 50 years, tritium from the body free water - about 10 days [5].

Specifics of the works at the tunnels (radiation-hazardous facilities) are subject to compliance with measures to ensure radiation safety preclude oral intake of radionuclides, so in these conditions, the main source of internal dose formation is the inhalation intake of radionuclides.

Mechanism of internal dose formation will depend on the concentrations of these radionuclides in the air and on the use of respiratory protection. However, given the dose coefficients for all of radionuclides listed, contamination with which is possible in these conditions (results of calculations based on initial data), with an equal volumetric activity in air, it can be assumed that isotopes of Pu, ^{241}Am and ^{90}Sr will have major influence on the dose. Also, when calculating doses from inhalation, it is necessary to consider the effect of tritium, radon and its decay products.

4. METHODOLOGY FOR CALCULATION OF DOSES

Since fissionable nuclear materials in the tunnels have not been found in significant quantities, we did not consider neutron radiation at dose assessment.

Overall total effective dose (when working at the tunnels at "Degelen" site) is defined as the sum of the following components [6]:

$$E = E_{\gamma} + E_{\beta} + \sum E_{DP} + \sum E_{i(inh)}, \text{ where} \qquad (1)$$

E_{γ} – dose of external γ-radiation;

E_{β} – dose of external β-radiation;

$E_{i(inh)}$ – expected dose from intake of an *i*-radionuclide through respiratory organ (except daughter products of radon and thoron);

E_{DP} – dose from intake of the daughter products of radon and thoron.

At dose calculations we employed a conservative approach. Calculation of doses was carried out during the work and the year.

4.1. Methodology for calculation of doses from external ionizing radiation

Dose from external ionizing radiation sources (SIR) was defined as the sum of the doses of external γ-and β-radiation.

Calculation of doses from external γ-radiation was based on measurements of equivalent dose rate (EDR) at the work site and in the field camp, taking into account the time spent there [6]:

$$E_{\gamma} = P_{\gamma} \cdot t \cdot K_{in}, \text{ where} \quad (2)$$

P_{γ} – measured equivalent dose rate of gamma radiation at the work site and in the field camp;

t – exposure time (7,2 hours - daily work, 16,8 hours - daily rest time);

K_{in} – correction for attenuation of gamma radiation in the human body, taking into account the uneven exposure of different organs (for the calculations it was taken as 1)

Calculation of effective dose from external sources of β-radiation was carried out by measurements; flux of β-particles at the work site and in the field camp - taking into account the time spent there [6]:

$$E_{\beta} = H_{\beta} \cdot W_t \cdot K_o, \text{ where} \quad (3)$$

$H_{\beta}=P_{\beta} \cdot t_{exp}$ – equivalent dose of beta radiation;

P_{β} – equivalent dose rate of beta radiation at the work site and in the field camp;

t_{exp} – exposure time (7.2 hours - daily work, 16.8 hours - daily rest time);

$W_t = 0.01$ - weighting factor for skin;

$K_0 = 0.5$ – attenuation of beta-radiation from clothing.

Calculation of equivalent dose rate of beta radiation is as follows:

$$P_{\beta i} = \sum N_{\beta Ei} \cdot e_{\beta Ei} \cdot \exp\left(-\mu_B \cdot d\right), \text{ where} \quad (4)$$

$N_{\beta Ei}$ – flux of beta-particles with E_i average energy, beta-particles/cm^2;

$\exp(-\mu_b \cdot d)$ – correction for attenuation of beta-particles in air (for the calculations it was taken as 0,9).

$e_{\beta Ei}$ – equivalent per unit fluence of beta-particle E_i of average energy dose from Table 8.4 NRB-99 [7], Sv•cm^2/h.

Of the considered radionuclides β-particles emitting ^{90}Y (2,28 MeV) have the highest energy. Given the conservative approach, to determine equivalent dose per unit fluence in calculating the dose from external sources of β-radiation it was assumed that all β-particles have energy of 2.28 MeV.

4.2. Methodology for calculation of doses due to inhalation of radionuclides

Determination of doses from internal intake of radionuclides for the staff was done for inhalation intake. Calculation of doses from internal intake of artificial radionuclides was determined by computational and analytical method based on the data obtained from laboratory tests of samples of air aerosols and water vapour, measurements of equivalent equilibrium volumetric activity of ^{222}Rn and ^{220}Rn and reference materials on dose coefficients and the annual volume of inhaled air of conditioned human [8].

Expected effective dose from intakes of radionuclides by inhalation (through respiratory organ) is determined by the following formula:

$$E_{i(inh)} = \left(C_{i(ws)} \cdot V_{staff} + C_{i(atm)} \cdot \left(V_{pop} - V_{staff}\right)\right) e_{(inh)i}, \text{ where} \quad (5)$$

$C_{i(atm)}$ – concentration of an i - isotope in the atmosphere of the residential zone, m^3;

$C_{i(ws)}$ – concentration of an *i* - isotope in the air of the work site, m^3;

V_{staff} – volume of air inhaled by staff at the work site (1.41 m^3/h, 10.16 m^3 - during the daily work [7]);

V_{pop} – volume of air inhaled by the population in the atmosphere of residential area (1.41 m^3/h, 23.7 m^3 - during the daily rest [7]);

$e_{(inh)i}$ – dose coefficient of an *i*-isotope when intaken by inhalation specified in NRB-99 in Table P1 for the staff and in Table P2 for the population, Sv/Bq [7].

Effective dose from intake of radon and its decay products E_{DP} is determined by the formula [6]:

$$E_{DP(staff)} = g_{DP(staff)} \cdot \left(V_n \cdot Cp_{ws} + \left(V_н - V_n\right) \cdot \left(Cp_{atm} \cdot 0{,}2 + Cp_{ls} \cdot 0{,}8\right)\right), \text{ where} \qquad (6)$$

$g_{DP(staff)}$ =6,6·10^{-9} Sv/Bq - conditional dose coefficient of ^{222}Rn for staff calculated on the basis of the limit of annual intake for staff;

Cp_{ls}, Cp_{atm} , Cp_{ws} – equivalent equilibrium volumetric activity of ^{222}Rn in living spaces, atmosphere and at the work site, respectively;

0.2; 0.8 – time fractions of being outdoors and indoors, respectively.

Effective dose from intake of ^{220}Rn daughter products is calculated by formula 6 with conventional dose coefficients of ^{220}Rn for staff $g_{DP(staff)}$ =3·10^{-8} Sv/Bq.

Calculation of doses from internal radionuclides inhalation was performed ignoring the respiratory protection. When calculating doses, given the conservative approach, the values of volumetric activities as per the measurements that are below the detection limit, were equated to the detection limit of the measurement devices.

5. ASSESSMENT OF INDIVIDUAL DOSES OF STAFF BY COMPUTATIONAL METHOD BASED ON THE MEASUREMENT OF RADIATION PARAMETERS AT WORK ON CREATION OF ADDITIONAL ENGINEERING PROTECTION

5.1. Dose from external ionizing radiation

Works to create additional protection were carried out on a shift-change work basis. Number of days per shift varied (maximum number of days per one shift was 25). Calculation of dose from external SIR was performed for each shift on the maximum values of radiation parameters registered in the shift at the work site and living space, taking into account the nature of changes in radiation conditions, working and rest times. With minor changes in radiation parameters the calculation doses per shift was held on the maximum values for the period of shift (conservative assessment). In the case of abrupt changes in radiation parameters for the period for shift when calculating we took into account the separate days with high values of radiation parameters. Dose over a period of work was calculated as the sum of the doses for shifts. Assessment of doses for the year was held at doses over the period of works from the assumption that works during the year were carried out under the

same conditions as for the period of work. Results of dose calculation from external SIR are presented in the table (Table 2).

Table 2.

Calculation of doses from external sources of ionizing radiation

Tunnel code	Duration, days	Total dose from external SIR for the period of works, mSv	Dose from external γ-radiation per year, mSv/year	Dose from external β-radiation per year, mSv/year	Total dose from external SIR for the year, mSv/year
H1	140	3.2	8.06	0.27	8.3
H2	149	0.9	2.27	0.02	2.3
H3	160	1.2	2.65	0.02	2.7
H4	105	2.4	7.76	0.75	8.5
V1	80	0.4	1.62	0.01	1.6
V2	86	0.5	1.94	0.01	2.0
V3	72	0.3	1.33	0.01	1.3
V4	95	0.5	2.02	0.02	2.0
V5	42	0.2	1.44	0.01	1.5

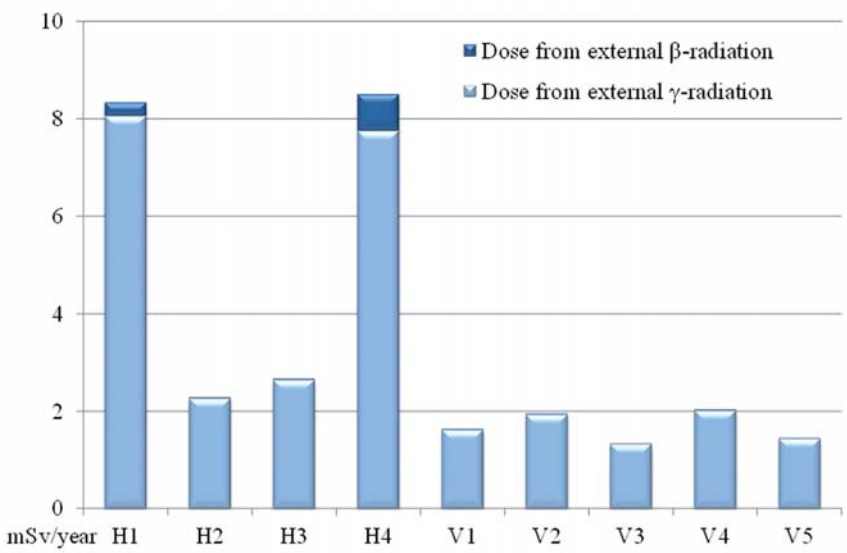
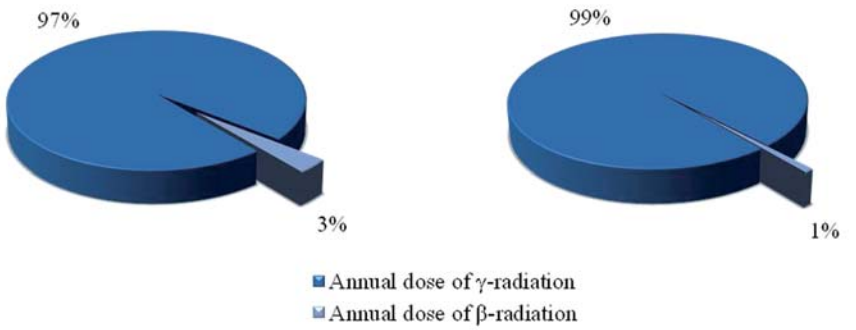


Figure 5. Calculation of doses from external sources of radiation

Calculations of annual doses show that the maximum dose from external SIR could be obtained during the works using horizontal method.

When assessing the contribution of dose from external gamma and beta radiation it is evident that during the vertical method of works the contribution to dose from external beta radiation is negligible at all the tunnels, whereas when working horizontally the contribution from external beta radiation may take significant values (Figure 6, Table 3).



(a) (b)

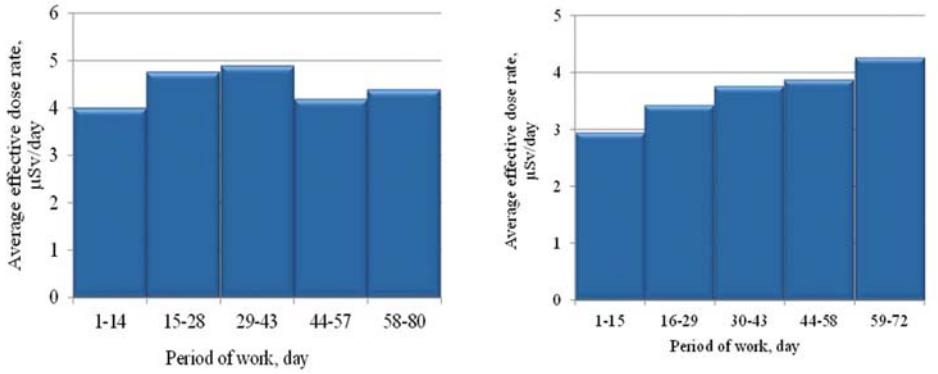
Figure 6. Averaged contribution to the dose from external gamma and beta radiation,
(a) at the tunnels with horizontal method of creating protection,
(b) at the tunnels with vertical method of creating protection

Table 3.

Contribution to dose from external gamma and beta radiation,%

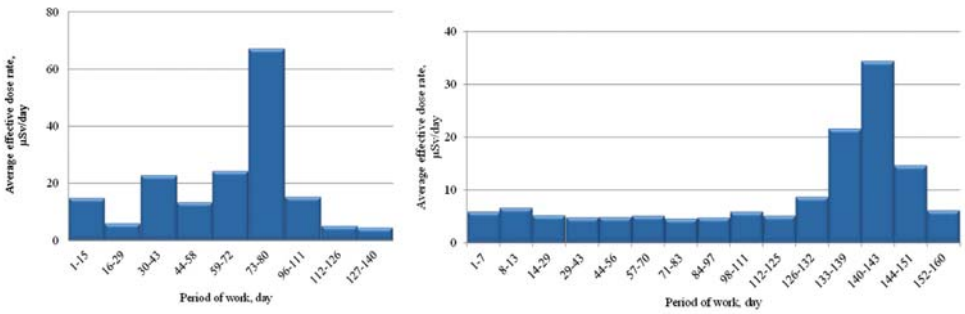
	H1	H2	H3	H4	V1	V2	V3	V4	V5
Contribution to dose from external gamma radiation	97	99	99	91	99	99	99	99	99
Contribution to dose from external beta radiation	3	1	1	9	1	1	1	1	1

Analysis of dose formations from external SIR on periods of dose calculation shows that for the tunnels with the vertical method changing in the average rate of the effective dose is of uniform character throughout the period (Figure 7). Formation of dose from external SIR at the tunnels with the horizontal method of work is not of a uniform character, upon that major formation of dose is in the second half and may be associated with the final stage of work on the construction of additional protection that is characterized by increased radiation hazard (Figure 8).



(a) (b)

Figure 7. Doses from external SIR at the tunnels with the vertical method of work, (a) tunnel V1, (b) tunnel V3



(a) (b)

Figure 8. Formation of doses from external SIR at the tunnels with the horizontal method of work, (a) tunnel H 1, (b) tunnel H 3

5.2. Internal dose from radionuclides

5.2.1. Calculation of internal radiation doses for ^{241}Am, $^{239+240}$Pu, ^{137}Cs +^{137m}Ba, ^{3}H, ^{222}Rn and ^{220}Rn

Calculation of doses due to inhalation of radionuclides was carried out using the measured values of ^{241}Am, ^{137}Cs +^{137m}Ba, ^{3}H, ^{222}Rn and ^{220}Rn. Calculation of doses due to inhalation of $^{239+240}$Pu was carried out both on the basis of the measured values, and on the basis of estimated data on the calculated ratio of $^{239+240}$Pu/^{241}Am for each tunnel. Calculation of doses due to inhalation of radionuclides is similar to the calculation of external SIR. Calculation was performed for each shift using the maximum values of radiation parameters registered in the shift at work site and living space, given the nature of changes in radiation conditions and time of work and rest. With minor changes in the radiation parameters, dose calculation was held using maximum values for the period of shift (conservative assessment). In the case of abrupt changes of radiation parameters for the period of shift when calculating there were taken into account separate days with high values of radiation parameters. Dose over a period of work was calculated as the sum of the doses for the shifts. Assessment of doses for the year was held using doses for the period of works from the assumption that works during the year were carried out under the same conditions as for the period of work. Results of dose calculation from are presented in the table (Table 4).

Table 4.

Calculation of doses from inhalation intake of radionuclides

Tunnel notation	Duration, days	Total dose from inhalation intake for the period of works, mSv	Total dose from inhalation intake for the period of year, mSv/year
H1	140	13.9	36
H2	149	4.0	10
H3	157	19.4	44
H4	104	4.0	14
V1	80	1.0	4

Tunnel notation	Duration, days	Total dose from inhalation intake for the period of works, mSv	Total dose from inhalation intake for the period of year, mSv/year
V2	86	1.4	6
V3	72	1.3	7
V4	95	1.1	4
V5	42	0.6	5

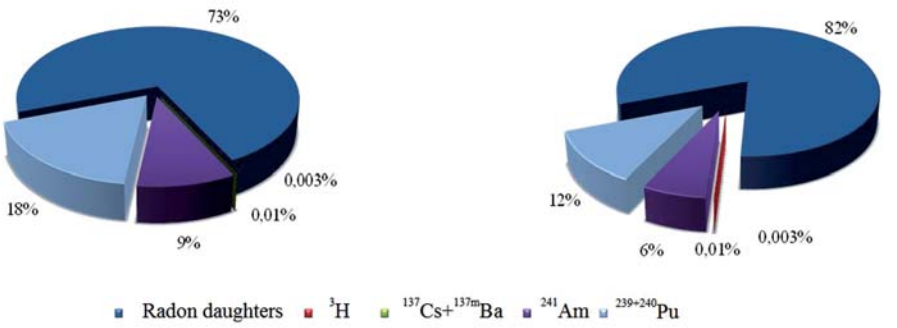
Analysis of different radionuclides contribution to dose formation shows that ^{222}Rn, ^{220}Rn, $^{239+240}$Pu and ^{241}Am have the major influence on the dose (Table 5).

Table 5.

Contribution of radionuclides to the formation of doses for various tunnels,%

	H1	H2	H3	H4	V1	V2	V3	V4	V5
Radon daughters	67	94	96	36	87	65	92	92	73
^{3}H	0.007	0.012	0.002	0.003	0.05	0.007	0.002	0.006	0.001
^{137}Cs+^{137m}Ba	0.0003	0.0009	0.002	0.02	0.002	0.008	0.0004	0.0002	0.002
^{241}Am	12	2	1	22	5	12	3	3	9
$^{239+240}$Pu	22	4	3	42	9	23	5	5	17

Calculation of the average contribution of different radionuclides to dose formation for tunnels with horizontal and vertical methods of work is shown in the figure (Figure 9).



(a) (b)

Figure 9. Averaged contribution from different radionuclides to the dose (a) at the tunnels with horizontal method of creating protection, (b) at the tunnels with vertical method of creating protection

Analysis of the averaged contribution of different radionuclides showed that mechanism of dose formation is similar for tunnels with different methods of works to create additional engineering protection.

5.2.2. Assessment of individual doses of staff from ^{241}Pu intake

Assuming that the ratio ^{241}Pu/$^{239+240}$Pu will be invariably in soil and air aerosols for one research subject, we can assess the dose from intakes of ^{241}Pu by measuring the specific activity of $^{239+240}$Pu.

According to the results of research conducted in IRSE NNC, it is known that the ratio $^{241}Pu/^{239+240}Pu$ for the various tunnels is ranging from 1.7 to 130, therefore, one cannot apply a uniform recalculation coefficient of the specific activity of ^{241}Pu by the specific activity of $^{239+240}Pu$. However, to assess the possible contribution to dose formation due to intake of ^{241}Pu we calculated doses for all the tunnels by the well-known ratio $^{241}Pu/^{239+240}Pu$ (3.5), defined for the tunnel B1. Dose values obtained from intake of ^{241}Pu and their comparative analysis with the doses due to ^{241}Am, are represented in the histogram (Figure 10).

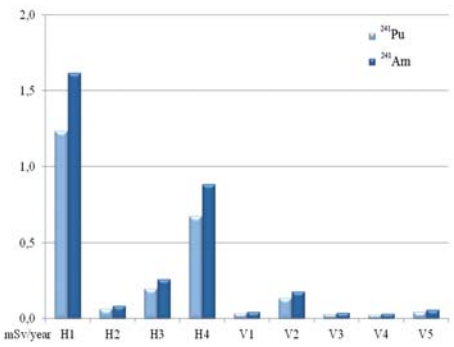


Figure 10. Contribution to the formation of doses for various tunnels due to internal intake of ^{241}Pu and ^{241}Am

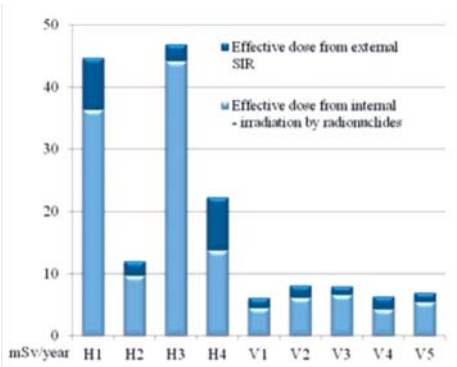


Figure 11. Calculation of total effective doses

Based on the comparison of doses from internal intake of ^{241}Pu and ^{241}Am it can be concluded that even with relatively small values of the ratio $^{241}Pu/^{239+240}Pu$ (3.5), doses from ^{241}Pu are comparable to doses from ^{241}Am. In the case of increasing values of this ratio up to the highest registered to date at "Degelen" (130) doses from internal intake of ^{241}Pu will greatly exceed the dose from intake of ^{241}Am.

5.2.3. Assessment of the contribution to individual doses received by personnel due to intake of other isotopes

To assess the contribution to dose formation from internal intake of radionuclides, which may contaminate the tunnels and adjacent areas in significant concentrations, but measurement of volumetric activity in air which has not been ($^{90}Sr+^{90}Y$, ^{121}Sn-m, ^{151}Sm, ^{155}Eu, ^{99}Tc, ^{238}Pu, ^{242}Pu), based on initial data on the test in the tunnels we determined their ratio at the moment and radionuclides, of which there are data on the volumetric activity in the air during the works (^{241}Am and $^{137}Cs+^{137m}Ba$). Based on these ratios we calculated the potential volumetric activities of $^{90}Sr+^{90}Y$, ^{121}Sn-m, ^{151}Sm, ^{155}Eu, ^{99}Tc, ^{238}Pu, ^{242}Pu. Calculation of radionuclide activity in the air was carried out using the maximum measured values of volumetric activity of ^{241}Am and ^{137}Cs in the air for each tunnel. Calculation of doses was carried out given a conservative approach, at the highest possible concentrations at a constant intake of radionuclides during the year.

Results of annual dose calculations are presented in tables (Table 6, Table 7).

Table 6.

Calculated values of doses based on measured values of $^{137}Cs+^{137m}Ba$, mSv/year

	H1	H2	H3	H4	V1	V2	V3	V4	V5
$^{90}Sr+^{90}Y$	$8\cdot10^{-3}$	$7\cdot10^{-3}$	$3\cdot10^{-1}$	$3\cdot10^{-1}$	$8\cdot10^{-3}$	$8\cdot10^{-3}$	$5\cdot10^{-3}$	$7\cdot10^{-3}$	$4\cdot10^{-3}$
^{99}Tc	$1\cdot10^{-5}$	$8\cdot10^{-6}$	$4\cdot10^{-4}$	$3\cdot10^{-4}$	$9\cdot10^{-6}$	$1\cdot10^{-5}$	$6\cdot10^{-6}$	$8\cdot10^{-6}$	$5\cdot10^{-6}$
^{155}Eu	$5\cdot10^{-7}$	$4\cdot10^{-7}$	$2\cdot10^{-5}$	$2\cdot10^{-5}$	$5\cdot10^{-7}$	$5\cdot10^{-7}$	$3\cdot10^{-7}$	$4\cdot10^{-7}$	$3\cdot10^{-7}$
^{121m}Sn	$3\cdot10^{-7}$	$3\cdot10^{-7}$	$1\cdot10^{-5}$	$1\cdot10^{-53}$	$3\cdot10^{-7}$	$3\cdot10^{-7}$	$2\cdot10^{-7}$	$3\cdot10^{-7}$	$2\cdot10^{-7}$
^{151}Sm	$5\cdot10^{-5}$	$4\cdot10^{-5}$	$2\cdot10^{-3}$	$1\cdot10^{-3}$	$5\cdot10^{-5}$	$5\cdot10^{-5}$	$3\cdot10^{-5}$	$4\cdot10^{-5}$	$2\cdot10^{-5}$
Total dose	$8\cdot10^{-3}$	$7\cdot10^{-3}$	$3\cdot10^{-1}$	$3\cdot10^{-1}$	$8\cdot10^{-3}$	$8\cdot10^{-3}$	$5\cdot10^{-3}$	$7\cdot10^{-3}$	$4\cdot10^{-3}$

Table 7.

Calculated values of doses based on measured values of ^{241}Am, mSv/year

	H1	H2	H3	H4	V1	V2	V3	V4	V5
^{242}Pu	$2\cdot10^{-2}$	$1\cdot10^{-3}$	$1\cdot10^{-3}$	$7\cdot10^{-3}$	$2\cdot10^{-4}$	$1\cdot10^{-3}$	$1\cdot10^{-4}$	$5\cdot10^{-4}$	$7\cdot10^{-4}$
^{238}Pu	$2\cdot10^{1}$	$1\cdot10^{0}$	$1\cdot10^{0}$	$7\cdot10^{0}$	$2\cdot10^{-1}$	$1\cdot10^{0}$	$1\cdot10^{-1}$	$5\cdot10^{-1}$	$7\cdot10^{-1}$
Total dose	$2\cdot10^{1}$	$1\cdot10^{0}$	$1\cdot10^{0}$	$7\cdot10^{0}$	$2\cdot10^{-1}$	$1\cdot10^{0}$	$1\cdot10^{-1}$	$5\cdot10^{-1}$	$7\cdot10^{-1}$

Calculations showed that $^{90}Sr+^{90}Y$ and ^{238}Pu have the greatest influence on the dose formation at internal intake. Maximum values of annual doses at a constant intake of radionuclides were $^{90}Sr+^{90}Y$ – 0.3 mSv/year, ^{238}Pu - 20 mSv/year (under the same conditions dose of ^{241}Am is 25 mSv/year, $^{137}Cs+^{137m}Ba$ – $4\cdot10^{-4}$ mSv/year).

Thus, one cannot neglect the contribution of ^{238}Pu, as its contribution to dose formation is comparable with the contribution from ^{241}Am.

Total contribution to the dose from internal intake of $^{90}Sr+^{90}Y$, ^{121}Sn-m, ^{151}Sm, ^{155}Eu, ^{99}Tc according conservative estimate does not exceed 0.5 mSv/year, upon that 99% of the dose is formed by intake of $^{90}Sr+^{90}Y$.

6. EXPECTED TOTAL EFFECTIVE DOSES

Calculation of total effective doses for the period of works and for the year is presented in Table 8 and Figure 11.

Table 8.

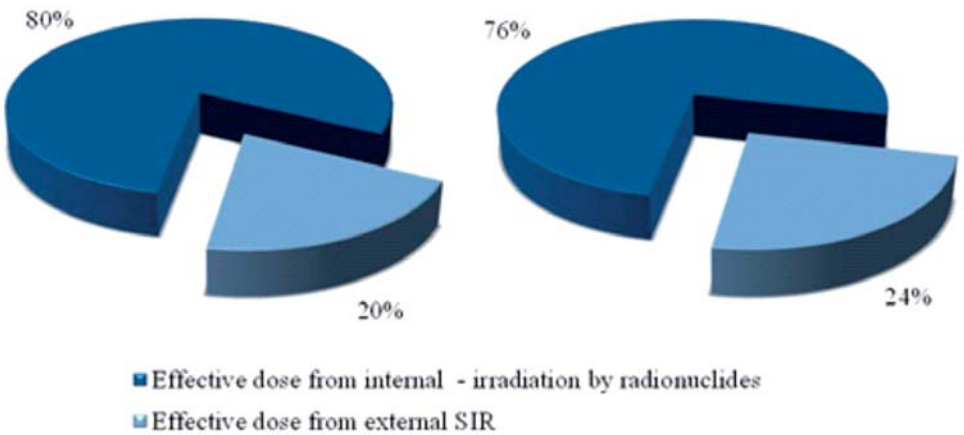
Calculation of total effective doses

Tunnel code	Total dose from inhalation intake of for, the period of works, mSv	Total dose from inhalation intake of for the year, mSv/year	Total dose from external SIR for the period of works, mSv	Dose from external γ-radiation per year, mSv/year	Total effective dose for the period of works, mSv	Total effective dose per year, mSv/year
H1	13.9	36	3.2	8.3	17.1	44.3
H2	4	10	0.9	2.3	4.9	12.3

Tunnel code	Total dose from inhalation intake of for, the period of works, mSv	Total dose from inhalation intake of for the year, mSv/year	Total dose from external SIR for the period of works, mSv	Dose from external γ-radiation per year, mSv/year	Total effective dose for the period of works, mSv	Total effective dose per year, mSv/year
H3	19.4	44	1.2	2.7	20.6	46.7
H4	4	14	2.4	8.5	6.4	22.5
V1	1	4	0.4	1.6	1.4	5.6
V2	1.4	6	0.5	2	1.9	8.0
V3	1.3	7	0.3	1.3	1.6	8.3
V4	1.1	4	0.5	2	1.6	6.0
V5	0.6	5	0.2	1.5	0.8	6.5

Calculation of effective dose for the works on the construction of additional engineering protection showed that annual doses, which can be intaken during the works using horizontal method, is significantly higher than those intaken during the vertical method works. Carrying out of works (under conditions similar to tunnel H1 and H2) without use of personal respiratory protection for 3 years can lead to overexposure of personnel (132-138 mSv for 3 years with allowed 100 mSv in any consecutive 5 years).

Calculation was done on the averaged contribution to dose formation from external sources of ionizing radiation and internal intake of radionuclides for tunnels with horizontal and vertical methods of work (Figure 12, Table 9).



(a) (b)

Figure 12. Averaged contribution to dose formation from external sources of ionizing radiation and internal intake of radionuclides, (a) tunnels with horizontal method of creating protection, (b) tunnels with vertical method of creating protection

Table 9.

Contribution to dose formation from external SIR and internal intake of radionuclide, %

	H1	H2	H3	H4	V1	V2	V3	V4	V5
Contribution to dose from external gamma radiation	81	81	94	62	73	76	83	68	79
Contribution to dose from external beta radiation	19	19	6	38	27	24	17	32	21

According to the calculation of contribution to dose formation from external sources of ionizing radiation and internal intake of radionuclides one can conclude that basic dose forming is due to internal intake for both horizontal method of creating additional protection and vertical one.

Works on the construction of additional engineering protection at the tunnels of "Degelen" site are carried out with the mandatory use of personal respiratory protection. Calculation of total annual effective dose was done, assuming that the minimum multiplicity of attenuation of the maximum allowable concentration of the personal protective equipment is equal to 10 (Table 10).

Table 10.

Calculation of total effective dose, taking into account the use of personal respiratory protection, mSv/year

Tunnel code	Total dose from inhalation intake	Total dose from external SIR	Total effective dose
H1	3.6	8.3	11.9
H2	1	2.3	3.3
H3	4.4	2.7	7.1
H4	1.4	8.5	9.9
V1	0.4	1.6	2
V2	0.6	2	2.6
V3	0.7	1.3	2
V4	0.4	2	2.4
V5	0.5	1.5	2

According to the results of total annual effective dose calculation, taking into account the use of personal respiratory protection, we can conclude that the maximum doses that might be untaken during the works to create additional engineering protection of the facilities under consideration will not exceed the doses to staff of the category A (20 mSv/year).

7. COMPARISON OF MEASURED DOSES FROM EXTERNAL SIR WITH CALCULATED DOSES

Calculated values of dose from external SIR were compared with measured doses of external radiation sources for some of the tunnels. Measurement of doses was carried out using thermoluminescent dosimeters. Results of the comparison are presented in the histogram (Figure 13).

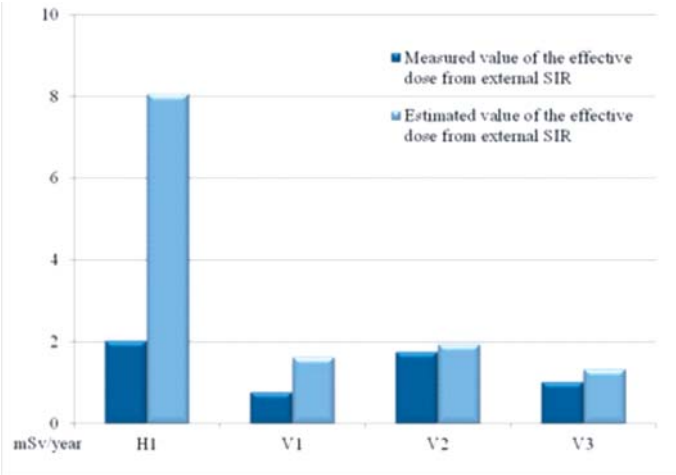


Figure 13. Comparison of the measured dose from external SIR with the calculated doses

The histogram shows that the calculated dose from external SIR in the vertical method of creating protection in most cases almost coincide with the measured values, whereas the measured value in the horizontal method of creating protection is 4 times less than the calculated value. This fact can be explained by the changes in the radiation situation at the site of conducting operations during various methods of creating protection:

- when applying a vertical method of creating protection the changes in the radiation environment is of uniform character, the values of radiation parameters that affect the formation of doses from external SIR, virtually do not change over time.
- when applying a horizontal method of creating protection the change in radiation situation is uneven, the values of radiation parameters change over time in a range of up to 2 orders of magnitude.

Since the calculation of the dose was performed using a conservative approach, i.e. supposing that person resides in a place with the highest values of radiation parameters, given the nature of the change in the radiation situation, the calculated value of dose from external SIR in the horizontal method of creating protection will be significantly higher than the measured doses, whereas in the vertical method of creating protection the calculated doses would be comparable with measured ones.

CONCLUSIONS

1. In assessing the dose from external SIR it is needed to consider the contribution from external beta radiation when working in tunnels with the horizontal method of creating protection.
2. At horizontal method of protection, the basic dose from external SIR is formed during the final phase of works, connected directly to the construction of additional protection and is characterized by increased radiation hazard.

3. Mechanism of dose formation from internal intake of radionuclides is the same as for both horizontal method for creating protection and the vertical one.
4. When assessing the doses from internal intake of radionuclide the influence of $^{137}Cs+^{137m}Ba$ and ^{3}H on dose can be neglected since their contribution does not exceed 0.01% of the total dose.
5. Natural radionuclides ^{222}Rn and ^{220}Rn have the greatest influence on the dose from inhalation intake of radionuclides. But we should not ignore the influence of such artificial radionuclides as ^{241}Am and $^{239+240}Pu$ on the formation of doses from inhalation.
6. Most expeditious and cost-effective way to assess the doses at internal intake of $^{239+240}Pu$ is the computational method by the measured values of ^{241}Am volume activity in the air.
7. When carrying out works to create additional engineering protection the contribution from ^{241}Pu must be taken into account. The contribution from ^{241}Pu can be counted by calculation of the measured ^{241}Am, it requires to measure ^{241}Pu in soil samples to calculate the ratio $^{241}Pu/^{241}Am$ at the preliminary examination.
8. For a more accurate assessment of doses from internal exposure it is needed to lower the detection limit of ^{241}Am (by increasing the time of aspiration, changes in sample preparation and measurement of all filters at the end of the works with the calculation of the average volumetric activity for the entire period of work).
9. When assessing the doses from internal intake of radionuclides it is necessary to consider the contribution of ^{238}Pu, as its contribution to the dose is significant. Contribution to the dose from ^{238}Pu can be taken into account by calculation of the measured ^{241}Am, it requires to measure ^{238}Pu in soil samples to calculate the ratio $^{238}Pu/^{241}Am$ at the preliminary examination.
10. To eliminate the systematic error in estimating doses it is necessary to take into account the contribution from the internal intake of $^{90}Sr+^{90}Y$.

Recommendations for optimization of radiation safety at work on the construction of additional engineering protection at the tunnels of "Degelen" site in order to prevent overexposure of personnel

1. Reduce the frequency of sampling to estimate the doses from internal intake of radionuclide ^{137}Cs $+^{137m}Ba$ and ^{3}H to 1 time per week.
2. For a more rapid assessment of doses from internal intake of both $^{239+240}$ Pu and ^{241}Pu to calculate the expected volumetric activity of both $^{239+240}$ Pu and ^{241}Pu in the air by the measured values of ^{241}Am volumetric activity in the air.
3. In a preliminary survey to measure both $^{239+240}Pu$ and ^{241}Pu in the soil samples to calculate ratios of $^{239+240}Pu/^{241}Am$, $^{241}Pu/^{241}Am$, $^{238}Pu/^{241}Am$.
4. Since the ratios of $^{239+240}Pu/^{241}Am$, $^{241}Pu/^{241}Am$ и $^{238}Pu/^{241}Am$ may vary when creating protection using horizontal method as far as penetration deep into the tunnel, it is necessary to re-determine these ratios during the key stages (opening the tube, opening the terminal cavity).
5. For a more accurate assessment of doses from internal exposure it is needed to lower the detection limit of ^{241}Am by increasing the volume of samples taken up to 1000 m^3.

6. Assessment of doses from internal intake of $^{90}Sr+^{90}Y$ by determining their volumetric activity using instrumental methods.

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"ДЕГЕЛЕҢ" АЛАҢЫ ШТОЛЬНЯЛАРЫНДАҒЫ ИНЖЕНЕРЛІК ҚҰРЫЛЫСТАРДЫҢ ҚОСЫМША ҚОРҒАУЫН ҚҰРУ БОЙЫНША ЖҰМЫСТАРДЫ ЖҮРГІЗУ КЕЗІНДЕГІ ЖЕКЕ ДОЗАНЫ ҚАЛЫПТАСТЫРУ

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Мақалада "Дегелең" алаңы штольняларындағы инженерлік құрылыстардың қосымша қорғауын құру бойынша жұмыстарды жүргізу кезіндегі қызметкердің жеке дозасын есептеу нәтижесі ұсынылған. Қосымша инженерлік қорғау құрылысы бойынша жұмыстарды жүргізу кезіндегі жеке дозаны қалыптастыруға әсер ететін негізгі факторлар анықталып, қызметкерге иондық сәулеленудің сыртқы көздерінен дозалық жүктемесі және радионуклидтердің ингаляциялық түсуінің салдары есептелген, қосымша инженерлік қорғау құрылысы бойынша жұмыстарды жүргізу кезіндегі радиациялық қауіпсіздікті қамтамасыз етуді оңтайландыру бойынша ұсыныстар әзірленген.

Түйін сөздер: полигон, «Дегелең», радионуклидтер, инженерлік құрылғыларды қорғау, штольня, радиациялық жағдай, радиациялық параметрлер, дозалардың қалыптасуы, сыртқы сәулелену, ішкі сәулелену.

ФОРМИРОВАНИЕ ИНДИВИДУАЛЬНЫХ ДОЗ ПРИ ПРОВЕДЕНИИ РАБОТ ПО СОЗДАНИЮ ДОПОЛНИТЕЛЬНОЙ ЗАЩИТЫ ИНЖЕНЕРНЫХ СООРУЖЕНИЙ НА ШТОЛЬНЯХ ПЛОЩАДКИ "ДЕГЕЛЕН"

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В статье представлены результаты расчетов индивидуальных доз персонала при проведении работ по созданию дополнительной защиты инженерных сооружений на штольнях площадки "Дегелен". Определены основные факторы, влияющие на формирование индивидуальных доз при проведении работ, проведен расчет дозовых нагрузок на персонал от внешних источников ионизирующего излучения и вследствие ингаляционного поступления радионуклидов, разработаны рекомендации по оптимизации процесса обеспечения радиационной безопасности при проведении работ по сооружению дополнительной инженерной защиты.

Ключевые слова: полигон, "Дегелен", радионуклиды, защита инженерных сооружений, штольня, радиационная обстановка, радиационные параметры, формирование доз, внешнее облучение, внутреннее облучение.

УДК 577.4: 504.064:539.16

RADIOECOLOGICAL SITUATION IN THE TERRITORY OF KARADZHAL FLUORITE DEPOSIT

S.A. Aidarkhanov, S.N. Lukashenko, S.B. Subbotin, V.I. Edomin

This paper presents multi-year radioecological research in the territory of Karadzhal fluorite deposit, located in the immediate vicinity of the underground nuclear explosions at the Semipalatinsk test site. It was determined that the content of artificial radionuclides in the surface area of the deposit is due to, mainly, global fallout. The values of radiation parameters, the content of radionuclides in soil, air, products of the deposit are safe. No radioactive contamination of equipment, vehicles, and premises for the staff was found. Main problematic issue is flow of groundwater contaminated with tritium to the boundaries of the deposit. Tritium concentration exceeds the intervention level several times. Also traces of radionuclides ^{137}Cs and ^{90}Sr in groundwater were found. With further development of the deposit we can not exclude the possibility of increase of radionuclides inflow into this deposit. There were given recommendations on creation of network of observation wells in order to obtain a reliable forecast of the radiation environment at the deposit.

Key words: deposit, "Degelen" test site, hydrogeology, radionuclide contamination, monitoring, underground nuclear explosions.

INTRODUCTION

This paper presents data of multi-year radioecological researches in the territory of Karadzhal fluorite deposit located in the immediate vicinity of the northern boundary of "Degelen" test site (Fig. 1) – one of underground nuclear explosion (UNE) test sites at the Semipalatinsk test site (STS) [1].

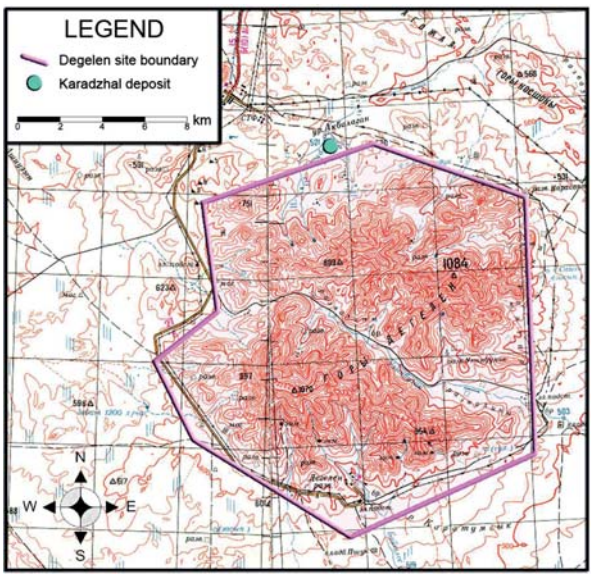

Figure 1. Location of Karadzhal deposit relative to "Degelen" test site

1. GENERAL INFORMATION ABOUT THE DEPOSIT

1.1. Physical and geographic characteristic of the area

Karadzhal deposit is located in East-Kazakhstan region in the territory of the former STS in 170 km to south-west from Semey city and in 120 km to south-west from Kurchatov city.

The deposit area is practically unpopulated. The nearest settlement Sarzhal is located in 60 km to south-east from the deposit. At a distance of 6 km there is a hard-surface road connecting "Degelen" test site with Kurchatov city.

Climate is sharply continental, arid with continuous wind. Continuous wind conditions high evapouration of water, and in cold season the redistribution of snow over the territory takes place. Snow is piled up in ravines of upland, especially in bush growing places. Evapouration from water surface reaches 1100 – 1600 mm a year.

1.2. Subsurface geology of the deposit

Main feature of Karadzhal deposit is its location in near-contact zone of Degelen mountain massif exerted a contact impact on effusive and sedimentary rocks of Paleozoic age. Relief of ore zone of north-western strike, where the deposit is confined to, is a gently ridgy plain having general slope to center of the deposit, where feebly marked bed of Karabulak River runs from the south to the north.

Karadzhal fluorite deposit was formed in outlier of sedimentary rocks among granites in the northern endo-contact of Degelen complex. Rocks composing this outlier are presented by sedimentary deposits, including carbonate ones of the Famennian suite that were intensively metamorphized and underwent a complex metasomatic transformations as a result of contact impact of granites. Commercial mineralization is presented by 64 quartz-fluorite ore bodies, among which 19 bodies concentrate main deposit reserves (92 %). Dip of ore bodies is traced to 10-80 m and they wedging-out by approaching to granites and in marbles slightly crossing the contacts of these rocks.

Approved by the State Committee for Reserves of the Republic of Kazakhstan, ore reserves by categories C1+C2 make up 1,545.6 thous. tons with average content of fluorspar 49.94%. Reserves are distributed non-uniformly by depth – their main part is deposited at depths up to 30 m (Fig. 2).

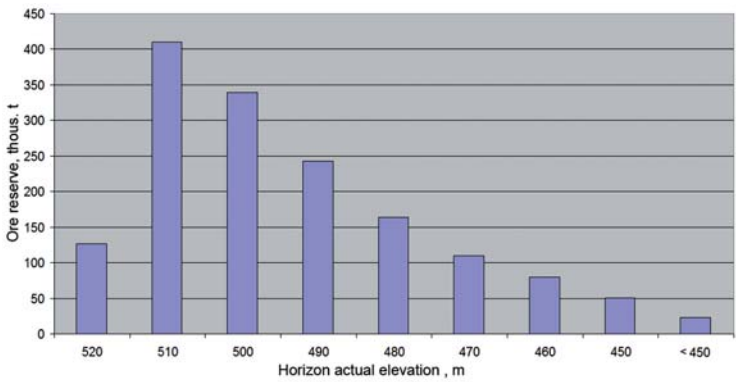

Figure 2. Distribution of the projected ore reserves of Karadzhal deposit by depth

1.3. Hydrography and hydrogeology of the area

Hydrogeological conditions of Karadzhal deposit are typical for territories of central Kazakhstan hummocky topography: small amount of atmospheric fallouts (170–460 mm a years, mean annual – 263 mm, in high-water year – 382 mm, in low-water year – 221 mm), prevalence of evapouration over precipitations, seasonal flow of surface waters that is not observed every year. Major part of surface flow takes place during the snowmelt period – April-May.

Degelen mountain massif is a part of regional hydrogeological system of left-bank of Irtysh river and is an area of recharge and transit of underground waters at the expense of infiltration of atmospheric fallouts. Besides, water-bearing horizons are partially discharged here through springs. There are pore, fracture and vein types of underground waters within the mountain massif.

Pore waters of diluvial-proluvial deposits are confined to crushed stone rocks with sand-clay filler that compose intermontane trails of debris cone, as well as submountain trail rimming the mountain massif. They are deposited among boulder-pebbles and sands filling the well stripped valleys of rivulets Uzynbulak, Karabulak, Baitles, Toktakushik and others. Depth of pore waters depends from season hydro-meteorological conditions. During the period of spring snowmelt and abundant summer rains their level is increased so much that they give birth to seasonal springs. In drought season the level is decreased to 2–6 m, and in submountain trails – to 10–12 m. These waters are recharged at the expense of infiltration of atmospheric fallouts through aeration zone.

Fracture waters are confined to exogenous fracture zone in upper part of Paleozoic foundation, in some valleys they are blocked by Neogene clays causing pressure buildup. In some cases the head of water exceeds the absolute mark of area and it results in self-discharge of wells (well 385 in valley of rivulet Baitles).

Vein type of waters is confined to tectonic deformations having hyperpermeability. All types of mentioned waters are hydraulically connected between each other at the major part of the territory and form an integrated hydrodynamic system.

About 15 active springs were found out at "Degelen" site and water discharges vary in the range from 0,5-0,6 dm^3/min to 1000 dm^3/min. Most watery springs are located at a height of 545-650 m. They are connected with main water-bearing fractures and are discharge points of fracture-vein waters. Besides the above-mentioned springs, there is a large quantity of seasonal springs (~30). Hydrophilous vegetation, soil moistening and erosional cuttings indicate on seasonal character of springs.

Karabulak rivulet, its bed cut by quarry being under development, is formed by junction of 3 waterways originated in northern flanks of Degelen mountains (Fig. 3). The first and second waterways flow together before the quarry, the joined waterway runs along the south boundary of the quarry via special bypass drainage trench. The third waterway runs to the east of the quarry territory and only then it joins with Karabulak rivulet.

According to long-term observations, watercourse in Karabulak rivulet takes place only during the short-term period of snowmelt. Riverbed is indistinct, often it does not appear at all – there is no cutting in weak soil and vegetable cover. Riverbed disappears in submountain trails at a distance of 2.5 km after the quarry and does not fall into a single waterway.

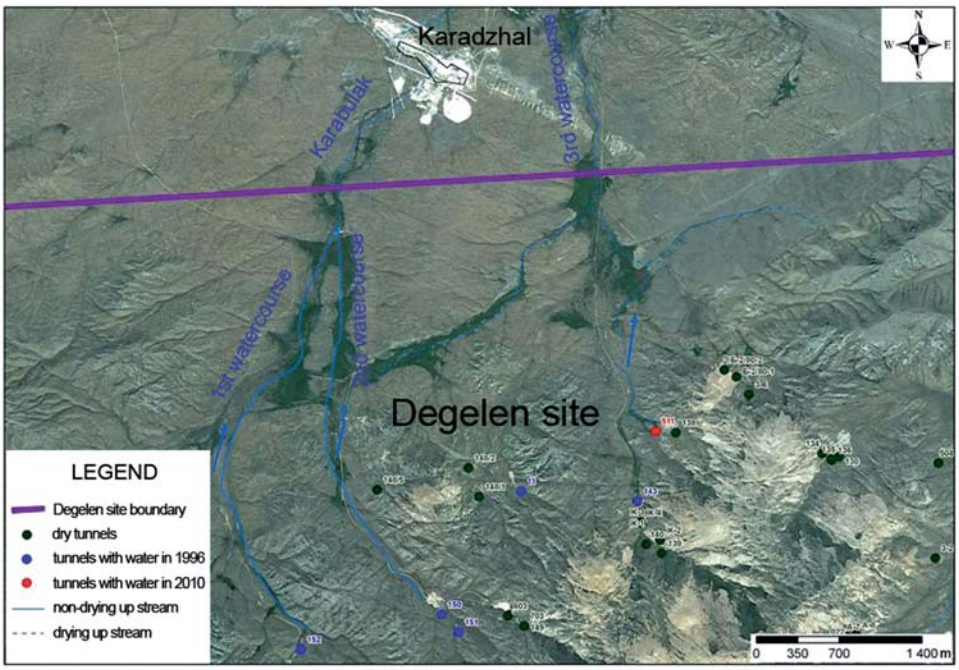

Figure 3. Location of Karadzhal deposit site and main sources of potential migration of artificial radionuclides with surface and underground waters.

1.4. Deposit development features

Shallow bedding of ore predetermines the open-cut deposit mining. The following deposit mining scheme was accepted – all-work lateral single-side with the mixed arrangement of dump. Front of overburden and winning operations moves parallel to the short axis of quarry field. Deposit development is carried out with application of drilling and blasting operations in the following order:

- drilling and blasting operations using drilling machine BTS-150B;
- excavation and loading of ore and overburden using excavators EO-5225, ET-25 and ET-18 having bucket capacity 2.5 m^3, 1.25 m^3 and 0.75 m^3, respectively;
- transportation of ore to ore storage using dump truck KrAZ of capacity 12 t;
- dumping of overburden and grading operations using bulldoser T-170.

Potential output of the quarry is 103 thous. tons of ore a year. Taking into account the stated output, the deposit operation life will be 16 years. Project quarry depth is 70 m.

General view of Karadzhal deposit quarry is presented in figure (Figure 4).

Figure 4. General view of Karadzhal deposit quarry.

Main elements of infrastructure of Karadzhal deposit are presented in figure (Figure 5).

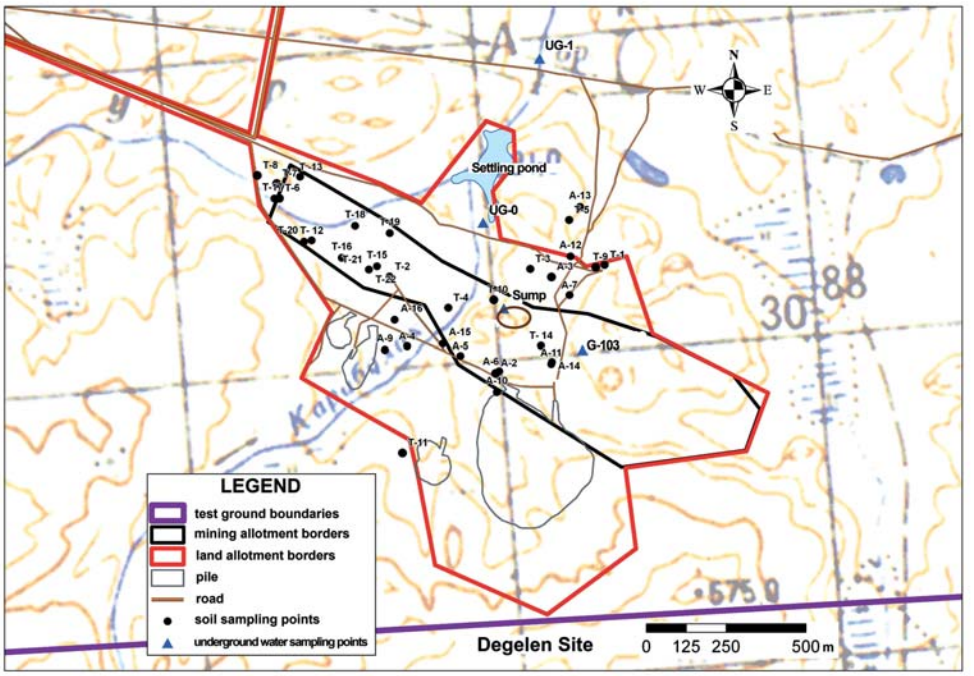

Figure 5. Main elements of infrastructure of Karadzhal deposit

Quarry is dewatered using open spillway. Quarry water formed at the expense of rain water, melt water, as well as inflow of underground water, is accumulated in a water header – a sump passing from horizon 510 m to 505 m. Volume of sump is 630 m^3, depth – 5 m, slope angles of sides are up to 30 degrees. A pumping unit equipped with three pumps CNS 38-88 is used for pumping water out of sump. Normal inflow makes up 50 m^3/h, and maximum (taking into account rain water) – 150 m^3/hour. The pumped-out water is supplied to the surface via pipeline of diameter 100 mm and discharged in settling pond UG-0 being formed by natural hollow in relief. The project provides the pumping-out of 262,324 m^3/year of quarry water, including the discharge of water on relief – 252,485 m^3/year, sprinkling of operation roads and dump – 9,839 m^3/year.

Discharge pipeline is equipped with flow meter VMH-100 to account the discharge water. Level of underground water is monitored using hydrogeological wells G-103 located near the south-east side of the quarry and G-105 located at a distance of 1.2 km to the north-east from the quarry, in a valley of the third waterway falling further in Karabulak rivulet.

According to the project, clarification of water from suspended particles makes up 98 %. After clarification, water runs by gravity from the settling pond to relief of locality (dry bed of Karabulak rivulet). When water runs further along the bed, approximately in 500 meters to the north, at the point of intersection of Karabulak rivulet bed with road, it stops in relief valley and forms a water body UG-1 that functions actually as pond-evapourator, although formally UG-1 can not be considered as pond-evapourator, since it is located out of boundary of the land allotment.

Mine infrastructure includes: quarry, waste dump, intermediate ore storage, ore blending site, storage of off-balance ore, grading plant, charge materials storage, saleable ore storage, operation roads, camp with housing, workshop, motor transport parking, gasoline station with storage of combustive-lubricating materials, bathhouse, cellar, toilet facility, cesspool.

Total number of the mine workers is 48 persons, shift work system, 24 persons per shift [1].

2. FACTORS AND MECHANISMS FOR FORMATION OF RADIATION ENVIRONMENT

In general, the radiation environment of any territory can be presented as superposition of two components:

1. Natural radioactivity of environment.
2. Artificial radionuclide contamination of environment.

2.1. Natural radioactivity of environment

Among natural radiation sources are *cosmic radiation* and *natural radioactive substances* distributed on the Earth surface and in its depth.

2.1.1. Radioactivity of rocks

Average values of specific activity of natural radionuclides in rocks according to reference data [2] are presented in table (Table 1).

Table 1.

Specific activity of natural radionuclides in rocks

Rocks	Specific activity, Bq/kg		
	^{238}U	^{232}Th	^{40}K
Magmatic			
Persilicic	60	80	1000
Intermediate	20	30	700
Basic	10	10	240
Ultrabasic	0.4	25	150
Sedimentary			
Limestone	30	7	90
Sandstone	19	10	370
Clay slate	44	45	700

High content of natural radionuclides is observed in igneous rocks of persilicic and alkaline composition rich in potassium. High radioactivity among sedimentary rocks is observed in clay and clay slate. Pure chemical and organic sediments – rock salt, gypsum, limestone, dolomite, quartz sand, chert, jasper – have low radioactivity.

With regard to the considered territory, values of specific activity of natural radionuclides (NRN) for persilicic magmatic rocks can serve as reference marks. Degelen intrusive complex was formed by such rocks (granites).

2.1.2. Soil radioactivity

Soil-forming rocks shall be considered as main source of radioactive elements in soils. Soils formed on persilicic magmatic rocks are relatively enriched by radioactive elements (uranium, radium, thorium, potassium), and they practically do not present in soils formed on basic and ultrabasic rocks. Almost everywhere clay soils are richer in radioisotopes than sandy ones.

Specific activity of main types of soils [2] is presented in table (Table 2).

Table 2.

Specific activity of natural radionuclides in soils

Main types of soils	Specific activity, Bq/kg		
	^{238}U	^{232}Th	^{40}K
Grey soil	31	48	670
Grey-brown	28	41	700
Chestnut	27	37	550
Chernozem	22	36	410
Grey wood	18	27	370
Sod-podsolic	15	22	300
Podsolic	9	12	150
Peaty	6	6	90

Principles of distribution of NRN in sedimentary rocks are also valid in many respects for soils that are close to rocks by material constitution. However there are components in soils relatively enriched with radioisotopes – this is a thin colloidal fraction involved in exchange-sorption processes, as well as organic component. Besides, abrupt displacement of radioactive equilibrium between uranium and radium towards the last one is observed in most soils due to leaching of uranium by underground water [2].

With regard to the deposit territory, values of specific activity of NRN for Chestnut soils typical for major part of the STS territory can serve as reference mark. To the south of the considered territory, in valleys of rivulets filling the intermountain troughs, there are meadow soils; however Karadzhal deposit site is actually located beyond the mountain terrain.

2.2. Artificial radionuclide contamination of environment

There are rather many and various sources of the artificial radionuclide contamination of the Earth: emergency emissions of nuclear power plants, nuclear weapon tests and nuclear explosions for peaceful purposes, research nuclear reactors, contamination of seas by nuclear-powered watercraft, radioactive wastes, etc.

With regard to the considered territory and taking into its geographic location, the artificial radionuclide contamination will be determined, first of all, by nuclear weapon tests carried out at the STS and resulted in formation of the following main radionuclides:

- fission products – ^{137}Cs, ^{137}Ba, ^{147}Pm, ^{151}Sm, ^{90}Sr, ^{90}Y, ^{99}Tc;
- products of activation by neutrons of environment – ^{60}Co, ^{152}Eu, ^{154}Eu, ^{3}H, ^{14}C, ^{36}Cl;
- unreacted part of the charge material – ^{238}Pu, $^{239+240}$Pu, ^{241}Pu, ^{241}Am.

Traditionally, most biologically important radionuclides are considered the following ones: ^{137}Cs, ^{90}Sr, ^{3}H, $^{239+240}$Pu.

Main potential sources of contamination of the considered territory are atmospheric fallouts causing the contamination of soils on day surface and UNEs at "Degelen" site resulted in formation of cavities from which radionuclides can be carried over by underground and surface waters.

2.2.1. Atmospheric fallouts and contamination of the day topsoils

2.2.1.1. Local atmospheric fallouts

In a period from 1949 to 1962, 30 ground tests were carried out at the site "Opytnoye pole". Surface nuclear tests resulted in high radioactive contamination of vast territories and formed radioactive tracks stretched to hundred kilometers from the explosion epicenter. The figure (Figure 6) demonstrates tracks identified on the ground.

As the figure shows, the axis of movement of the cloud from explosion carried out on 02.08.55 lies near "Degelen" test site, it may well be so that edge parts of the cloud could pass over the territory of Karadzhal deposit and cause some influence on content of the artificial radionuclides in soils of the territory. This influence, however, can not be substantial, since the explosion of 02.08.55 does not relate to radiation-important event [3].

Thus, factor of local atmospheric fallouts due to surface tests at the STS shall not play essential role in formation of radiation environment at the deposit territory.

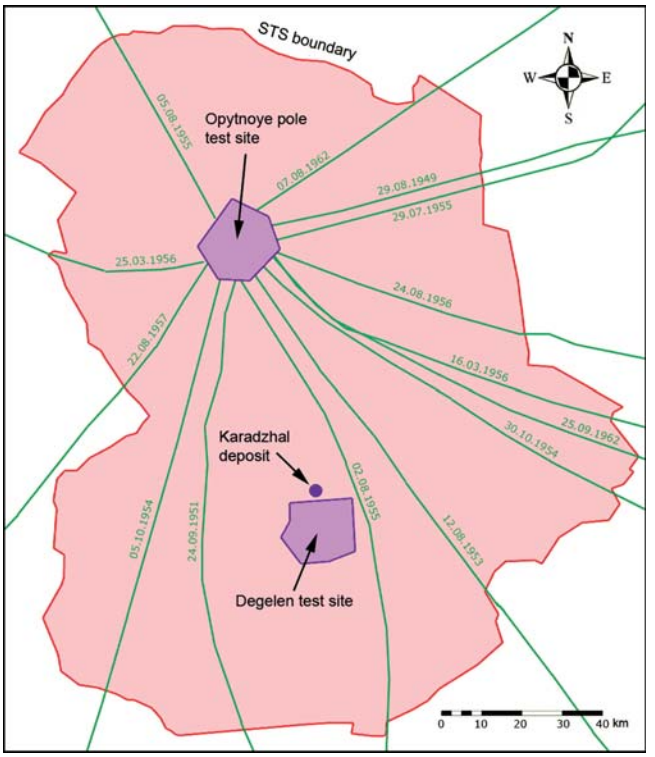

Figure 6. Attitude position of movement axes of radioactive clouds of the ground nuclear explosions carried out at the site "Opytnoye pole"

2.2.1.2. Global atmospheric fallouts

During the conduction of air nuclear tests, major part of radioactive products is erupted in stratosphere, where the mixing of vertical air layers is weak, and the precipitation is slow. Radioactive aerosols of microscopic sizes (~$4*10^{-5}$ cm) remain in stratosphere as a part of radioactive cloud from several months to several years, and winds carry this cloud over the Earth (stratospheric transfer).

Artificial radionuclides fall on surface layer of soil from atmosphere with precipitations and dry fallouts. Density of these so called global fallouts depends from geographic latitude of locality, from time passed after emission of artificial radionuclides (ARN) in atmosphere, from season and from meteorological factors.

According to reference data [4], specific activity of the artificial radionuclides in soils caused by global fallouts makes up: ^{137}Cs – 4-29 Bq/kg, ^{90}Sr – 1-19 Bq/kg, $^{239+240}$Pu – 0.02-5 Bq/kg, ^{151}Sm ~ 1.2 Bq/kg, ^{99}Tc ~ 0.2 Bq/kg. Total (cumulative) ARN sediments are redistributed with the lapse of time in natural landscapes. This takes place due to:

- horizontal migration – washout by atmosphere precipitations from highlands to lowlands, wind-induced transfer;
- vertical migration – with the lapse of time ARNs penetrate deep into soil, and some part contained in vegetable cover moves to the upper layer of soil (after plant fading).

Natural migration brings to still more spread of concentrations of artificial radionuclides in the upper (subject of study as a rule) layer of soil. Therefore, the range can be extended with confidence at the least 2-3 times as much from average one.

Estimates of potential contamination of soils with the artificial radionuclides can serve as background to observe the effect of other factors and to clarify their reasons.

2.2.2. Underground nuclear explosions and migration of the artificial radionuclides with underground and surface waters

UNEs were carried out at Degelen mountain massif in horizontal mine openings – tunnels (or adits, as also referred to in other literature). For a period from 1961 to 1989 there had been mined 181 tunnels, where 209 nuclear tests were carried out.

UNE cavities contain a huge amount of radioactive substances. If a cavity is dry, it actually represents a storage of radioactive wastes. In case of penetration of underground water in a cavity, it became an agent carrying radionuclides out of cavities and surrounding shatter zones.

All tunnels of "Degelen" site are located near the level of underground waters or in filtration zone. Water-bearing tunnels are located at a height of 600–700 m above sea level. Results of surveys demonstrate that before closure of tunnels at the end of 90-th, radioactive products had been carried out to day surface by rivulets from ~30 tunnels. Within beds and overflow lands of these rivulets there were found areas of radioactive contamination, where values of radiation parameters were much higher than parameters in tunnels from which they run out. Some tunnels (from 8 to 12, depending from season and amount of precipitations) still remain water-bearing in our days, after closure of tunnels, since water penetrates through sealing structures. Laboratory analyses of water samples show that radionuclide carry-over continues. Concentration of ^{3}H in some waterways reaches 460 kBq/kg, ^{137}Cs – 780 Bq/kg, ^{90}Sr – 1,800 Bq/kg, $^{239+240}Pu$ – 2.3 Bq/kg [5].

As a result of sorption of radionuclides from water by soils of banks and overflow lands of rivulets, they accumulate in certain places. At the end of path, all waterways flood rather vast territories forming water-logged grounds, where considerable amount of radionuclides had been concentrated for many years.

Tunnels meeting the conditions below represent an immediate danger to the territory of Karadzhal deposit:

1. Location on the northern flanks of Degelen mountains near water collection of Karabulak rivulet, the quarry territory is located in the valley of this rivulet.
2. Presence of waterways from tunnels at present (radionuclide carry-over by surface waterways) or in the past, before closure of tunnels (there is a risk that cavity of the sealed tunnel was flooded and radionuclides can be carried over by underground water).

There are 6 tunnels meeting the given conditions: 150, 151, 152, 011, 143, 511. Their attitude position relative to the quarry is presented in figure (Figure 3).

When radionuclides are carried over by underground water from cavities and shatter zone, there are various mechanisms of their further propagation and contamination of the territory:

- contamination of air due to dust formation on the contaminated soils;
- mechanical transfer of the contaminated soils;

- penetration of the radionuclide contaminated underground and surface waters in the deposit quarry;
- contamination of air by tritium due to evapouration and transpiration from surface and underground waters.

2.2.2.1. Contamination of air due to dust formation on the contaminated soils

Long-term surveys at STS [6] have showed that even on large-scale sources of radionuclide contamination of day surface, such as "Opytnoye pole", the considerable radioactive contamination of surface air is possible only in case of anthropogenic dusting (movement of motor vehicles and other works causing the considerable dust formation), as well as steppe fire.

The deposit opencast mining operations refer exactly to works causing the considerable dust formation, hence it may result in internal irradiation due to penetration of radionuclides in organism via respiratory organs and it will be required to monitor the content of the artificial radionuclides in air.

2.2.2.2. Mechanical transfer of the contaminated soils

The above mentioned radionuclide contamination of soils of the near-tunnel areas and in the valleys of waterways originated from tunnels of "Degelen" site, in the immediate vicinity of the deposit, makes possible the further transfer of contamination by people or machinery over operation roads, the developed territories, camp, etc. Thus, content of radionuclides in soil of the near-tunnel area of the tunnel 177 makes up for ^{137}Cs, $^{239+240}Pu$ and ^{90}Sr – $3.3*10^4$; $2.6*10^4$ and $1.4*10^5$ Bq/kg, respectively. This mechanism of transfer can hardly make a considerable contribution in formation of radionuclide contamination of the territory, it will rather regard to particular individuals exposed to such contamination. Therefore it will be required to impose restrictions on motor transport routes.

2.2.2.3. Penetration of the radionuclide contaminated underground and surface waters in the deposit quarry

Main direction of radionuclide carry-over beyond mountain massif is migration of radionuclides by water arteries of Degelen – rivulets Uzynbulak, Toktakushik, Baitles and Karabulak that are located in the area of influence of waterways from tunnels. Karabulak rivulet crosses Karadzhal deposit located in the northern part of "Degelen" site at a distance of about 4 km from UNE test site (Figure 3).

As it was mentioned, Karabulak rivulet is formed by junction of 3 waterways. Portal of tunnel 152 is located in the valley of the first waterway (from the west to the east), portals of tunnels 150 and 151 are located in the valley of the second waterway, and portals of tunnels 011, 143, 511 are located in the valley of the third waterway.

The first and second waterways are the least contaminated, since their tunnels are not water-bearing now. Specific activity of ^{3}H in water below the point of junction of the first and second waterways makes up about 30 kBq/kg, ^{137}Cs – up to 0.22 Bq/kg, ^{90}Sr – up to 0.07 Bq/kg. Derivation of waterway from the quarry using bypass drainage trench will exclude the risk of inflow of the contaminated surface water in the quarry. However, at the same time the risk of inflow of the contaminated underground water is not excluded.

The third waterway of Karabulak rivulet is the most contaminated in comparison with other two waterways, since now there is a water-bearing tunnel 511 in its valley. Maximum val-

ues of specific activity of ^{137}Cs and ^{90}Sr reach 700 and 400 Bq/kg. At present, the valley of the third waterway is located to the east from the quarry territory, but the quarry will expand to the east and in the near future it will come under influence of the third waterway. At the same time the risk of inflow of the contaminated both surface and underground water in the quarry rises.

2.2.2.4. Contamination of air by tritium due to evapouration and transpiration from surface and underground waters

In contrast to other artificial radionuclides, tritium is not absorbed by host rocks and migrates by water practically with the water flow velocity. Tritium contamination of surface or shallow underground waters may result in its escape into atmosphere by evapouration or plant transpiration. Concentration of tritium in underground waters within Degelen mountain massif reaches 10^5 Bq/kg. Content of tritium in air of Degelen mountain massif reaches 240 Bq/m^3, its average content is 41 Bq/m^3 [5].

With regard to Karadzhal deposit, study of this mechanism is important, since underground waters will inflow in the quarry during its development, and under some conditions, for example, windless weather, the concentration of tritium in air may reach the value representing a danger to people.

2.2.2.5. Interconnection of contamination sources, mechanisms and objects

Summarizing all above stated, main factors that may have an effect on radiation situation – potential sources of environmental contamination, intermediate transfer agents, mechanisms of their action, as well as their interaction with objects of environment can be presented in the form scheme shown in figure (Figure 7).

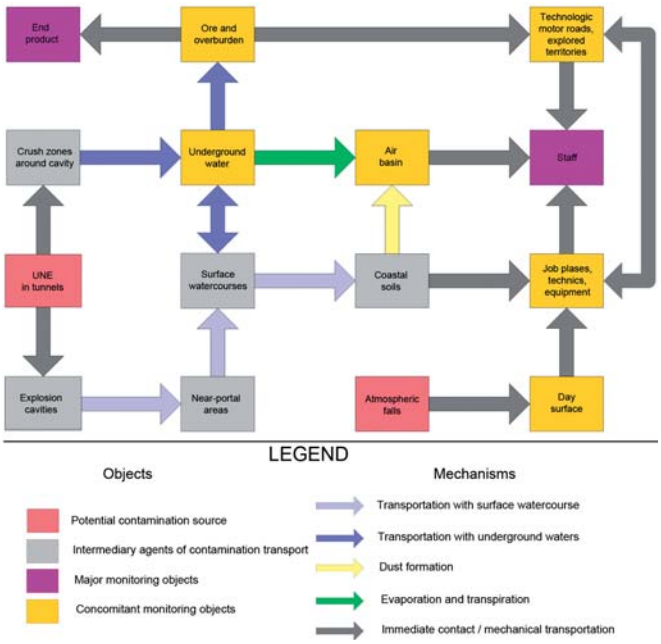

Figure 7. Main factors that may have an effect on radiation situation and their interaction with the monitored objects

3. RADIOECOLOGICAL SURVEYS IN THE TERRITORY OF THE DEPOSIT

Proceeding from the above stated, the following set of objects to be studied and topical for the considered territory was determined:

- soil of day surface areas;
- soil of the developed territories, dumps, motor roads;
- underground and surface waters;
- ore and host rocks;
- air aerosols;
- water vapours;
- workplaces and equipment;
- personnel (urine, individual dosimetry);
- finished product (concentrate).

Radioecological surveys included the conduction of field and laboratory works.

The following equipment was used for conduction of works:

- aspiration unit designed for aerosol sampling,
- standard chemical equipment for preparation of samples;
- liquid-scintillation spectrometer TriCarb 2900 TR for determination of ^{3}H concentration;
- gamma-spectrometer Ortec GMX20P4 for determination of specific activity of gamma-emitting radionuclides;
- radiometer SRP-68-01, dosimeter-radiometer RKS-01-SOLO, dosimeter RKS-01G-SOLO for measurement of values of radiation parameters.

Measurement of radiation parameters included the following:

- route gamma-radiation survey of the day surface, dumps and operation motor roads in the mode "Search";
- radiometric measurement of equivalent dose rate (EDR) and density of surface contamination by α- and β-emitting radionuclides in fixed points;
- radiation monitoring of workplaces and equipment;
- EEVA measurement of radon and thoron in industrial rooms, the quarry and camp;
- random radiation survey.

Sampling consisted in taking samples of soil and ore, underground and surface waters from observation wells, sump, contiguous rivulets, air aerosols, urines, water vapours in air, finished products (concentrate).

Laboratory researches included the determination of specific activity of gamma-emitting radionuclides, radiochemical analysis for determination of specific activity of ^{90}Sr, $^{239+240}Pu$ and beta-spectrometric analysis for determination of ^{3}H concentration.

Attitude position of sampling points is presented in figure (Figure 5).

This paper presents the analysis of results of all radioecological investigations and the assessment of current radioecological state of the deposit territory.

4. RESULTS

By now a large volume of actual material has been accumulated as a result of works carried out at the deposit. Volume of material (quantity of measurements and laboratory analyses) is presented in the table (Table 3).

Table 3.

Volume of actual material on all types of field and laboratory research works

Type of measurement or sample	1998	2003	2004	2005	2006	2007	2008	2009
	Results of radiation monitoring, quantity of measurements							
Radiation monitoring in fixed points of day surface	√*	-	√	-	-	3	5	3
Radiation monitoring of the developed territories dumps, motor roads	-	-	-	-	9	5	5	3
Radiation monitoring of workplaces and equipment	-	-	-	24	98	120	116	131
Random individual radiation survey	-	-	-	-	-	19	33	16
EEVA measurement of radon and thoron in industrial rooms, the quarry and camp	-	-	-	-	-	18	-	-
	Results of laboratory analyses, quantity of samples							
Soil of day surface areas	√	-	√	5	-	3	5	3
Soil of the developed territories, dumps, motor roads	-	-	-	-	9	5	5	3
Underground and surface waters	√	√	√	18	4	12	20	20
Ore and host rocks	-	-	-	-	4	65	5	8
Air aerosols	-	-	-	3	4	4	4	5
Water vapours	-	-	-	4	14	45	60	98
Urines	-	-	-	-	-	88	40	36
Finished product (concentrate)	-	-	-	-	-	20	39	26
Note: * - sign √ means that there are already generalized data, exact quantity of measurements or samples is unknown								

4.1. Results of radiation monitoring of target objects

Radiation monitoring of the day surface was carried out in the mode "Search". The places with maximum EDR values were selected as points of soil sampling for conduction of comprehensive analysis of the content of natural and artificial radionuclides. Results of radiation monitoring of the day surface are presented in the table (Table 4).

Table 4.

Results of radiation monitoring and laboratory analyses of soil samples taken in fixed points of the day surface

Type of measurement	2005	2006	2007	2008	2009
	Averaged value (minimum – maximum)				
Surface α-radiation density, particles/(min*cm^2)	-	-	< 0.2	< 0.2	< 0.2
Surface β-radiation density, particles/(min*cm^2)	-	-	18 (17-19)	14 (10-16)	< 10
EDR on ground surface, μSv/h	-	-	0.14 (0.14-0.15)	0.15 (0.12-0.19)	0.13 (0.12-0.13)

Obtained results indicate that radiation parameters of the day surface do not exceed the background values typical for the given locality. Thus, relative stability of radiation parameters values was fixed for the whole period of observations and there are no reasons to expect any abnormally high values.

A state of operation roads and the developed territories was studied according to the same scheme. Results of radiation monitoring of operation roads and the developed territories are presented in the table (Table 5).

Table 5.

Results of radiation monitoring and laboratory analyses of soil samples taken in the developed territories, dumps, motor roads

Type of measurement	2005	2006	2007	2008	2009
	Averaged value (minimum – maximum)				
Surface α-radiation density, particles/(min*cm^2)	-	< 0.3	< 0.2	< 0.2	< 0.2
Surface β-radiation density, particles/(min*cm^2)	-	5 (4-7)	11 (10-12)	8 (7-9)	< 10
EDR on ground surface, μSv/h	-	0.11 (0.09-0.14)	0.14 (0.11-0.20)	0.11 (0.10-0.12)	0.13 (0.12-0.14)

Obtained results indicate that radiation parameters of the developed territories, dumps, motor roads do not exceed the corresponding values for the day surface. There was no radionuclide contamination of the site in the course of development of the deposit.

Measurement of radiation parameters in workplaces, in rooms of temporary and permanent attendance, on working cloths and skin of personnel was also carried out. Results of radiation measurements are presented in the table (Table 6).

Table 6.

Results of radiation monitoring of workplaces and equipment

Type of measurement	2005	2006	2007	2008	2009
	Averaged value (minimum – maximum)				
Surface α-radiation density, particles/(min*cm^2)	< 0.3	< 0.3	< 0.2	< 0.2	< 0.2
Surface β-radiation density, particles/(min*cm^2)	9.3 (3-15)	8.6 (4-15)	8.2 (5-16)	7.7 (4-15)	9.1 (5-14)
EDR on ground surface, μSv/h	0.13 (0.07-0.19)	0.11 (0.05-0.18)	0.11 (0.05-0.19)	0.09 (0.04-0.18)	0.10 (0.05-0.18)

EDR values in the deposit territory were in the range from 0.09 to 0.13 μSv/h. The measured values of β-particles flux density were <10 particles/(min*cm^2). Values of α-particles flux density in all points of survey did not exceed the detection limit of measurement instrumentation applied (<0.2-0.3 particles/(min*cm^2)). This fact demonstrates that personnel and motor transport had complied with limits set for movement routes and had not been exposed to casual radionuclide contamination in potential hazardous places.

Minor variations of values of radiation parameters are connected with the fact that surveys have been carried out at different times and by different groups, and also the applied dosimeters and radiometers differed from each other both by ionizing radiation detection principles and other technical characteristics effecting on results of measurements (detector type and area, detector anisotropy, energy nonlinearity of detector efficiency, etc.). When measuring the values close to the lower threshold of detection limit, all these parameters may bring to high uncertainty of results – up to ± 30%.

According to results of surveys it was defined that radiation parameters of all target objects of the deposit territory do not exceed the background values of the given territory. For reasons given, further radiation monitoring of the target territory is not necessary.

4.2. State of soils

In order to study the dynamics of development of radioecological situation, the day surface of the deposit territory is subjected to annual survey of mining lease areas being involved in development according to mining plan. Soil samples for laboratory analyses are taken in points of maximum values of radiation parameters detected during the radiometric survey in the mode "Search". Sampling is performed 0–5 cm deep using "spike" technique.

Results of laboratory analyses of soil samples taken from the day surface areas are presented in the table (Table 7). Values of minimum significant specific activity (MSSA) according to the Radiation Safety Norms NRB-99 are presented for comparison reasons.

Table 7.

Results of laboratory researches of the day topsoil samples

Radionuclide	2004	2005	2007	2008	2009
	Averaged value of specific activity (minimum – maximum), Bq/kg				
^{40}K	(120-430)	750 (480-1020)	-	570 (500-700)	620 (450-900)
^{226}Ra	(25-56)	31 (27-35)	-	54 (35-90)	55 (50-60)
^{232}Th	(6-59)	44 (18-79)	-	17 (7-20)	32 (20-45)
^{241}Am	-	0.6 (<0.3-0.9)	<1	<1	<1
^{137}Cs	(<0.9-33)	16 (<0.4-51)	<1	2.4 (<1-6)	14 (<1-25)
^{90}Sr	(2-3.5)	12 (<9-15)	11 (11-11)	5 (5-5)	13 (12-14)
^{239,240}Pu	(<1-17)	24 (11-36)	-	10 (10-10)	7 (0.5-13)

The obtained results show that contents of natural radionuclides in the day topsoils do not exceed the average natural background (Table 2), and contents of the artificial radionuclides do not exceed the background of global fallouts, except $^{239+240}$Pu, its content is slightly higher than the background one. Most likely this is connected with the passage of radioactive tracks from ground nuclear weapon tests at the site "Opytoe pole". On the whole it can be stated that the day topsoil of the target territory does not represent radiation hazard to personnel of the deposit.

Results of laboratory analyses of soil samples of the developed territories, dumps and motor roads are presented in the table (Table 8).

Table 8.

Results of laboratory analyses of soil samples of the developed territories, dumps and motor roads

Radionuclide	2006	2007	2008	2009
	Averaged value of specific activity (minimum – maximum), Bq/kg			
^{40}K	-	-	690 (300-1300)	510 (400-650)
^{226}Ra	-	52 (45-59)	43 (20-60)	70 (65-80)
^{232}Th	-	55 (33-67)	32 (10-65)	32 (20-40)
^{241}Am	0.9 (<0.5-2)	< 1.5	< 1	< 1

Radionuclide	2006	2007	2008	2009
	Averaged value of specific activity (minimum – maximum), Bq/kg			
^{137}Cs	25 (2-46)	10 (<1-36)	7 (<1-15)	4 (<1-10)
^{90}Sr	16 (10-22)	14 (8-20)	-	-
$^{239,240}Pu$	12 (6-18)	-	-	-
^{60}Co	< 1.7	< 1	< 1	-

According to results of works it was established that contents both natural and the artificial radionuclides in soils of the developed territories, dumps and motor roads generally do not exceed the corresponding values for the day surface, i.e. there was no radionuclide contamination of the area in the course of development of the deposit. However, according to results of works in 2006, there were points with increased values of $^{239,240}Pu$. This also confirms indirectly the expressed assumption about the passage of radioactive track over the deposit territory.

When a deposit is located in the immediate vicinity of the nuclear weapon test site, a temporary variation of radiation parameters of rocks and ore is possible due to migration of radionuclides with underground waters and due to anomalies not revealed during the survey. In order to monitor these processes, rock and ore sampling is performed for further conduction of gamma-spectrometric analysis to determine the specific activity of natural and the artificial radionuclides.

Results of laboratory analyses of ore and host rock samples are presented in the table (Table 9).

Table 9.

Results of laboratory analyses of ore and host rock samples

Radionuclide	2006	2007	2008	2009
	Averaged value of specific activity (minimum – maximum), Bq/kg			
^{40}K	390 (200-810)	-	75 (29-140)	390 (80-780)
^{226}Ra	23 (16-38)	76 (22-390)	19 (7-32)	95 (50-220)
^{232}Th	21 (14-34)	21 (5-130)	4 (2-7)	36 (25-59)
^{210}Pb	120 (86-170)	-	-	-
^{241}Am	< 0.4	< 2	< 1	< 1
^{137}Cs	< 0.5	20 (<0.7-39)	< 1	1.9 (<1-5)
^{90}Sr	-	7 (<2-11)	-	-
^{60}Co	< 1	< 3	< 2	-

Contents of natural radionuclides in ore and host rock do not exceed the corresponding values for the given rock. Presence of the artificial radionuclides ^{90}Sr, ^{137}Cs draws the attention. Most likely this is connected with the mining technology, while excavating the overburden, areas of the day surface, where presence of the given radionuclides was detected, could mix with ore.

Also random radiation monitoring of the deposit product to detect possible radioactive contamination is carried out in the framework of these surveys. Results of laboratory analyses of the finished product (concentrate) samples are presented in the table (Table 10).

Table 10.

Results of laboratory analyses of the finished product samples

Radionuclide	2007	2008	2009
	Averaged value of specific activity (minimum – maximum), Bq/kg		
^{40}K	-	250 (23-480)	270 (200-350)
^{226}Ra	34 (22-71)	14 (4-54)	22 (10-30)
^{232}Th	10 (<2-15)	8 (2-18)	10 (4-15)
^{241}Am	< 2	< 1	< 1
^{137}Cs	< 2	< 1	< 1
^{238}U	-	< 8	-
^{60}Co	< 2	< 3	-

According to results of laboratory analyses it was established that the deposit product is not exposed to radioactive contamination and can be used without limits.

Thus, concentration of natural radionuclides in soils of the target territory corresponds to natural background of this area (predominately rocks of granite variety).

Content of the artificial radionuclides is comparable with background of global fallouts and is connected with the conduction of nuclear tests, except minor excess by $^{239+240}Pu$ that was detected in some points of the day surface. This can be explained by influence of edge tracks of nuclear tests at the site "Opytnoye pole".

4.3. State of air medium

Radioactive contamination of the territory by products of nuclear explosions represents danger only to internal irradiation. During the deposit development, main path of radionuclide ingress is inhalation that is conditioned by dust formation at conduction of exploration, mining and construction works accompanied by breakdown of soil layer. Due to opencast and through aeration of the working area, the concentration of radionuclides in air of the working area under certain circumstances may make considerable contribution in formation of personnel irradiation dose due to inhalation. These circumstances can be caused by natural factors (dust storms, etc.), as a result radioactive products may get in air from nuclear explosion areas.

Results of laboratory analyses of air samples are presented in the table (Table 11).

Table 11.

Results of laboratory analyses of air samples

Radionuclide	Acceptable volumetric activity ($AVA_{saturation,}$ (NRB-99), Bq/m^3	2005	2006	2007	2008	2009
		Volumetric activity, Bq/m^3				
^{40}K	31	$< 1.5*10^{-5}$	$< 4*10^{-5}$	-	-	-
^{226}Ra	0.03	$< 1*10^{-5}$	$< 1*10^{-5}$	-	-	-
^{232}Th	$4.9*10^{-3}$	$< 2*10^{-6}$	$< 1*10^{-5}$	-	-	-
^{210}Pb	0.11	$< 3*10^{-4}$	$< 4*10^{-3}$	-	-	-
^{241}Am	$2.9*10^{-3}$	$< 2*10^{-7}$	$< 4*10^{-7}$	$< 7*10^{-7}$	$< 2*10^{-4}$	$< 1*10^{-6}$
^{137}Cs	27	$< 1*10^{-6}$	$< 3*10^{-6}$	$< 1*10^{-6}$	$< 1*10^{-5}$	$< 1*10^{-6}$
^{90}Sr	2.7	-	-	$< 5*10^{-6}$	-	-

According to results of analyses it was noticed that there was no presence of the artificial radionuclides in air of the object for the whole period.

Water vapour sampling at the deposit territory was carried out right near sump, where maximum content of tritium in air was expected. Part of samples was taken in living and working areas, in places of long-continued stay of personnel. Results of analyses of tritium concentration in air are presented in the table (Table 12).

Table 12.

Results of laboratory analyses of air samples

Radionuclide	$AVA_{saturation}$ (NRB-99), Bq/m^3	2005	2006	2007	2008	2009
		Averaged value of volumetric activity (minimum – maximum), Bq/m^3				
^{3}H	1900	9.3 (2-25)	3.8 (0.5-13)	24 (33-234)	6.3 (<0.1-42)	9.6 (<0.1-165)

The increased concentration of tritium in 2007 in comparison with other years can be explained by sampling in windless weather. Nevertheless, these values, as well as other ones in other years, are considerably lower the permissible annual average activity of tritium making up 1900 Bq/m^3.

Results of laboratory analyses on determination of volumetric activity of natural and the artificial radionuclides in air samples have demonstrated that they are lower in several degrees than annual average volumetric activity in air inhaled by population and they do not represent the radiation hazard.

Values of tritium concentration in air also did not exceed the permissible annual average volumetric activity of tritium for population. However, taking into account considerable content of tritium in underground and drainage waters of the quarry (up to 50 kBq/kg), it is not ruled out that under certain weather conditions, for example, in windless weather, considerable amount of tritium may ingress in air of the deposit working area.

4.4. Assessment of underground and surface water contamination by the artificial radionuclides

Main points of monitoring of underground and surface waters at the deposit territory are hydrogeological wells G-103 and G-105, the quarry sump and pond-evapourator

UG-1. Water samples were taken according to standard technique in the sealed polyethylene vessels. Before filling, single-trip plastic laboratory vessels for samples were rinsed out by stratal water not less than three times. Volume of samples for determination of the content of artificial radionuclides ^{137}Cs, ^{90}Sr and $^{239+240}$Pu was not less than 10 liters, for determination of the content of ^{3}H – 1.5 liters. Results of laboratory analyses of surface water samples for determination of the content of ^{137}Cs, ^{90}Sr are presented in the figure (Figure 8).

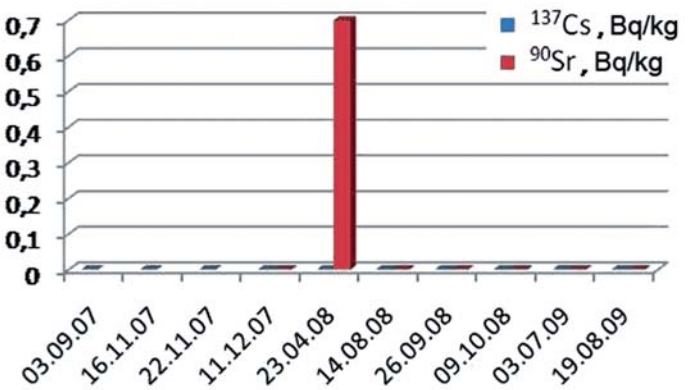

Figure 8. Content of ^{137}Cs and ^{90}Sr in quarry water (sump)

Results of laboratory analyses of underground water samples for determination of the content of ^{137}Cs, ^{90}Sr are presented in the figure (Figure 9).

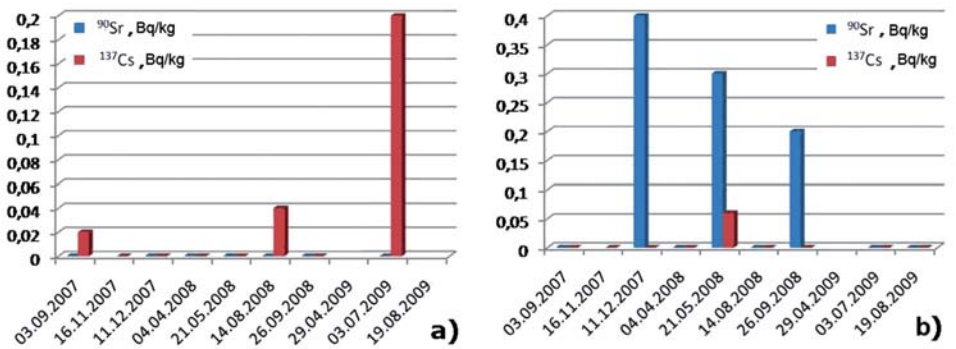

Figure 9. Content of ^{137}Cs and ^{90}Sr in underground waters a) well G-103; b) well G-105

Analysis of laboratory results has demonstrated that some water samples are characterized by presence of the artificial radionuclides ^{137}Cs, ^{90}Sr in amounts considerably lower than intervention levels (NRB-99, ^{137}Cs - 11 Bq/kg, ^{90}Sr – 5 Bq/kg). However, their presence in the deposit area requires the conduction of additional precise surveys. Some random water samples contained the artificial radionuclide $^{239+240}$Pu. As a result it was established that concentration of $^{239+240}$Pu in water is lower than detection level making up 2 mBq/kg.

Main contaminant of underground and surface waters is tritium. Concentration of tritium in most water samples exceeds several times the intervention level making up 7700 Bq/kg (NRB-99).

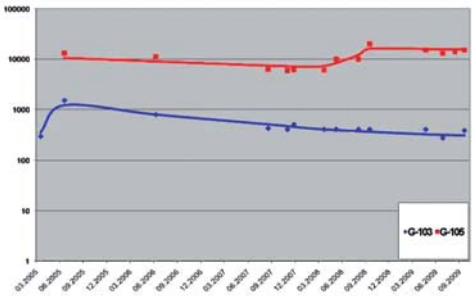

Figure 10. Dynamics of tritium concentration in underground water.

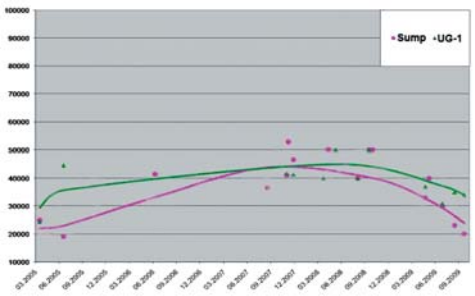

Figure 11. Dynamics of tritium concentration in surface water.

Tritium concentration in underground water for the observed period has remained practically invariable. At the same time, specific activity of tritium in surface water of the quarry depends from the level of dilution of the tritium contaminated underground water by melt water and atmospheric fallouts and from amount of atmospheric fallouts. In a period of low amount of precipitations, the annual average concentration of tritium is increased and vice-versa.

5. DOSIMETRIC CONTROL OF STAFF

Individual radiation monitoring is carried out using dosimeters DTL-02 that fix the exposure dose from external gamma radiation. Individual dosimeters were given to the deposit personnel spending maximum time according to its duty in working area. It is necessary to note that employees of "Ulba-FtorComplex" Ltd Company are not personnel of "A category" and individual radiation monitoring is not obligatory for them. However, presence of the contaminated areas in territory of "Degelen" site, relatively free access to them, probable presence of radioactive contamination seats not revealed during the survey requires the need in conduction of such type of monitoring.

The obtained results allowed us to state that effective dose of external gamma radiation, total from natural background and possible effect of the artificial radionuclide contamination, did not exceed 1 mSv per year. According to NRB-99, effective dose from natural background is not regulated, and from the induced sources it shall not exceed 1 mSv a year. It is obvious that in this case there was no excess of permissible values regulated by NRB-99.

Calculation of dose due to internal irradiation by tritium was carried out according to [7]. For this purpose the specific activity of tritium in urine of personnel working in the deposit was determined. (Figure 13).

Slightly increased values of tritium in 2007-2008 are connected with application of different techniques of determination of tritium in urine, without preliminary sample preparation and with distillation. When the technique without preliminary sample preparation was applied, natural radionuclides contained in urine and luminophors could make a contribution

in count rate. Thereby overrated results of analyses were obtained. After the conduction of preliminary preparation and distillation of samples, contribution of interfering radionuclides was deducted and it allowed us to obtain more precise results of analyses.

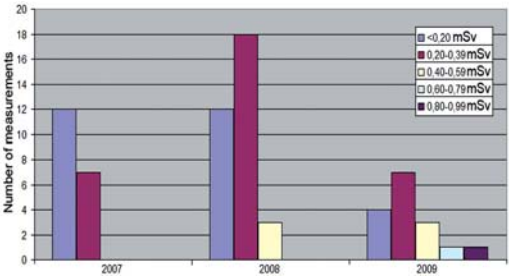

Figure 12. Results of individual radiation monitoring

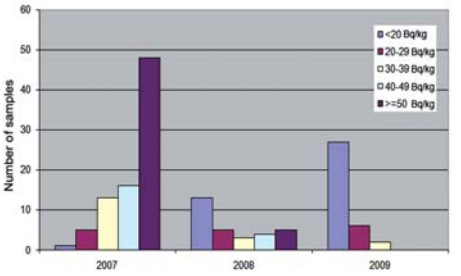

Figure 13. Results of determination of tritium content in urine samples for 2007-2009

Results of calculation of internal irradiation doses of personnel working in Karadzhal deposit are presented in the table (Table 13).

Table 13.

Results of laboratory analyses of urine samples and calculation of internal irradiation doses due to tritium penetrated in organism

Analyzed component	2007	2008	2009
	Averaged value (minimum – maximum)		
^{3}H in urine, Bq/kg	65 (17-1160)	53 (10-300)	16 (10-60)
Irradiation dose, mSv/year	$9.7*10^{-5}$ ($2.6*10^{-5}$-$1.7*10^{-3}$)	$7.9*10^{-5}$ ($1.5*10^{-5}$-$4.5*10^{-4}$)	$2.3*10^{-5}$ ($1.5*10^{-5}$-$9.0*10^{-5}$)

The obtained data demonstrate that using any methods of measurement there was no overexposure of personnel due to inhalation penetration of tritium. Tritium inhaled with air makes in total annual effective dose of external and internal irradiation less than 0.1% of contribution from the permissible dose limit for population making up 1 mSv.

6. DISCUSSION

As a result of researches it was established that the content of the artificial radionuclides in the day surface of the deposit territory is conditioned mainly by global fallouts and probably by contingency radiation tests during the conduction of nuclear tests at "Degelen" and "Sary-Uzen" sites.

Today local areas of radiation contamination located at "Degelen" and "Sary-Uzen" sites can be potential sources of secondary radiation contamination.

Content of natural radionuclides in soil cover is typical for soils of the given territory. There was detected the presence of the artificial radionuclides, however their concentration is quite comparable with background of global fallouts.

The artificial radionuclides were not detected in air of the object. Content of tritium in air of the object does not exceed the permissible values established by radiation safety norms (NRB-99) and is typical for "Degelen" site. However, it is not ruled out that the contribution of tritium in radiation dose under certain conditions may be substantial.

Values of radiation parameters in all measurement points remain practically invariable. Radioactive contamination of working surfaces of equipment, transport vehicles and personnel rooms was not detected.

Approach of underground and surface waters contaminated by the artificial radionuclides to boundaries of the deposit remains the main problem issue. Tritium – the artificial radionuclide is a major radioactive contaminant of underground and surface waters of Karadzhal deposit. Unique feature of this contaminant is its ubiquitous presence in underground waters.

Since UNEs in "Degelen" tunnels were stopped in 1989, only "long-lived" radionuclides are of import now. Among them traditionally four radionuclides ^{3}H, ^{90}Sr, ^{137}Cs and $^{239+240}Pu$ are biologically most important (hazardous) and their amount is the largest one. At present attention of researchers is also paid to additional artificial radionuclides – ^{241}Am, ^{238}Pu, ^{241}Pu, ^{36}Cl, ^{99}Tc etc., however now their consideration is beyond the limits of subject of the current paper.

Radionuclide ^{3}H is not absorbed by host rocks and, hereupon, it passed the path from areas of tests to the deposit territory before start of observations. Although dynamics of its concentrations in various sampling points has not yet stabilized, overall increase of ^{3}H content in surface and underground waters is not observed. Probably, concentration of tritium will decrease after several years due to decay processes.

$^{239+240}Pu$ represents a special hazard in case of air and ground nuclear explosions. In case of underground tests that took place at Degelen mountain massif, practically all plutonium is confined in rock melt forming glassy mass at the bottom of cavity. About 70% of ^{90}Sr and ^{137}Cs is also located there. Within first decades after explosion this source of radionuclides does not make substantial contribution in formation of radiation situation, since leaching process runs very slowly. Second source of radionuclides being formed at explosion is radioactive aerosol-dust component localized on broken rock in collapse column above the cavity and in fractures of the fractured zone around the cavity. After explosion this source contains all tritium formed and about 30% of ^{90}Sr and ^{137}Cs, which start actively redistributing and migrating in environment under influence of underground water and penetrating atmospheric fallouts.

The quarry of Karadzhal deposit represents absolute horizontal drain intercepting underground water flows running from tunnels 151, 152 and 511. Each of these tunnels was subjected to nuclear explosion of power 20 ktons and ~10^{15} Bq of tritium and ~10^{14} Bq of ^{90}Sr and ^{137}Cs was produced. Tritium carry-over from the shatter and fractured zones of these tunnels is a reason of its high concentration in drainage waters of the quarry. For a while there are no considerable amounts of ^{90}Sr and ^{137}Cs in these waters, since these radionuclides move much slowly than tritium due to sorption on walls of water channels and the sorption capacity of rock block between tunnels 151, 152, 511 and the quarry is not exhausted yet.

It is difficult to assess precisely the time of saturation of the mentioned rock block with cesium and strontium because of lack of information about internal structure of Degelen mountain massif, filtration and sorption properties of rocks, water migration channels. Thus,

it is not ruled out that appearance of ^{90}Sr and ^{137}Cs in the quarry is a thing of the nearest future. If this happens, a picture of radioactive contamination of the deposit would qualitatively change. Cloth and footwear of personnel, instruments and machinery will acquire indelible contaminants and become sources of external irradiation; the deposit product will not meet the radiation safety requirements and will be unsuitable for further processing.

Future of the deposit may have two options of development – optimistic and pessimistic. According to the first option, the deposit reserves will be depleted before the artificial radionuclides appear in considerable amounts in underground waters inflowing in the quarry. According to the second option, there will be need in construction of special protective objects – drainage workings or antifiltration barriers between the quarry side and Degelen mountain massif. This is an expensive measure and it may be economically reasonable only in case of rise of real threat to radiation situation.

In the course of development of the deposit, the quarry will deepen. Design depth that will be reached at the end of development is 70 m. At the same time, depression cone of considerable size will arise in pond of fracture water at the deposit territory, and this will result in increase of filtration rate of underground water in the quarry and, hence, in acceleration of ^{90}Sr and ^{137}Cs movement from underground nuclear test areas.

In order to carry out additional radioecological surveys to study processes of propagation of the artificial radionuclides by underground waters to boundaries of the deposit, it is necessary to create a network of observation wells. By development of proposals on determination of well locations, three-dimensional structure of filtration flows shall be taken into account. According to results of radioecological researches, the artificial radionuclides migrate at "Degelen" site both by surface waterways and by underground water. Underground waters of "Degelen" test site are represented by three types:

- underground waters of fractured-stratal type confined to zones of exogenous efflorescence of crystal rocks composing an intrusive massif. The form continuous cover blocked in rivulets valleys by sedimentary deposits;
- underground waters of fractured-vein type circulating through fracture zones of high permeability;
- porous-stratal waters locally spread in loose sediments filling rivulets valleys. They have a character of flows concurring with slope of creek valleys and, most probably, they directly contact with underflow and surface waters rivulets.

The following is proposed for assessment of possible passage of radionuclides with underground waters from Degelen mountain massif to the quarry being under development:

1. To drill 5 wells in line every 200-250 m across the flow of underground water at a distance of 50 m from the quarry side in order to assess the spatial propagation of radionuclides with underground waters of fractured-stratal type.
2. To drill 1 well in the fracture zone from the south-east side within ore body in order to assess the propagation of radionuclides with underground waters of fractured-vein type.
3. To drill 3 wells along rivulet bed every 50, 150 and 250 m from the quarry side towards Degelen mountain massif in order to assess the contamination of underflow waters of Karabulak rivulet.

Hydrogeological wells shall be drilled to the depth of occurrence of water-bearing horizon within the deposit (down to 45 m), wells of depth to 20 m can be used as monitoring ones.

CONCLUSIONS

Results of radioecological surveys in Karadzhal deposit area presented in this paper have allowed us to perform the assessment of current radiation situation of the deposit territory.

With regard to series radiation monitoring issues, the situation at the site is favorable now.

Values of radiation parameters determining hazard of external and internal irradiation (EDR, α- and β-particles flux density) do not exceed the permissible levels regulated by NRB-99. Content of natural and the artificial radionuclides in soils corresponds to natural level and background of global fallouts. Therefore it is proposed to exclude or to reduce substantially this type of monitoring during the conduction of works on radioecological monitoring.

Doses received by personnel from external gamma radiation do not exceed the permissible levels regulated by NRB-99. Content of the artificial radionuclides in air is lower by several degrees than the permissible annual average volumetric activity in inhaled air for population regulated by NRB-99.

Main problematic issue is propagation of underground waters contaminated by the artificial radionuclides, in particular, tritium of concentration up to 50 Bq/kg, to boundaries of the deposit. An issue regarding the value of personnel body burden due to tritium inhalation requires additional study. In this connection it is reasonable to increase the scope of monitoring works, as well as to assess doses received taking into account workplace, time of stay at the mine, etc.

For the time being the content of ^{137}Cs and ^{90}Sr in underground and surface waters occurs in the form of tracks of concentrations close to the detection limit and much less than the interference level, $^{239+240}Pu$ was not detected. At the same time, now it is not possible to make reliable prediction regarding the inflow of underground water contaminated by the artificial radionuclides in the deposit quarry. In this connection it is reasonable to increase both volume and quantity of samples, including samples from newly drilled wells. Otherwise this circumstance may result in unmonitored propagation of the artificial radionuclides, contamination of the deposit product, process equipment, machinery, etc., as well as in irradiation of personnel.

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ҚАРАДЖАЛ ФЛЮОРИТТІ КЕНОРНЫНЫҢ АУМАҒЫНДАҒЫ РАДИОЭКОЛОГИЯЛЫҚ ХАЛ-АХУАЛ

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Осы жұмыста Семей сынақ полигонындағы жерасты ядролық жарылыстары жүргізілген орынға тікелей жақын орналасқан Қаражал флюорит кен орнының аумағындағы көп жылғы радиоэкологиялық зерттеу деректері берілген. Кен орнының аумағындағы жерүстіндегі радионуклидтердің күндізгі техногенді құрамы, негізінен, көп мөлшердегі түсулерге байланысты. Топырақтағы, ауадағы, кен орнының өнімдеріндегі радиациялық параметрлер мәнінің, радиоуклидтер құрамының қауіпі жоқ. Жабдықтардың, көлік құралдарының, қызметкерлер тұратын жайлардың радиоактивті ластануы тіркелмеген. Кен орны шекарасына тритийдің аралас су деңгейінен көп есе жоғары шоғырлануымен ластанған жерасты суларының түсуі негізгі проблемалы мәселе болып табылады. Сондай-ақ жерасты суларында ^{137}Cs и ^{90}Sr радионуклидтері болуының іздері байқалған. Кен орнының ары қарайғы әзірленуі кезінде карьерге осы радионуклидтердің түсуінің ұлғаюы мүмкін. Кен орнындағы радиациялық жағдайдың шынайы болжауын алу мақсатында бақылау ұңғымалары желісін құру бойынша ұсыныстар берілді.

Түйін сөздер: кен орны, "Дегелең" алаңы, гидрогеология, радионуклидті ластану, мониторинг, жерасты ядролық жарылыстары.

РАДИОЭКОЛОГИЧЕСКОЕ СОСТОЯНИЕ ТЕРРИТОРИИ ФЛЮОРИТОВОГО МЕСТОРОЖДЕНИЯ КАРАДЖАЛ

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В настоящей работе представлены данные многолетних радиоэкологических исследований на территории месторождения флюоритов Караджал, расположенного в непосредственной близости от мест проведения подземных ядерных взрывов на Семипалатинском испытательном полигоне. Установлено, что содержание техногенных радионуклидов на дневной поверхности территории месторождения обусловлено, в основном, глобальными выпадениями. Значения радиационных параметров, содержания радионуклидов в грунтах, воздухе, продукции месторождения не представляют опасности. Радиоактивного загрязнения оборудования, транспортных средств, помещений пребывания персонала не зафиксировано. Основным проблемным вопросом является поступление к границам месторождения подземных вод, загрязненных тритием с концентрацией, превышающей уровень вмешательства в несколько раз. Также в подземных водах отмечены следы присутствия радионуклидов ^{137}Cs и ^{90}Sr. При дальнейшей разработке месторождения не исключено увеличение поступления в карьер этих радионуклидов. Даны рекомендации по созданию сети наблюдательных скважин с целью получения достоверного прогноза развития радиационной обстановки на месторождении.

Ключевые слова: месторождение, площадка "Дегелен", гидрогеология, радионуклидное загрязнение, мониторинг, подземные ядерные взрывы.

CHAPTER: ADVANCED STUDIES AT STS

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USE OF PLANTS AS INDICATORS FOR NEAR-SURFACE RADIOACTIVE CONTAMINATION AT NEAR-PORTAL AREAS OF THE TUNNELS AT THE FORMER "DEGELEN" TEST SITE

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The article presents the results of the study of vegetation and the nature of its radioactive contamination at the near-portal area of the tunnels at the former "Degelen" test site. There has been assessed possibility of using the plants as indicators of near-surface contamination at the near-portal area of the test site.

Key words: nuclear tests, tunnels, radioecology, radioactive contamination, radionuclides, vegetation cover, plants, accumulation coefficient (A_c).

INTRODUCTION

During Semipalatinsk Test Site operation, Degelen experimental site was a place used for underground nuclear explosions in horizontal tunnels. Testing included an explosion with further opening of a tunnel which leads to partial escape of radioactive contamination to original ground. Later there was a liquidation of tunnel portal performed and reclamation of work directed at decrease of radioactive contamination at tunnel entrance which resulted in partial burial of radioactive contamination under technogenic soil.

Currently in most cases tunnel entrances do not have surface radioactive contamination, although there not enough information about deep soil layers. Vegetation capable of accumulating of radionuclides from soil may serve as indicators of subsurface contamination. During recent years it has been determined that in conditions of meadow biogeocenosis widely distributed in the areas of radioactive tunnel stream at Degelen site, radionuclide accumulation from soil by vegetation changes within wide range. Accumulation coefficient values range (A_c) for ^{90}Sr, determined as ratio of radionuclide content to vegetation mass unit [1], varies from 0,1 to 1, A_c ^{137}Cs – from 0,001 to 1 (in the area of tunnel #177) and from 0,01 to 10 (in the area of tunnel #176). ^{241}Am A_c value on the average do not exceed tenth, $^{239+240}Pu$ A_c is within 0,0001 to 0,01 [2]. Although even with such differences in accumulation for the same radionuclide, knowing specific activity of radionuclides in vegetation estimates can be done on expected radionuclide content in soil. With that radioactive contamination characteristics of thicker massive subtopsoil layer may be done with perennial vegetation (trees, bushes) with well developed root system.

1. MATERIALS AND RESEARCH METHODS

Studies were performed in the area of several tunnels entrances of former test site Degelen marked as – B, P, C, N, B_1, A, F, G, O, I, E, R and Q. Study of area vegetation was performed by different methods of geobotanic description with identification of main

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types of vegetation and species [3]. To sample plants in the area of each tunnel two areas were chosen, one was located on technogenic earth-deposits on a portal and second one – at some distance from it. Plant samples represented upper part (current year shoots) of studied species of bushes and trees. Soil sampling was made at the portal area of tunnels taking into consideration local relief, location of natural and technogenic objects, access roads and also areas with higher radiation background found during gamma-survey. [4]. Soil sampling area was 100-200 cm^2, depth - 5 cm. Samples was taken and packaged in accordance with requirements to soil sampling for general and local [5].

Analysis for radionuclides specific activity measurements in soil and plant samples was performed in accordance with standard methods at certified lab equipment [6, 7]. Measurement of specific activity of ^{137}Cs and ^{241}Am radionuclides was performed at Canberra GX-2020 gamma spectrometer, ^{90}Sr – at beta spectrometer "Progress" (soil) using radiochemical isolation (of a plant). Detection limit for ^{137}Cs was 1 bq/kg (for plant samples) and 4 bq/kg (for soil sample), ^{241}Am – 0,3 Bq/kg and 1 Bq/kg, ^{90}Sr – 1 Bq/kg and 100 Bq/kg. Measurements error for ^{137}Cs and ^{241}Am did not exceed 10–20 %, ^{90}Sr – 15–25 %.

Spatial interpolation method of "Kriging" was used to map ^{137}Cs and ^{241}Am distribution. Colour differentiation of different contamination levels was used to simplify map utilization. To reflect all data in accordance with their spatial distribution and in order to process pictures according to requirements, ArcGis software was used.

2. RESULTS AND DISCUSSION

2.1. Vegetation study

Performed study identified that coenotic composition of vegetation in the area of surveyed tunnels of former Degelen test site is defined by environmental conditions and most of all, by smothered relief. In the area of surveyed tunnel, steppe plants prevail represented by fescue-stipa (fescue (*Festuca valesiaca*), stipa (*Stipa sareptana*, *Stipa lessingiana*)) cenosis with motley grass and petrophyte options.

Motley grass is represented by yellow cress (*Erysimum sisymbrioides*), five-fingers (*Potentilla bifurca*, *Potentilla dealbata*), bedstraw (*Galium boreale*), jerusalem sage (*Phlomis tuberosa*), onion (*Allium subtilissimum*), toadflax (*Linaria vulgaris*), goldendrop (*Onosma simplicissimum*), alyssum (*BertEEVA incana*), speedwell (*Veronica spuria*), pink (*Dianthus squarrosus*), milfoil (*Achillea nobilis*), sedge (*Carex supina*), wormwoods (*Artemisia austriaca*, *Artemisia rupestris*). The following plants are rather rare: knotweed (*Polygonum aviculare*), alfalfa (*Medicago falcata*), vetch (*Vicia cracca*), nightshade (*Solanum dulcamara*), willow herb (*Chamaenerium angustifolium*), blooming sally (*Epilobium adnatum*), goosefoot (*Chenopodium glaucum*), campion (*Silene wolgensis*), sweet clover (*Melilotus albus*), hyssop (*Hyssopus macranthus*). Gramineous plants are represented by the following plants: wild rye (*Leymus angustus*), stipa (*Stipa sareptana*), fescue (*Festuca valesiaca*), wheat-grass (*Agropyron cristatum*), bluegrass

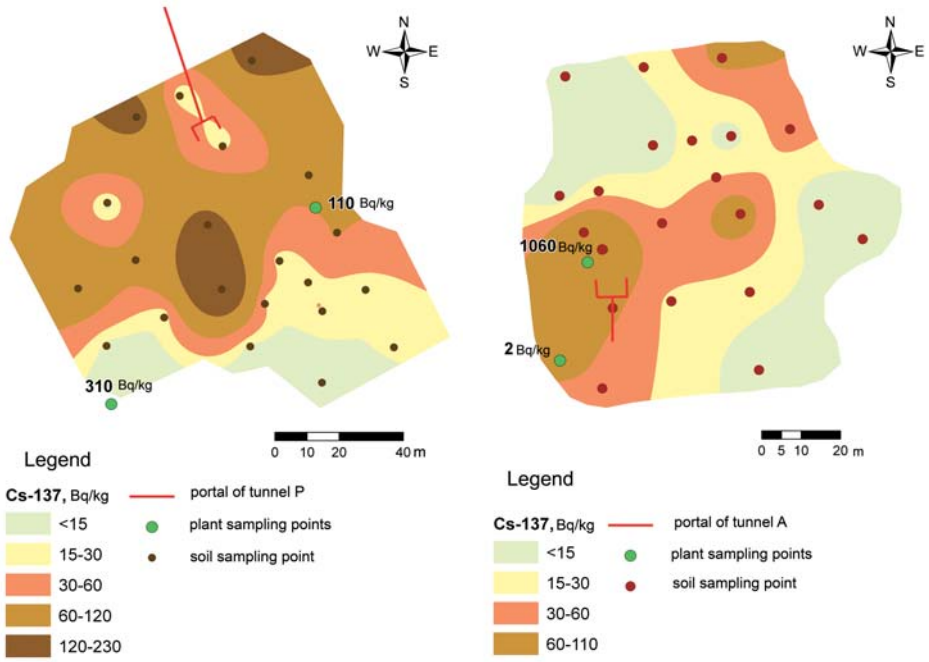
(*Poa stepposa*), foxtail (*Alopecurus arundinaceus*), brome grass (*Bromus inermis*), June grass (*Koeleria cristata*), bristle grass (*Setaria viridis*). For rocky hills bushes are typical – honeysuckle (*Lonicera microphylla*), spiraea (*Spirea trilobata*), pea shrub (*Caragana camilli-schneideri*), brier (*Rosa spinosissima*), ziziphora (*Ziziphora clinopodioides*), and stonecrop (*Sedum hybridum*). In some area the following plants are found – prairie-weed (*Pentaphylloides floribunda*), juniper (*Juniperus sabina*), globe thistle (*Echinops ritro*), speedwell (*Veronica incana*) and five-finger (*Potentilla acaulis*). In lower relief areas with additional moistening, phytocenosis elements are found: reed grass (*Calamagrostis arundinacea*), five-finger (*Potentilla dealbata*), willow herb (*Chamaenerium angustifolium*), reed (*Phragmites australis*), burnet (*Sanguisorba officinalis*), raspberry (*Rubus idaeus*), wormwood (*Artemisia pontica*) and licorice (*Glycyrrhiza glabra*).

Destroyed areas of tunnel portals filled with granite gravel and construction debris are occupied by wormwoods (*Artemisia vulgaris*, *A. sieversiana*, *A. marschalliana*, *A. rupestris*, *A. dracunculus*) and also spiraea (*Spiraea hypericifolia*), birch (*Betula pendula*), ephedra (*Ephedra distachya*) and dodartia (*Dodartia orientalis*). Main plants covering tunnel entrance are wormwoods (*Artemisia marschalliana*, *A. dracunculus*). Rarely sheep fescue and are found (*Festuca valesiaca*), stipa (*Stipa sareptana*), motley grass representatives: speedwell (*Veronica spuria*), pink (*Dianthus squarrosus*) and other. Raising technogenic hills are partially covered with bushes – honeysuckle (*Lonicera microphylla*), pea shrub (*Caragana camilli-schneideri*), brier (*Rosa glabrifolia*), spiraea (*Spiraea hypericifolia*, *S. trilobata*).

2.2. Characteristics of vegetation radioactive contamination and evaluation of possible subsurface radioactive contamination at tunnel portal areas

To study radioactive contamination of vegetation the following trees and bushes with well developed root system were chosen as study objects: birch (*Betula pendula*), spiraea (*Spiraea hypericifolia*, *S. trilobata*), cotoneaster (*Cotoneaster melanocarpa*), brier (*Rosa glabrifolia*, *R. spinosissima*), honeysuckle (*Lonicera microphylla*), raspberry (*Rubus idaeus*), pea shrub (*Caragana frutex*) and osier (*Salix triandra*). In all samples of studied plants specific activity of ^{241}Am, ^{137}Cs and ^{90}Sr was determined. Data on radionuclides content in plants and value ranges of their specific activity (actual and estimated) in soils of portal areas of surveyed tunnel are presented in table (Table 1).

Received data allows evaluating range of possible radionuclides content in soil and making assumptions on presence or absence of subsurface radioactive contamination at portal areas. For example, increased ^{137}Cs concentration in plants in the area of P and A tunnels entrances, in some cases, significantly higher its specific activity in subtopsoil at these tunnels portals areas. (Figure 1).



a b

Figure 1. Areal distribution of ^{137}Cs radionuclides by P (a) and A (b) tunnels

Taking into consideration ^{137}Cs content in plants and possible range of A_c for this radionuclides can estimate its specific activity in soils in the area around P tunnel, it alternates from 11 to 310 kBq/kg, in the areas of tunnel A – from 0.2 to 1,060 kBq/kg, which means that it exceed in two times maximum ^{137}Cs values in surface layer around P and one order – in the area of tunnel A (Table 1). Based on this, presence of certain subsurface contamination is expected in the areas of tunnels B, G, O and E where calculated values of ^{137}Cs specific activity in soil may exceed its estimated content for surface layer by 1-2 orders (Table 1).

Similar situation is also typical for ^{241}Am, which highest specific activity in plants (from 3 Bq/kg to 6 Bq/kg) is found in the area of tunnels Q, A, F and B_1 (Figure 2).

As one can see from the Figures, ^{241}Am concentration in plants in many cases matches its concentration level in soil which contradicts estimated ^{241}Am A_c for Degelen site, which does not exceed <0.0001-0.1. But taken quantitative values of ^{241}Am specific activity in plants received in the areas of tunnels P, B_1, A, F, E and Q, and A_c data, it may be assumed that its concentration in soil in the area of these tunnels may be tens of kBq/kg and more, which is by 2-3 orders higher than received maximum values of ^{241}Am specific activity for topsoil layers (Table 1).

Presence of certain subsurface contamination caused by ^{90}Sr radionuclide is most expected around tunnels P, N and R, where calculated range of its specific activity in soil is by 1-2 orders higher than ^{90}Sr concentration established for topsoil layer. Presence of such subsurface contamination is also possible in the areas of tunnels A, G, O and I, where estimated values of ^{90}Sr specific activity in soil is also somewhat higher than established ones (Table 1).

Table 1.

Specific activity of ^{241}Am, ^{137}Cs and ^{90}Sr in studied plants, specific activity of ^{241}Am, ^{137}Cs and ^{90}Sr in soil at tunnel portals areas

Tunnel reference label	Plant species	Point	Specific activity, Bq/kg								
			^{241}Am			^{137}Cs			^{90}Sr		
			plants	soil		plants	soil		plants	soil	
				actual	assumed*		actual	assumed*		actual	assumed*
B	Spiraea hypericifolia	1	<0.9	<1 - 320	<9000	5 ± 1	4 - 100	<0.1 - 5000	7 ± 1	<100 - 1300	7 - 300
	Spiraea trilobata	2	<0.6			<1.3			30 ± 1		
P	Cotoneaster melanocarpa	1	<0.8	<1 - 110	<20000	110 ± 10	5 - 2200	11 - 310000	34 ± 2	<100 - 330	34 - 3950
	Betula pendula	2	2 ± 1			310 ± 10			395 ± 5		
C	Spiraea hypericifolia	1	<0.8	<1 - 50	<8000	2 ± 1	3 - 48600	0.1 - 2000	7.2 ± 0.4	<100 - 2480	7 - 250
	Spiraea hypericifolia	2	<0.3			0.6 ± 0.2			25 ± 1		
N	Betula pendula	1	<1	<1 - 20	<13000	99 ± 4	45 - 49000	8 - 99000	165 ± 2	<100-3200	165 - 66000
	Rosa glabrifolia	2	<1.3			77 ± 4			6600 ± 100		
B_1	Spiraea hypericifolia	1	<1.4	<0.3 - 160	<60000	<1.3	<1 - 2100	<0.2	26.8 ± 0.4	<100 - 13000	27 - 370
	Lonicera microphylla	2	6 ± 2			<1.8			37 ± 1		
A	Spiraea trilobata	1	<1	<0.3 - 140	<30000	2 ± 1	9 - 459400	0.2 - 1060000	26 ± 1	<100 - 25000	26 - 28500
	Betula pendula	2	3 ± 2			1060 ± 10			2850 ± 30		
F	Rosa glabrifolia	1	3 ± 2	<1 - 150	<30000	4 ± 2	6 - 28000	<0.2 - 4000	79 ± 1	<100 - 8700	79 - 880
	Spiraea trilobata	2	<1.4			<1.6			88 ± 1		
G	Spiraea hypericifolia	1	<0.6	<1 - 300	<13000	2 ± 1	14 - 450	<0.2 - 2000	30 ± 1	<100-540	30 - 610
	Rubus idaeus	2	<1.3			<1.8			61 ± 1		
O	Spiraea trilobata	1	<0.8	<1 - 5	<9000	3 ± 1	<0.7 - 110	<0.1 - 3000	22 ± 1	<100	22 - 350
	Spiraea trilobata	2	<0.9			<1			35 ± 1		
I	Spiraea hypericifolia	1	<1.3	<1 - 470	<30000	<1.2	4 - 180	<1200	22 ± 1	<100	20 - 220
	Spiraea hypericifolia	2	<3			<0.8			20 ± 1		
E	Rosa glabrifolia	1	<1.4	<1 - 410	<20000	<1.5	4 - 100	<0.2 - 2000	23 ± 1	<100 - 520	8 - 230
	Rosa spinosissima	2	2 ± 1			2 ± 1			8.4 ± 0.3		
R	Betula pendula	1	<0.9	<1 - 3	<14000	<1.3	4 - 130	<1300	12 ± 1	<100-400	12 - 19100
	Spiraea hypericifolia	2	<1.4			<0.9			1910 ± 20		
Q	Caragana frutex	1	<1.1	<1 - 60	<40000	<1.7	<2 - 640	<1800	4 ± 1	<100 - 730	4 - 80
	Salix triandra	2	4 ± 2			<1.8			8 ± 1		

Remark: * Specific activity of radionuclide in soil = Specific activity of radionuclide in plants / A_c, where

A_c ^{241}Am = <0.0001-0,1;

A_c ^{137}Cs = 0.001-10;

A_c ^{90}Sr = 0.1-1

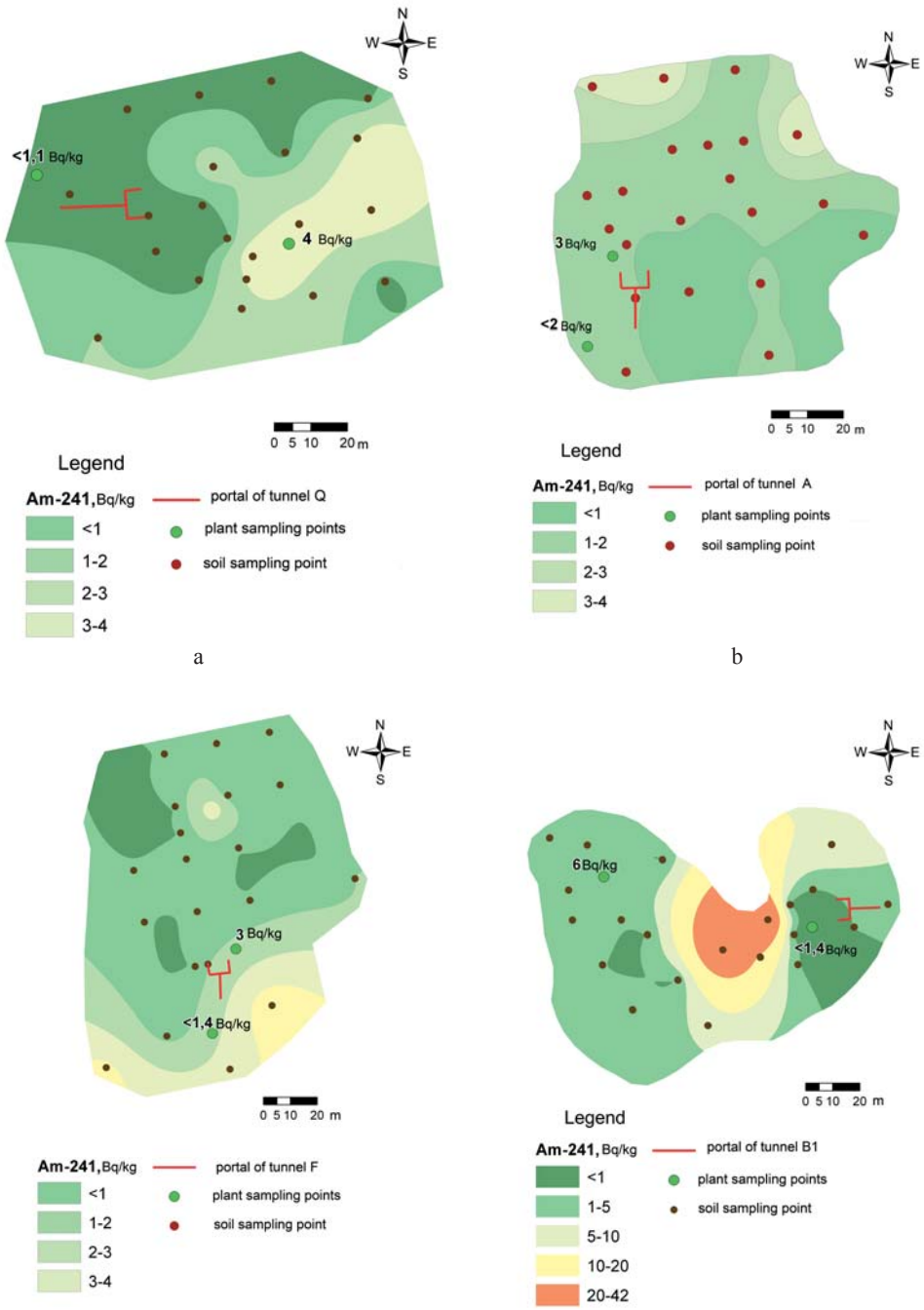


Figure 2. Areal distribution of ^{241}Am radionuclide at the area of tunnels Q (a), A (b), F (c) and B_1 (d)

CONCLUSIONS

Based on the performed study, it has been identified that in the area of all surveyed tunnels, mainly steppe vegetation prevails represented by fescue feather-grass species with motley grass and petrophytes and in case of lower relief areas, characterized by additional moistening, pratal phytocenosis is registered. Natural growth of vegetation on destroyed tunnel entrances filled with granite gravel and partially with construction debris is represented mainly by southernwood, bushes and weeds.

Presence of radionuclides in studied plants, especially presence of ^{90}Sr, which is specified by a smallest range of A_c of this radionuclide, in some cases is an indicator of contamination in deeper soil layers of corresponding territories. This circumstance confirms possibility of using plants as indicators of subsurface radioactive contamination which in its turn may become an important addition to a radioactive contamination study of the surface layer of soil around tunnels of former test site "Degelen".

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ӨСІМДІКТЕРДІ БҰРЫҢҒЫ "ДЕГЕЛЕН" СЫНАҚ АЛАҢДАРЫНДАҒЫ ШТОЛЬНЯЛАРДЫҢ ПОРТАЛАЛДЫ АЛАҢДАРЫНЫҢ ЖЕРБЕТКІ РАДИОАКТИВТІ ЛАСТАНУ ИНДИКАТОРЛАРЫ РЕТІНДЕ ПАЙДАЛАНУ

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Мақалада бұрынғы "Дегелең" сынақ алаңдарындағы кейбір штольнялардың порталалды алаңдарының ауданындағы өсімдік жамылғысы мен оның радиоактивті ластану сипатын зерттеу нәтижелері ұсынылған. Өсімдіктерді порталалды алаңдардағы жербеткі ластанудың индикаторлары ретінде қолдану мүмкіндігі бағаланған.

Түйін сөздер: ядролық сынақтар, штольнялар, радиоэкология, радиоактивті ластану, радионуклидтер, өсімдік жамылғысы, өсімдіктер, жинақталу коэффициенті (Жк).

ИСПОЛЬЗОВАНИЕ РАСТЕНИЙ В КАЧЕСТВЕ ИНДИКАТОРОВ ПРИПОВЕРХНОСТНОГО РАДИОАКТИВНОГО ЗАГРЯЗНЕНИЯ НА ПРИПОРТАЛЬНЫХ ПЛОЩАДКАХ ШТОЛЕН БЫВШЕЙ ИСПЫТАТЕЛЬНОЙ ПЛОЩАДКИ "ДЕГЕЛЕН"

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В статье представлены результаты исследования растительного покрова и характера его радиоактивного загрязнения в районе припортальных площадок некоторых штолен бывшей испытательной площадки "Дегелен". Оценена возможность использования растений в качестве индикаторов приповерхностного загрязнения на припортальных площадках.

Ключевые слова: ядерные испытания, штольни, радиоэкология, радиоактивное загрязнение, радионуклиды, растительный покров, растения, коэффициент накопления (Кн).

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SPECIES OF RADIONUCLIDES IN SOILS OF WATERCOURSE ECOSYSTEMS AT DEGELEN MASSIF

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The paper presents data on studying ^{137}Cs, ^{90}Sr, $^{239+240}$Pu and ^{241}Am species in meadow soils of watercourse ecosystems of the tunnels #176 and #177 of "Degelen" testing ground of former Semipalatinsk nuclear test site. It was revealed that despite the spatial delimitation, there is considerable variation for radionuclide species in soil. It is shown that ^{137}Cs and $^{239+240}$Pu radionuclides in soils of studied ecosystems are mainly in the soil-bound form, while ^{90}Sr and ^{241}Am present there in exchange and mobile forms. Correlation analysis made us possible to assess the possible influence of physical and chemical properties of studied soils on content of ^{137}Cs and ^{90}Sr radionuclides.

Key words: soil, species of radionuclides, sequential leaching, physical and chemical properties of soil, tunnel, Degelen mountain massif.

INTRODUCTION

Determining factor of radionuclides migration ability in the system soil - plant, soil - surface and ground water and causing potential danger to human is radionuclide species in soils.

Conventional approach to determination of mobility and bioavailability of radioactive contaminants in soil involves sequential extraction methods. These methods imply a sequence of progressively more harsh chemicals and it is assumed that each of them will selectively leach the contaminants bound in specific components of the soil. Selectivity and reproducibility of chemical extraction procedures vary depending on the type of extractant, characteristics of the subject studied; so one needs to keep this in mind when provides valuation of bioavailability. Ratio of individual forms of artificial radionuclide compounds is to be studied in a unified system, which can be used to obtain an integral estimate of radionuclides mobility in soil.

Objective of this study was to investigate the distribution of artificial radionuclides in the meadow soils of spatially bounded watercourse ecosystems of Degelen Mountain between fractions that are different in physical and chemical mobility and bioavailability employing sequential extraction of radionuclides.

1. MATERIALS AND METHODS

As the subject of the investigation we have chosen watercourse ecosystem at tunnels #176 and #177 of Degelen massif. Investigated ecosystem of the watercourse at tunnel #176 is a left-handed part of Baitles brook valley, separated from the mainstream by small elongated adrift upland with outcrops of rocks. Streambed width of the watercourse from the tunnel is mostly less than 1m. Ecosystem territory of the tunnel #177 is a right-handed part of the tributary of Uzynbulak brook valley separated from the mainstream by a small elongated adrift upland and body of road. Width of the watercourse at tunnel #177 is mostly less than 1 m.

In the area of tunnel # 176, samples were taken along the watercourse streambed. We incorporated 30 research sites with soil sampling to a depth of 20 cm and with an area of not less than 100 cm^2. Distance between the most distant points was about 630 m. Width of the watercourse streambed, in general, did not exceed 1 m. The figure (Figure 1) shows the layout of soil sampling points.

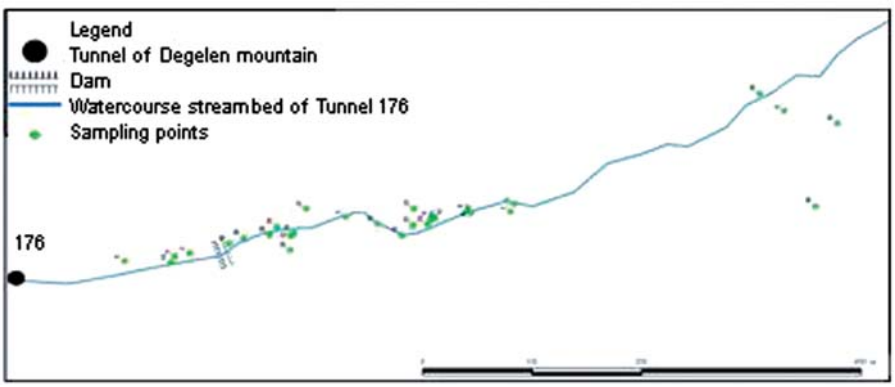


Figure 1. Sampling locations near the tunnel #176

Scheme of soil sampling points in the ecosystems of tunnel #177 is shown in the figure (Figure 2). Samples were also taken along the streambed of the watercourse. 36 research sites were organized with soil sampling to a depth of 20 cm and with an area of not less than 100 cm^2. Sampling points of soil were tied to the points of plant sampling, taken to study parameters of radionuclide accumulation in plants.

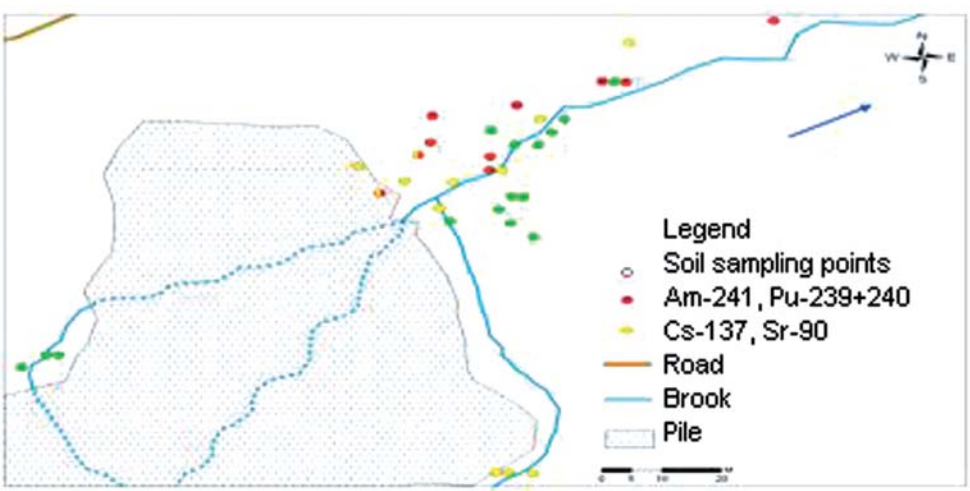


Figure 2. Sampling locations near the tunnel #177

Investigation of radionuclide species was done by sequential extraction; the works flow chart is shown in the figure below (Figure 3).

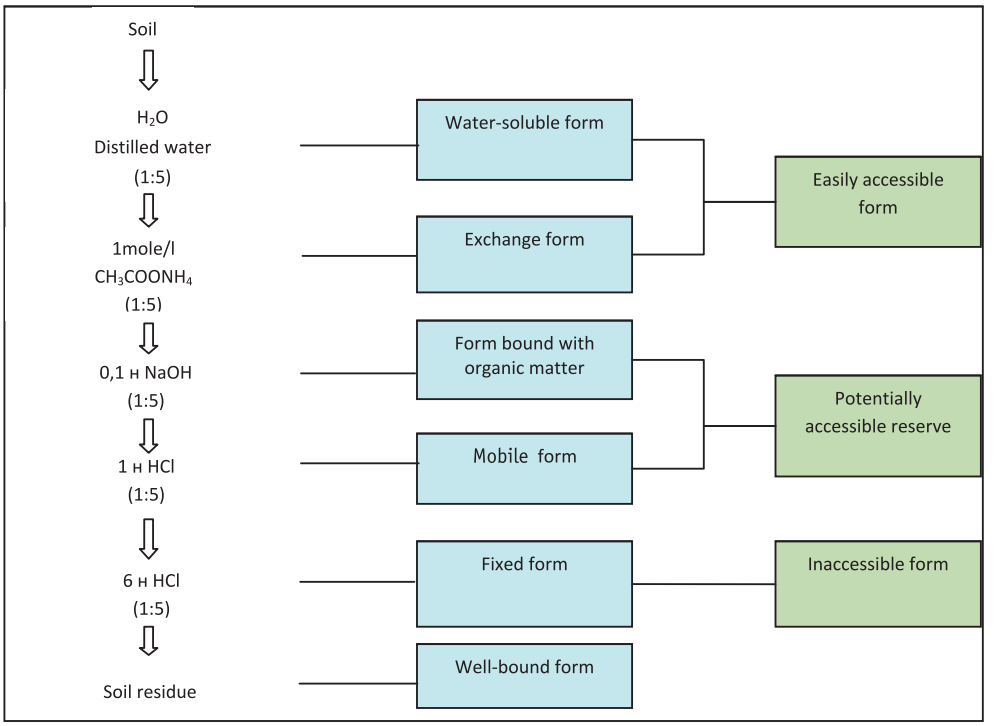


Figure 3. Algorithm for determination of radionuclide species in soil

Based on the objectives of radioecological research, the study of radionuclide species was carried out by determining the forms influencing the intensity and mechanisms of radionuclide migration. To assess the bond strength of radionuclides ^{90}Sr, ^{137}Cs in ecosystem soils in the area of tunnel #176 of ecosystems studied, we determined relative content of different radionuclides in soil by sequential extraction with water (water-soluble), solution of ammonium acetate (exchange), 1M HCl (mobile) and 6M HCl [1, 2, 3, 4] (fixed). Ratio of soil and leaching solution at all stages of the experiment was to 1:5. In studies of radionuclide forms of ^{90}Sr, ^{137}Cs in ecosystem soil of the tunnel #177 we used a modified sequential extraction scheme.This scheme included single out and determination of relative amount of additional new forms of radionuclides - forms bound with soil organic matter (isolated 0.1n with solution of NaOH), in particular, in the fractions I - humic acid and fulvic acid (HA and FA) and their salts - free or loosely bound with the mineral part of soil, soluble in 0,1 n solution of NaOH (by Tyurin) [5]. As it is known soluble humic substances can interact with cations of heavy metals, as well as with artificial radionuclides, forming humates and fulvates. These reactions are subject to the laws of formation and behaviour of simple or complex heteropolar salts and have an impact on migration, accumulation and penetration of toxic elements into plants. More reduced modified sequential extraction scheme was applied to study radionuclides forms ^{241}Am and $^{239+240}Pu$. This scheme provides for the exclusion of water-soluble forms (contents of which, as shown by studies of soil at tunnel #176, is below the minimum detectable activity of the used techniques) and fixed

forms of radionuclides (extracted with 6M HCl solution), as a rule, does not affect the migration of radionuclides.

To determine the radionuclide forms soil samples were pre-dried and sieved through a sieve with a mesh size of 1 mm. Of the prepared samples we took analytical samples weighing 100 g by quartering.

Content of ^{137}Cs and ^{241}Am in soil samples and extracts was determined by instrumental gamma-ray spectrometry. Specific activity of ^{90}Sr in the soil samples was determined by non-destructive method - a direct instrumental measurement of effective specific activity of ^{90}Sr on beta-spectrometer with "Progress" software. Volumetric activity of ^{90}Sr and $^{239+240}Pu$ in the extract samples were determined by radiochemical method.

In soil samples along with the content of radionuclides we determined physicochemical parameters:

- pH value of the aqueous extract;
- absorption bases, Ca, Mg (trilonometry);
- humus (carbon) by the method of Tyurin in modification of Nikitin;
- content of water-soluble substances (HCO_3, Cl, SO_4, Ca, Mg, K, Na);
- granulometric analysis by Kachinskiy`s method;

Determination of these indicators was conducted by conventional methods [6, 7, 8, 9].

2. RESULTS AND DISCUSSION

2.1. Characteristics of soils in the studied ecosystems

Out of the general description of soil and the data obtained, we can note that soils at 2 sites belong to the meadow soils. Soils are shallow. Thickness of the soil layer in the streambed does not exceed 40-60 cm, in the coastal zone - 20-40 cm. They have quite high contents of humus, especially in the center of streambed, well washed from soluble salts and carbonates. pH of the aqueous suspension is rarely neutral, alkalescent or alkaline. Cation of Ca dominates in the soil-absorbing complex. By its mechanical structure, the soil of the watercourse ecosystem around the tunnel #176 is mainly loamy and sandy loam, rarely medium loam, while at the site of the watercourse at tunnel #177 – medium and heavy loam dominate among the unconsolidated sediments. This is due to a decrease in slope, and as a consequence, lowers drainage condition of the area. All investigated meadow soils of the valleys differ by the incorporation on the entire soil profile of a large landwaste: grit, coarse sand and coarse dust. It should be noted that in meadow soil in ecosystem of the watercourse at tunnel #176 content of silt particles is in the range of 0.1-11%, and in the soil in ecosystem of tunnel #177 subjected to less drainage, silt content reaches 19-31%. Thus, decrease in slope of the area, and, accordingly, the flow rate leads to a change in the natural drainage condition of the area, which is reflected in the soil indicators: number of fine particles increases, there is accumulation of soluble salts in the surface horizons, number of fine humus increase.

The following table (Table 1) shows the results of determination of physical and chemical parameters of the investigated soils.

Table 1.

Physical and chemical parameters of the soils investigated

Soil characteristics	tunnel 176		tunnel 177	
	interval	averagee	interval	averagee
Concentration of organic matter, C, %	7 - 23	14	11 - 21	16
pH of aqueous suspension (actual acidity)	6.8 – 7.9	7.5	7.1 - 7.9	7.5
Amount of salt, %	0.02 – 0.3	0.045	0.05 - 0.6	0.2
Total of exchange bases, mg-eq/100 g	17 – 40	27	28 - 52	41
Granulometric composition: alphitite content (soil particles with size of <0.01 mm), % including: silt particles (<0.001 mm), % fine dust (0.001-0.005 mm), % medium dust (0.005-0.01 mm), % coarse dust particles content (0.01-0.05 mm), %	 15 – 37 0.1 – 11 0.2 – 12.3 3.3 – 14 12.6 – 41	 23 5 9 9 29	 32 - 59 19 - 31 3.4 - 25 3.3 - 15.4 3.2 - 48.4	 42 24 9.4 9.4 35
Specific activity of ^{137}Cs in soil, Bq/kg	26 – 450,0	27,4	16 – 8,7	1,9
Specific activity of ^{90}Sr in soil, Bq/kg	610 – 33,0	11,9	1,8 – 117,1	72,0
Specific activity of ^{241}Am in soil, Bq/kg	-	-	200 – 680	400
Specific activity of $^{239+240}$Pu in soil, Bq/kg	-	-	2,9 – 12,2	6,3

2.2. Ratio of radionuclide species

In soil samples, taken in the ecosystem of watercourse at tunnel #176, determination of the species was carried out for two radionuclides - ^{90}Sr and ^{137}Cs since they mainly determine the radioactive contamination of the area. Out of the 30 soil samples analyzed for ^{90}Sr, complete quantitative data were obtained for 14 samples, for ^{137}Cs - for 10 samples.

Species ratio for ^{90}Sr and ^{137}Cs is presented in the table (Table 2). Radionuclide concentrations are expressed as a percentage of the total content of all studied species.

Table 2.

Species of ^{90}Sr and ^{137}Cs in soil of ecosystems in vicinity of tunnel #176, % of the total content of all forms

Species	^{137}Cs, % (n=10)		^{90}Sr, % (n=14)	
	range	average	range	average
Water-soluble + exchange	0.5 – 35.6	13.1	32.7 – 65.1	54.5
Mobile	1.4 – 24.4	10.2	14.7 – 54.5	35.2
Fixed	15.1 - 77.4	57.3	0.7 – 17.1	6.4
Well-bound	0.1 – 79.4	19.4	1.73 – 7.6	3.9
n = number of quantitative values				

Investigation results show that the main content of ^{137}Cs in soil of ecosystems at the tunnel #176 is characterized by the lowest bioavailability - fixed (57.3 %) and well-bound (19.4 %). Water - soluble form is an average of 13.1 %, mobile – 10.2 % of the total content of all forms.

For ^{90}Sr a different pattern of distribution is typical. Major content of this radionuclide is concentrated in easily accessible forms (water + exchange – 54.5 %, mobile – 35.2 %, on average). Fixed and well-bound forms contain much less radionuclide, on average, 6% and 4%, respectively.

Investigation of radionuclide species in soil of the ecosystem at tunnel #177 was held for four radionuclides - ^{90}Sr, ^{137}Cs, ^{241}Am and $^{239+240}$Pu. Out of the 36 taken soil samples to study the species of ^{90}Sr, ^{137}Cs in soil 12 samples were analyzed, as for ^{241}Am and $^{239+240}$Pu - 10 samples of soil. Upon that complete quantitative data on the content of radionuclide forms of ^{137}Cs were obtained for 3 samples, ^{241}Am - for 6 samples, for $^{239+240}$ Pu – for all the 3 samples in connection with small yields of $^{239+240}$Pu during the radiochemical separation. As mentioned above, in the soil of ecosystem in the area of tunnel #177 we determined exchange, organic, mobile, fixed and well-bound forms of ^{90}Sr and ^{137}Cs. For radionuclides ^{241}Am and $^{239+240}$Pu we used reduced scheme to extract exchange, organic, mobile and well-bound forms.

The table below (Table 3) shows the assessing data on ratios of ^{90}Sr, ^{137}Cs, ^{241}Am and $^{239+240}$ Pu forms in soils in vicinity of the tunnel #177.

Table 3

Radionuclide species (^{90}Sr, ^{137}Cs, ^{241}Am and $^{239+240}$Pu) in soil of ecosystems in the area of tunnel #177, % of the total content of all forms

Forms	^{137}Cs, % (n=3)		^{90}Sr, % (n=12)		^{241}Am, % (n=6)		$^{239+240}$Pu, % (n=3)	
	range	average	range	average	range	average	range	average
Water-soluble + exchange	0.1 - 1.9	0.8	43.0 - 69.5	53.0	2.4 - 5.6	3.7	0.6 - 3.8	1.6
Organic	0.1 - 0.9	0.4	1.4 - 2.4	2.0	0.8 - 4.7	2.2	2.1 - 12.5	6.3
Mobile	0.6 - 1.6	1.1	24.1 - 40.0	33.0	62.8 - 83.3	77.5	0.1 - 14.9	6.2
Fixed	11.8 - 14.9	13.4	4.4 - 10.9	8.0	-	-	-	-
Well-bound	80.8 - 86.2	84.1	2.2 - 5.7	4.0	10.4 - 30.7	16.6	72.1 - 92.4	85.9
n = number of quantitative values								

For ^{137}Cs there is pattern typical for this radionuclide. The table shows that easily accessible caesium content on average is less than 1 %. Main content of caesium is in the well-bound form (84.1 %), less high value characterizes the fixed form (13.4 %). ^{137}Cs presents in mobile and exchange forms in trace amounts and in form bound to HA and FA (1,1 %, 0,8 % and 0,4 %, respectively). Predominance of easily accessible forms is typical for ^{90}Sr (water-soluble + exchange - 53 %, mobile - 33 %). Fixed forms are characterized by lower values of radionuclide content - 8 %, well-bound - 4 %, on average. Content of organic forms of radionuclide ^{90}Sr is insignificant - 2 % of the total content of all forms. For a complete study on the distribution of radionuclides in the organic matter it is necessary to define the group and fractional composition of humus. Results obtained earlier, provide an opportunity to make a preliminary assessment of the group and fractional composition of humus in the soils studied. Content of humic acids exceeds the fulvic acids. Majority of them is related

with moving calcium, less significant relates to the free humic acids and only a small amount is closely bound with the mineral part of soils.

According to the investigation, radionuclide $^{239+240}$Pu in soils of tunnel #177 in significant quantities, 86 % of all forms, is contained in well-bound forms. Radionuclide content in mobile and organic forms is equally low, bound with HA and FA. A minor part of $^{239+240}$ Pu, on average, 2 % of all forms, is in exchange form.

Migration regularities of $^{239+240}$Pu and ^{241}Am are similar, however, in contrast to $^{239+240}$Pu, ^{241}Am has a fairly good solubility and, therefore, has greater mobility in the environment (as compared to plutonium). Preliminary data on the ratios of radionuclide forms of ^{241}Am can say that the radionuclides in the soil is mainly in mobile form.

Comparative study indicates that in meadow soil in ecosystem of tunnel watercourse the ratio of radionuclides forms of ^{90}Sr and ^{137}Cs are significantly different. ^{137}Cs in the soil is mainly in well-bound state. In contrast to ^{137}Cs, ^{90}Sr is in the exchange form; apparently, determine the good bioavailability of ^{90}Sr.

Data obtained show that despite the fact that the studies were conducted within a spatially limited ecosystem, one type of soil and other near-identical conditions, there is considerable variation in the contents of these radionuclide forms.

For a more graphic representation the results are shown as a histogram of the empirical frequency distributions of radionuclide species. The figure below (Figure 4) is the histogram of distribution of the results by frequency of ^{90}Sr and ^{137}Cs forms in the soils studied (these radionuclides were studied better than others).

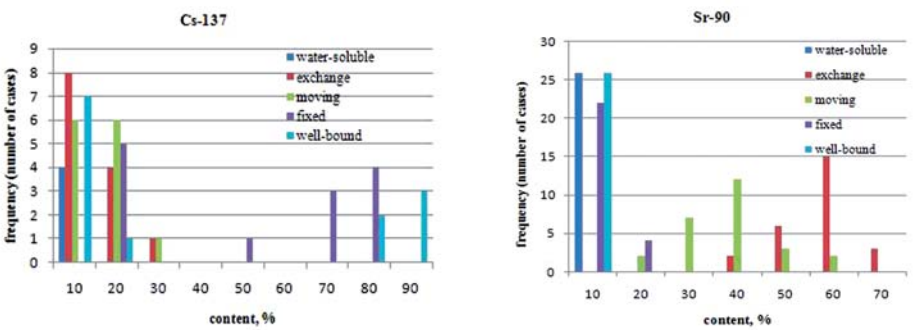


Figure 4. Frequency distribution of the ^{90}Sr and ^{137}Cs species in soils of the ecosystem of the tunnels #176 and #177

The widest range of variation is typical for the content of the fixed forms of ^{90}Sr, which reaches two orders of magnitude. The variation of range of water-soluble forms lies within an order.

A more significant variation in the content of forms is typical for caesium, in comparison with ^{90}Sr. Well-bound, exchange and mobile forms have the largest ranges of variation, reaching two orders of magnitude.

To assess the variability of the contents of ^{137}Cs and ^{90}Sr species and to solidify the shown empirical distributions, it makes sense to give some statistical numerical indicators. The most common and illustrative are average values, standard deviations and coefficients of variation (Table 4).

Table 4.

Summarized variation-statistical indicators of the content of ^{137}Cs and ^{90}Sr species in meadow soil in watercourse ecosystems of tunnels #176 and #177

Radionuclide forms	Tunnel #176						Tunnel #177		
	^{137}Cs			^{90}Sr			^{90}Sr		
	A_{am}, %	σ	K_{var}, %	A_{am}, %	σ	K_{var}, %	A_{am}, %	σ	K_{var}, %
water-soluble	2.14	2.9	140	1.1	0.5	46	1.4	1.0	76
exchange	13.1	8.9	69	53.4	8.9	17	51.6	6.6	13
organic	-	-	-	-	-	-	1.8	0.4	23
mobile	10.2	6.2	60	35.2	11.5	33	33.1	4.7	14
fixed	57.4	23.8	42	6.4	4.7	73	8.2	1.6	20
well-bound	19.3	30.2	156	3.9	1.8	47	3.9	1.0	26
A_{am} – arithmetic middling, σ – standard deviation, K_{var} – coefficient of variation									

As the table shows, there is considerable variability of content of both radionuclides forms in soil of tunnel #176, particularly it is expressed in the contents of water-soluble forms of ^{137}Cs (K_{var} = 140%), and well-bound forms (K_{var} = 156%) and fixed (K_{var} = 71%) forms of ^{90}Sr. Soils of tunnel #177 are characterized by considerable variability of contents of water-soluble forms of ^{90}Sr (K_{var} = 76%).

2.3. Assessment of possible impact of physical and chemical properties of meadow soils on radionuclide species in soil

Biological mobility of radionuclides (ability to migrate through the food chain) depends, on the one hand, on their physical and chemical properties, on the other hand, on properties of the soil, among which the crucial one is its type, mineral composition, acidity, organic matter content, humidity, duration of its use in agro-ecosystems, etc.

Assessing the impact of physical and chemical properties of soils on the relative content of species was done only for ^{137}Cs and ^{90}Sr in soils of the two studied ecosystems at tunnels # 176 and #177 due to the lack of quantitative data on $^{239+240}$Pu and ^{241}Am.

The table below (Table 5) shows the correlation coefficients between some soil characteristics and content of radionuclide forms of ^{90}Sr and ^{137}Cs in soils of tunnels #176 and #177.

Table 5.

Correlation of relative content between radionuclide species and physical-chemical properties of soil

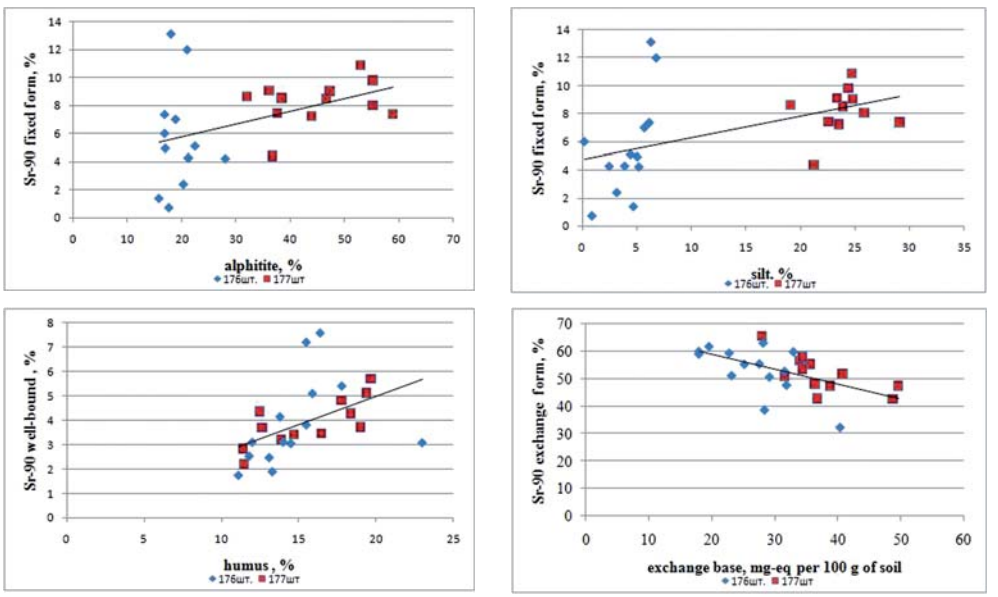
Forms	Humus, %		pH		Total salts, mg-eq per 100 g of soil		Total exchange bases, mg-eq per 100 g of soil		Alphitite, %	
	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
water-soluble	0.39	*	0.22	*	0.42	*	0.25	*	-0.004	*
exchange	-0.36	-0.19	-0.36	0.49	-0.14	-0.45	-0.58	-0.07	-0.03	-0.36
organic	0.06	-	-0.27	-	0.12	-	-0.31	-	0.29	-

Forms	Humus, %		pH		Total salts, mg-eq per 100 g of soil		Total exchange bases, mg-eq per 100 g of soil		Alphitite, %	
	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
mobile	0.14	-0.15	0.3	0.36	-0.03	-0.57	0.24	-0.18	-0.2	-0.31
fixed	0.12	-0.30	0.02	0.44	0.07	-0.62	0.34	-0.52	0.28	-0.67
well-bound	0.48	0.28	0.02	-0.50	0.04	0.65	0.27	0.43	0.08	0.63
* – no quantitative data										

Positive correlation is observed between the contents of well-bound forms of ^{90}Sr and humus content in soils of the two studied ecosystems, correlation coefficient is 0.48. Also there is a trend for positive influence of soluble salts content on the content of water-soluble forms of ^{90}Sr (K_{corr} = 0.42). We have revealed the tendency of inverse effect of absorbed base content on the content of ^{90}Sr exchange forms. When content of exchange bases increases in the soil, content of exchange forms of ^{90}Sr decreases (K_{corr} =- 0.58).

The research results allow assessing the influence of physico-chemical properties of soils on the behaviour of ^{137}Cs. The most significant correlations are observed between the content of well-bound radionuclide forms and content of water-soluble salts and alphitite in the soil.

Identifying the correlation relationship between physicochemical properties and ratios of radionuclide forms of ^{90}Sr is possible when summarizing the results on the two tunnels. The figure below (Figure 5) shows how ^{90}Sr species depend on physical and chemical properties of soil near two tunnels.



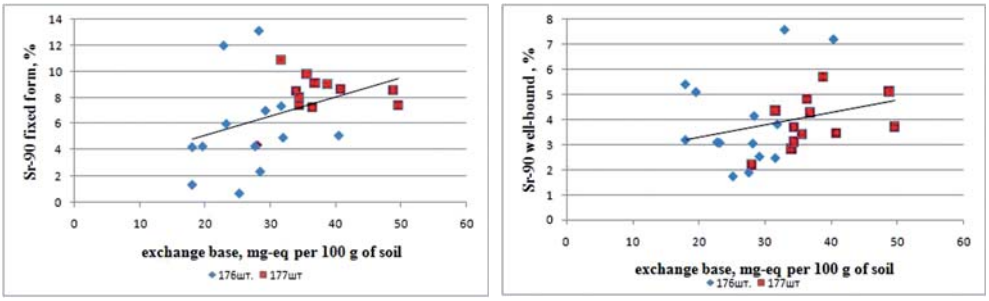


Figure 5. ^{90}Sr species as a function of physical and chemical properties of soils

Data show that granulometric composition has definite impact on the content of ^{90}Sr species. In particular, content of alphitite and silt fraction has a direct influence on the content of fixed forms of the radionuclide. Granulometric composition of soils largely determines their absorbing capacity. Sorption capacity of soil is directly proportional to the degree of dispersion of soil particles, since separate fractions of soils differ not only in size of the particles, but also in physical, chemical properties, mineralogical composition. When particle size decreases, the content of silicon oxide decreases, amount of humus and exchange cations of calcium, magnesium and potassium also decreases. When fraction of soil decreases in size, the radionuclides are more strongly bound. Radionuclides of clay fractions are bound most firmly.

Our investigation shows that humus content also has a direct influence on the content of well-bound forms. It is known that the organic matter has significant influence on behaviour of ^{90}Sr in the soil. Distribution and mobility of ^{90}Sr is largely determined by the quantitative and qualitative composition of humus. ^{90}Sr preferably binds to fulvic acid. Strontium is present in soils, mainly in the form of complex systems, which include calcium, iron and aluminium. Depending on whether they are in a soluble or insoluble state in the soil, the proportion of strontium is increasing or decreasing in a moving state.

Issue on the impact of exchange bases on the ratio of radionuclide forms is poorly understood. Our research results show that the content of exchange bases in the soil has the inverse effect on the mobility of radionuclide ^{90}Sr. Thus, increase in content of exchange bases leads to an increase in the content of exchange forms of radionuclide and a decrease in its inaccessible forms - fixed and well-bound.

It is possible that the correlation between concentrations of radionuclides and various forms of soil particles are a kind of indirect indicator, and, most likely, the reason lies in the contrast of mineralogical and chemical composition, absorption capacity, sorption capacity and other characteristics of different soil particles. Quantifying the effect of individual soil properties for the content of radionuclides forms is a complex task since the majority of physical and chemical parameters are closely linked together and the impact of each property depends on the influence of the entire complex.

Soil acidity has huge impact on the mobility of radionuclides, but in the studied soils this pattern has not been detected due to insufficient range of variation of this factor within the considered ecosystems.

2.4. Assessment of possible impact of some macro-elements and stable isotope forms on radionuclide species in soil

Presence of exchange cations in soil – element-carrier and stable isotopes – may have great impact on the availability of radionuclides to plants.

Being isotopes of chemical elements, radionuclides are characterized by the same chemical properties as the stable isotopes of these elements, as well as the chemical properties of elements-analogs. However, the identity of radionuclide behaviour and their stable analogs can be observed only in the case of equilibrium distribution of physical and chemical species of introduced forms of radionuclides and natural forms of their stable analogs in the soil, i.e. when the isotopic exchange in phases and components of the soil is complete. Therefore, the time to reach equilibrium distribution depends not only on physical and chemical properties of soil, reaction and soil solution, amount and composition of soil colloids, moisture content, etc., but, in the first place, on the initial physical-chemical state of radionuclides and speed of its transformation under specific landscape-geochemical conditions.

To compare the behaviour of radionuclides with the behaviour of macro-elements and stable isotopes in soils the table (Table 6) shows the data on content of ^{90}Sr, ^{137}Cs radionuclide species, macro-elements and stable isotopes, in % of total content of all forms.

Table 6.

^{90}Sr, ^{137}Cs species, macro-elements and stable isotopes in soils in the ecosystems of the tunnel #176%, % of the total content of all forms

Forms	Alcali metal				Alkaline-earth metals				
	^{137}Cs, %	Cs, %	K, %	Na, %	^{90}Sr, %	Sr, %	Ca, %	Mg, %	Ba, %
Water-soluble	2.1	*	0.3	0.5	1.1	1.2	1.2	0.5	0.04
Exchange	13.1	0.6	2.4	0.4	53.4	29.4	32.8	6.4	2.1
Mobile	10.2	0.4	0.7	0.1	35.2	15.9	10.6	4.9	5.0
Fixed	57.4	16.9	4.4	0.2	6.4	9.4	8.1	40.2	2.8
Well-bound	19.3	82.0	92.3	98.9	3.9	43.9	47.2	48.0	90.0
* – no quantitative data									

No clear identity of elements behaviour, as expected, from the data has been revealed, but a definite trend is seen for both radionuclides. Behaviour of radionuclide ^{137}Cs, to some extent, is similar to the behaviour of its chemical analogue K and Cs stable isotope. It is quite understandable that the majority of such elements as Na, K, and Cs are in well-bound forms, usually in the crystal lattice of the primary and secondary minerals. The main content of ^{137}Cs is observed in the fixed forms that are also inaccessible to plants. Significant difference of ^{137}Cs is in its relatively higher content in easily-accessible form.

It is known that the binding and distribution of strontium radionuclides in the soil components, is mainly determined by the laws of isotopic carrier behaviour - stable strontium, as well as chemical analogue - stable calcium. As the table shows, the behaviour of stable Sr is almost identical to the behaviour of Ca. The highest content in soil is in the well-bound form, on average 44-47% of the total, 31-34% are easily accessible forms (water soluble + exchange); content of fixed forms and mobile forms is much lower (8.1-9.4% and 10.6-15.9%, respectively). Radionuclide ^{90}Sr is more soluble and, therefore, has greater

bioavailability than its chemical counterparts. Proportion of well-bound forms of radionuclide ^{90}Sr is exactly the contrary, minimal, only 4% of the total content of all forms. The most significant radionuclide forms of ^{90}Sr are easily accessible (55%) and mobile (35%), content of the fixed form is much lower (6%).

CONCLUSION

The studies yielded the following results:

There have been revealed that the ratios of species of ^{137}Cs and ^{90}Sr in meadow soil in ecosystems of watercourses at the tunnels are significantly different. ^{137}Cs in the soil is mainly in well-bound and fixed states. In contrast to ^{137}Cs, ^{90}Sr main content accounts for the exchange forms, apparently, determining the good bioavailability of ^{90}Sr.

Preliminary results revealed the difference in the behaviour of transuranic radionuclides ^{241}Am and $^{239+240}$ Pu in soils. Main content of radionuclide ^{241}Am is concentrated in mobile form, $^{239+240}$ Pu - in well-bound form that is inaccessible for plants.

Within a single spatially limited ecosystem, a single soil type and at the identity of other conditions, the contents of artificial radionuclide forms varies in a considerable range in the meadow soil: for ^{137}Cs – from 5 up to 790 times, ^{90}Sr - from 1.5 up to 24 times.

As a result of the correlation analysis we managed to reveal a trend of a weak influence of some physical-chemical properties of soil on the relative content of artificial radionuclides forms of ^{137}Cs and ^{90}Sr in the investigated meadow soils.

There have been assessed the analogy between ^{137}Cs and ^{90}Sr behaviour in the soils and their chemical analogs and stable isotopes.

In general, detailed work was carried out to study various radionuclide forms in the soils of two ecosystems (the tunnel watercourses of Degelen massif) with virtually identical physical and chemical soil characteristics and with different types of contamination.

Conclusions based on the results obtained can be used to assess and forecast species of these radionuclides in meadow soils of Degelen massif as a whole, taking into account the variability of soil characteristics and nature of radioactive contamination.

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ДЕГЕЛЕҢ ТАУЛЫ МАССИВІНЕН АҚҚАН СУЛАРДЫҢ ТОПЫРАҚ ЭКОЖҮЙЕСІНЕН РАДИОНУКЛИДТЕРДІ ТАБУ ФОРМАСЫ

А.М. Қабдырақова, А.Е. Құндызбаева, С.Н. Лукашенко

Бұл мақалада бұрыңғы Семей сынақ полигонының "Дегелең" сынақ алаңындағы № 176 және № 177 штольнялардан аққан сулардың экожүйесіндегі жайылымдық топырақтан ^{137}Cs, ^{90}Sr, $^{239+240}$Pu және ^{241}Am техногенді радионуклидтерін табу формасына жүргізілген зерттеулер жайлы деректер келтірілген. Зерттелетін экожүйенің кеңістіктегі шектеуіне қарамастан, топырақтан радионуклидерді табу формасында қомақты түрде өзгеріс (вариациясы) бар екені анықталды. Зерттелетін экожүйенің топырағында ^{137}Cs және $^{239+240}$Pu радионуклидтерін табу формасында тығыз байланысқан форма, ^{90}Sr және ^{241}Am радионуклидтерінде алмасушы және құбылмалы форма сәйкесінше көбірек табылған форма болып табылады. Өзара байланыстылық талдама жүргізу нәтижесінде ^{137}Cs және ^{90}Sr радионуклидтерін табу формасының құрамына зерттелетін топырақтың физико-химиялық қасиетінің әсер ету мүмкіндігі бағаланды.

Түйінді сөздер: топырақ, радионуклидтерді табу формалары, сілтіден жүйелі айыру, топырақтың физика-химиялық қасиеттері, штольня, Дегелең таулы массиві.

УДК 577.4: 577.391:504.53:539.16

ФОРМЫ НАХОЖДЕНИЯ РАДИОНУКЛИДОВ В ПОЧВАХ ЭКОСИСТЕМ ВОДОТОКОВ ГОРНОГО МАССИВА ДЕГЕЛЕН

А.М. Кабдыракова, А.Е. Кундузбаева, С.Н. Лукашенко

В работе приводятся данные исследования форм нахождения техногенных радионуклидов ^{137}Cs, ^{90}Sr, $^{239+240}$Pu и ^{241}Am в луговых почвах экосистем водотоков штолен № 176 и № 177 испытательной площадки "Дегелен" бывшего Семипалатинского испытательного полигона. Выявлено, что, несмотря на пространственное ограничение исследуемых экосистем, существует значительная вариация содержаний форм нахождения радионуклидов в почве. Показано, что преобладающей формой нахождения радионуклидов ^{137}Cs и $^{239+240}$Pu в почвах исследуемых экосистем является прочносвязанная форма, радионуклидов ^{90}Sr и ^{241}Am – обменная и подвижная формы, соответственно. В результате проведенного корреляционного анализа оценено возможное влияние физико-химических свойств исследуемых почв на содержание форм нахождения радионуклидов ^{137}Cs и ^{90}Sr.

Ключевые слова: почва, формы нахождения радионуклидов, последовательное выщелачивание, физико-химические свойства почв, штольня, горный массив Дегелен.

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PECULIARITIES OF ARTIFICIAL RADIONUCLIDES ACCUMULATION IN PLANTS AT THE AREA OF TUNNEL WATERCOURSES OF "DEGELEN" SITE

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The article presents the generalized results of studying the peculiarities of artificial radionuclides accumulation in plants at the area of tunnel watercourses of "Degelen" site (under meadow biogeocenosis conditions). There was revealed the nature of redistribution (by organs) and the dynamics of radionuclide content in aboveground parts of plants, including the main phases of vegetation. The accumulation parameters were determined for radionuclides ^{137}Cs, ^{90}Sr, $^{239+240}$Pu and estimated ^{241}Am transit from the soil into aboveground plant parts. We traced the instability of the accumulation of ^{137}Cs and ^{90}Sr by plants during the 2 growing seasons. The assessment has been made for the influence of some physico-chemical properties of soils, forms of radionuclides occurrence and their stable isotopes and macroelements on radionuclides accumulation by plants. There has been considered the possibility of reducing the accumulation parameters of ^{137}Cs in plants by inserting stable Cs in soil.

Key words: radioecology, radioactive contamination, radionuclides, stable isotopes, forms of radionuclides occurrence, macroelements, soils, physical and chemical properties of soil, plants, phase of vegetation, accumulation coefficients (A_c).

INTRODUCTION

A long-term monitoring of the contaminated areas in Chernobyl and South Ural allowed providing a detailed estimation of radionuclides accumulation parameters, both for cultivated and natural plants of these regions. The main outcomes of that research are represented by the data on radionuclides in-plant accumulation, influenced by a number of factors, such as: physico-chemical properties of radionuclides, agrochemical characteristics of soils and the biological features of plants [1, 2]. Multiple regularities occurred between the radionuclides bioavailability and soil characteristics (humus, soil texture, exchange capacity and other), parameters of radionuclide build up in plants and biological characteristics of plants, as well as many others were experimentally determined.

However, with due consideration of the environment conditions that are typical for this region only, the research study of radionuclides specific accumulation in plants of natural biogeocenoses, is, indisputably, of great importance for the former Semipalatinsk test site (STS) even today. Such research becomes especially vital in view of the large-scale activity on transferring the site lands for economic use. In this connection, a need has intensified for the reliable data on parameters of radionuclides transition from the regional soil to aboveground plant parts, especially, in regard of such poorly studied radionuclides, as $^{239+240}$Pu and ^{241}Am. Data on these radionuclides are extremely rare available, even in the IAEA integrated materials [3].

From the point of view of artificial radionuclides accumulation study, the meadow biogeocenosis plants that are widely spread in the flood-plain areas of contaminated tunnel watercourses of Degelen rock massif are of particular interest. These plants are distinctive in an

extremely high radionuclide-accumulating capacity, which was determined by contents of the most accessible for plants radionuclide species in the vegetable layer, on the one part, and by formation peculiarities of the meadow species root system, on the other part, [4].

The obtained parameters of radionuclides accumulation in plants under study will further be used in computing the doses for the STS area population and also would serve as a basis for the development of both, specific practical recommendations for the solution of a land cover contamination problem in the investigated area and for the development of an action plan to decrease the radionuclide content in products, produced under the radiation pollution conditions.

1. MATERIALS AND METHODS OF RESEARCH

The research was conducted at the former STS territory in the area of tunnels 176 and 177 of "Degelen" testing ground (having the same name as Degelen mountain massif).

1.1. Research methods

Vegetation cover study

The areal vegetation study was performed using specific methods of geo-botanical description, including determination of the main vegetation types, as well as the estimation of projective cover and species composition of plants [5]. Defining of the vegetation cover productivity was accomplished through a cut-sample method at three grounds of 1 м^2 each simultaneously with usage of the description methods (Figure 1, a). For this purpose herbaceous plants were cut at a height of 1-3 cm, the tall grasses – at a 4-6 cm high, while current annual increments only were cut or plucked of subshrubs. To determine the dry weight productivity the cut plants were dried out (sublimated) to an air-dry (fragile) condition and then weighted.

a b c

Figure 1. Determination of vegetation productivity through the cut-sample method (a), meadow herbs (b), shrubs (osier (*Salix triandra*)) in the watercourse flood-plain area (c)

Integral radiation parameters

Measurements of radiation parameters, such as the β-particles flux density and equivalent dose rate (EDR) that are normally used in the selection of research areas with higher levels of radioactive pollution were accomplished in the course of field works, in accordance with the standard procedures [6].

Physical and chemical analysis of soils

Soil texture was determined by the pipette-method, which was exploited to define quantitative compositions in percentage ratio of soil fractions that include one or another

group of various size particles [7]. The Tiurin's technique, modified by Nikitin, was applied to determine the humus content of soils [7]. Measurement of the pH level was accomplished through a method based on the rock-water extract's pH gauging with an electrode system that includes an indicator glass electrode, which potential is determined by the hydrogen ions activity in solution, and a subsidiary flowing comparison electrode with a known potential [8].

Defining of the most accessible for plants radionuclide species, their stable isotopes and microelements

Species of radionuclides and microelements in soils were determined, using a method of successive soil leaching with various chemical agents and defining their content in the obtained extracts [9]. For this purpose we have sequentially treated with various leaching solutions a 100 g air-dry soil sample, sifted through a 1 mm-hole sieve, and obtained correlative extracts as a result. The soil-leach solution ratio was maintained at the level of 1:5 at all experimental stages. The radionuclide forms, which are the most easily accessible by plants, were extracted with water (water-soluble form) and 1M ammonium acetate solution (exchange form).

Radionuclide analysis

We have carried out a number of analyses to measure specific activity of radionuclides in soil and plant samples using the calibrated lab equipment in accordance with the standard technique specifications [10, 11]. The specific activity of ^{137}Cs and ^{241}Am radionuclides was determined with the gamma-ray spectrometer Canberra GX-2020, while the ^{90}Sr specific activity was measured with the beta-ray spectrometer "Progress", the same for $^{239+240}$Pu was determined through the radiochemical separation, followed by measuring with the alpha-ray spectrometer Canberra. The ^{137}Cs radionuclide detection limit equaled to 1 Bq/kg (for plant samples) and 4 Bq/kg (for soil samples), ^{241}Am detection limit – 0.3 Bq/kg and 1 Bq/kg, $^{239+240}$Pu – 0.1 Bq/kg and 1 Bq/kg, accordingly, while the ^{90}Sr detection limit reached 200 Bq/kg. The measurement uncertainty for ^{137}Cs and ^{241}Am radionuclides did not exceed 10-20 %, for ^{90}Sr – 15-25 % and for $^{239+240}$Pu – 30%.

Element analysis

Element analyses for the presence of microelements, such as Na, K, Ca, Mg, and Cs, as well as the Sr stable isotopes in soil samples were performed, applying the inductively coupled plasma mass spectrometry method and using the "ELAN 9000" device together with a computer and specially developed software.

Calculation of accumulation coefficients (A_c***)***

Calculation of accumulation coefficients (A_c), that are required for the quantitative description of parameters of the radionuclides transition from the soil to aboveground plant parts, represent the ratio of radionuclide contents in the unit mass of plants and the soil mass unit [4].

1.2. Subject of research

Soils

Soils under investigation are referred to meadow soils, which are thin (the soil depth in watercourse area is limited to 40-45 cm and in the bank zone to 20-25 cm), fairly humous (black) due to an insignificant period of formation, especially in the core watercourse area, well washed of readily soluble salts and carbonates.

Plants

The main biomass of meadow geocenosis plants is accumulated in the contaminated tunnel watercourse area, where due to extra humidification the full variety of meadow vegetation is represented (willow herb (*Chamaenerium angustifolium*), thistle (*Cirsium arvense*), tansy (*Tanacetum vulgare*), reed grass (*Calamagrostis arundinacea*), nettle (*Urtica dioica*), speedwell (*Veronica spuria*), mint (*Mentha interrupta*), sorrel (*Rumex confertus)*, geranium (*Geranium collinum*), burnet (*Sanguisorba officinalis*), larkspur (*Delphinium dictyocarpum*) and other) (Figure 1, b). The projective vegetation cover occupies 90 % of the flood-plain area, where the productivity makes 3-5 centner/ha. Brushwoods (Figure 1, c), including osier (*Salix triandra*, *S. viminalis*) and brier (*Rosa spinosissima*), Ggrow along the nullas (watercourses), while the reed (*Phragmites australis*) prevail in zones of the maximum wetting.

Vegetation cover of the foothill plain is presented by the steppificated meadow coenoses (wild rye (*Leymus angustus*), alfalfa (*Medicago falcata*), plantain (*Plantago cornuti*), yarrow (*Achillea asiatica*), cinquefoil (*Potentilla bifurca*, *P. dealbata*), dodartia (*Dodartia orientalis*), eryngo (*Eryngium planum*), daisy (*Galatella biflora*), centaury (*Centaurea ruthenica*) and other). The projective cover varies from 50% to 70 %, productivity is 2-3 centner/ha.

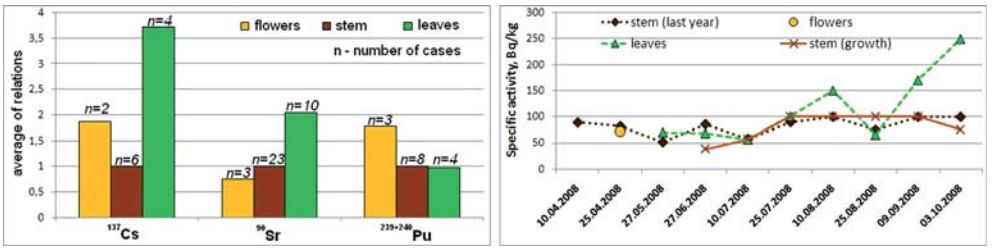
In the context of a radioecological investigation the most interesting objects of "Degelen" site include members (specimens) of the meadow vegetation, associated with higher humidity areas of channel watercourses, which are normally characterized by a higher radioactive contamination, as well as shrubs that are good in terms of instantiation of the deeper subsurface layers owing to a deeply extending root system. Therefore, we have selected some widespread species of meadow motley grasses (tansy (*Tanacetum vulgare*), willow herb (*Chamaenerium angustifolium*), thistle (*Cirsium arvense*), wild rye (*Leymus angustus*)), shrubs –brier (*Rosa spinosissima*) and osier (*Salix triandra*) as test objects for the present research work, because they are characterized by the relatively high biomass, required for a number of experiments.

2. RESULTS AND DISCUSSION

2.1. Nature of redistribution and dynamics of radionuclide contents in aboveground plant parts studied

Three monitoring sites have been established for studying the nature of redistribution and dynamics of radionuclide contents in aboveground parts of plants in the area of surface contaminated watercourses near the tunnel 177. Intermittent samplings (every 15 days) of particular osier (*Salix triandra*) organs (generative (flowers) and vegetative (leaves and stems, including stems (last year) and stems (growth)) were carried out on each site through the whole vegetative season (from April through October). We have determined the specific activity of radionuclides ^{137}Cs, ^{90}Sr, ^{241}Am and $^{239+240}$Pu in all samples.

A column chart (Figure 2, a) indicates the average values of relations between the ^{137}Cs, ^{90}Sr and $^{239+240}$Pu contents in vegetative (stems and leaves) and generative (flowers) organs of the osier (*Salix triandra*), as per the data obtained from the three monitoring sites through the whole research period, normalized to the concentration in footstalks. Specific activity values of ^{241}Am radionuclide in plants proved to be below the detection limit.



a b

Figure 2. Average values of relations between the ^{137}Cs, ^{90}Sr and $^{239+240}$Pu contents in various organs of osier (*Salix triandra*) (a); dynamics values of the ^{90}Sr specific activity in the osier (*Salix triandra*) (b)

The ^{137}Cs and ^{90}Sr radionuclides concentration in generative organs (flowers) is lower than in vegetative ones (leaves and stems), even though the ^{137}Cs content in flowers overcomes its concentration in stems. At the same time, the highest concentrations of both radionuclides were registered in leaves. Significant values of the $^{239+240}$Pu specific activity were registered in flowers in particular (1.4-2.3 Bq/kg), and comparatively lesser amounts were found in vegetative organs (0.6-1.8 Bq/kg).

Due to deficiency of a fair number of ^{137}Cs, ^{241}Am and $^{239+240}$Pu significant activity values, the dynamics of radionuclide contents in some osier (*Salix triandra*) organs through the vegetative season is represented in the chart above (Figure 2, b) only for one monitoring site and solely for the ^{90}Sr radionuclide. A definite trend of the ^{90}Sr concentration volume growth has been observed in leaves and stems of the osier (*Salix triandra*) height increment. However, the ^{90}Sr specific activity values vary in the last year stems, on an average, within the accuracy limits. Generally, the dynamics of ^{90}Sr concentration in the surveyed vegetative organs of osier (*Salix triandra*) have the fairly complex character, which, perhaps, may be explained by both, the growth conditions (in the immediate contaminated watercourse) and physiological peculiarities of the investigated plant.

During the vegetative season we have additionally investigated dynamics of the ^{137}Cs and ^{90}Sr radionuclides accumulation in aboveground parts of individual species of climatic meadow grasses in the area of tunnel # 176, depending on the main phases of vegetation. Therefore, intermittent samplings (in every 15 days) were conducted in the aboveground parts of plants (wild rye (*Leymus angustus*) and tansy (*Tanacetum vulgare*)) at 6 testing grounds. Dynamics of the average values of ^{90}Sr specific activity in each plant species is expressed in a graph below (Figure 3).

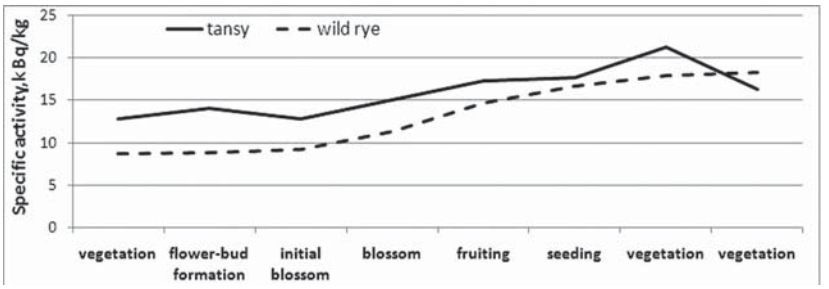
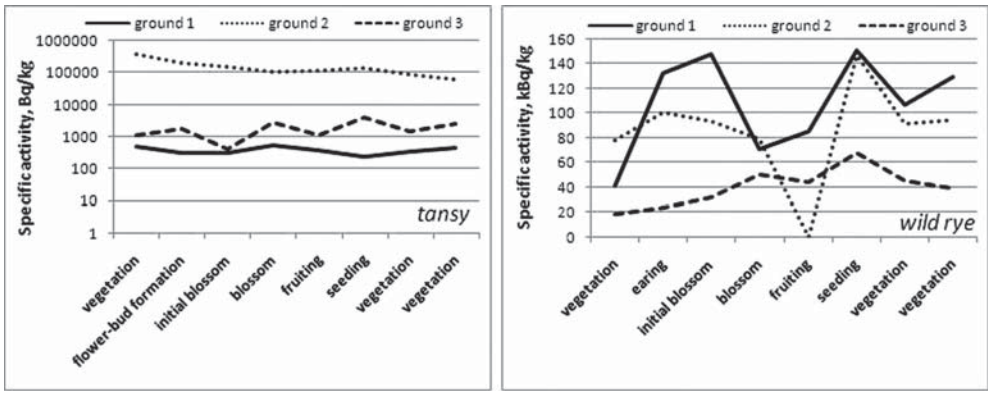


Figure 3. Dynamics of the average values of ^{90}Sr specific activity in the examined plants for various main vegetation phases.

Analyzing the obtained data, we have noted that ^{90}Sr contents in the studied species of plants increase dramatically at the initial blossoming stage and build up through the entire vegetative period. Maximum accumulations fall on autumn months, as follows, in tansy – in September and in wild rye – in September-October.

A completely different situation occurred in regard to the ^{137}Cs accumulation dynamics in the investigated species of plants (Figure 4).



a b

Figure 4. Dynamics of ^{137}Cs concentration in plants (tansy (a) and wild rye (b)), depending on main phases of vegetation

Relying on the findings, we can say that the dynamics of ^{137}Cs content vary in different species of the examined plants, as well as in different test areas. However, by the end of the vegetative season the accumulation of ^{137}Cs in aboveground parts was observed only in wild rye. As a whole, dynamics curves of the ^{137}Cs radionuclide content in the aboveground parts of plants under investigation have far more complex character, comparing to curves reflecting the ^{90}Sr dynamics.

2.2. Determination of quantitative parameters of radionuclides transfer from soils to aboveground parts of plants

For determining the quantitative parameters of radionuclides transfer from soils, characterized by similar physical and chemical properties, to aboveground parts of various plant species, as well as from soils with differential characteristics to specific species of plants we have arranged 5 testing grounds (3 м^2 each) along the tunnel 176 watercourse nulla. Samples of the aboveground plant biomass were taken from individual species of each ground (tansy (*Tanacetum vulgare*), daisy (*Galatella biflora*), inula (*Inula salicina*), fowl-grass (*Poa trivialis*), speedwell (*Veronica spuria*), meadow rue (*Thalictrum simplex*), willow herb (*Chamaenerium angustifolium*), reed (*Phragmites australis*), sedge (*Carex caespitosa*), thistle (*Cirsium arvense*), nettle (*Urtica cannabina*)). A soil sample included 3 specimens that were taken using a method of "shots" to a depth of the most rooting layer (~20 cm). An arithmetic average of the 3 soil specific activity values of each ground has been used to calculate the A_c. Results of the radionuclide analysis are tabulated in Table below (Table 1).

Table 1.

Specific Activity of ^{137}Cs and ^{90}Sr in Plant and Soil Samples

Testing ground #	Species of plants	Radionuclide specific activity, kBq/kg			
		^{137}Cs		^{90}Sr	
		plants	soil	plants	soil
1	tansy (*Tanacetum vulgare*)	34.4±0.2	12.5±0.1	46±5	31±4
	daisy (*Galatella biflora*)	47.0±0.1		19±3	
	inula (*Inula salicina*)	39.0±0.3		77±9	
	fowl-grass (*Poa trivialis*)	20.3±0.2		27±4	
	speedwell (*Veronica spuria*)	22.0±0.1		48±6	
2	tansy (*Tanacetum vulgare*)	73.3±0.3	37±0.3	42±5	21±3
	daisy (*Galatella biflora*)	32.1±0.1		37±5	
	inula (*Inula salicina*)	23.3±0.2		64±8	
	fowl-grass (*Poa trivialis*)	31.0±0.1		19±3	
	speedwell (*Veronica spuria*)	72.4±0.2		32±4	
3	tansy (*Tanacetum vulgare*)	222.0±0.3	30.6±0.3	36±5	20±3
	daisy (*Galatella biflora*)	238.0±0.5		33±4	
	inula (*Inula salicina*)	58.0±0.5		50±6	
	fowl-grass (*Poa trivialis*)	71.0±0.7		13±2	
	speedwell (*Veronica spuria*)	94.0±0.2		44±5	
4	daisy (*Galatella biflora*)	5.0±0.1	4.00±0.04	19±3	16±2
	meadow rue (*Thalictrum simplex*)	0.60±0.03		22±3	
	willow herb (*Chamaenerium angustifolium*)	14.0±0.1		50±6	
	reed (*Phragmites australis*)	25.0±0.2		35±4	
	sedge (*Carex caespitosa*)	1.55±0.03		13±2	
	thistle (*Cirsium arvense*)	7.2±0.1		79±9	
5	reed (*Phragmites australis*)	37.1±0.3	81.1±0.7	34±4	23±3
	sedge (*Carex caespitosa*)	35.0±0.3		22±3	
	thistle (*Cirsium arvense*)	76.2±0.3		90±10	
	nettle (*Urtica cannabina*)	74.0±0.2		126±15	

For carrying out a comparison analysis of the ^{137}Cs and ^{90}Sr accumulation volumes in plants, considering their biological features and physical-chemical properties of soils, we have calculated accumulation coefficients (A_c), as well as coefficients of variations (V) that reflect standard deviations, expressed as the arithmetical average percentage, against the A_c parent population (general totality), for the selected species of plants and different testing grounds (Table 2).

Table 2.

^{137}Cs and ^{90}Sr accumulation coefficients (A_c) and coefficients of variation (V)

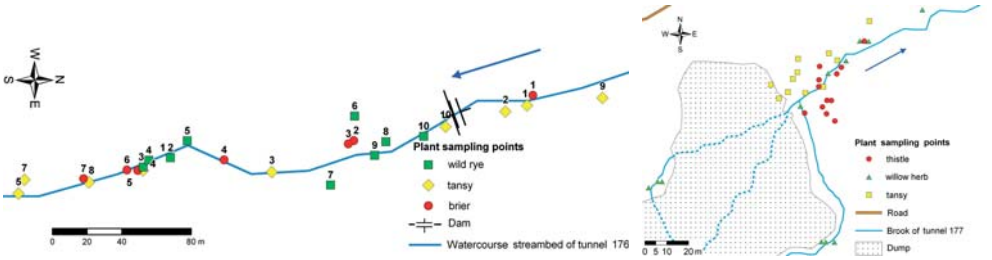
Plant species	A_c of ^{137}Cs					V, %	A_c of ^{90}Sr					V, %
	1*	2	3	4	5		1	2	3	4	5	
tansy (*Tanacetum vulgare*)	2.8	2,0	7.2	-	-	70	1.5	2.0	1.8	-	-	16
daisy (*Galatella biflora*)	3.8	0.9	7.8	1.3	-	92	0.6	1.8	1.7	1.2	-	41
inula (*Inula salicina*)	3.1	0.6	1.9	-	-	66	2.5	3.1	2.5	-	-	13
fowl-grass (*Poa trivialis*)	1.7	0.8	2.3	-	-	46	0.8	0.9	0.6	-	-	17
speedwell (*Veronica spuria*)	1.8	2.0	3.1	-	-	31	1.5	1.5	2.2	-	-	21
meadow rue (*Thalictrum simplex*)	-	-	-	0.2	-	-	-	-	-	1.4	-	-
willow herb (*Chamaenerium angustifolium*)	-	-	-	3.8	-	-	-	-	-	3.1	-	-
reed (*Phragmites australis*)	-	-	-	6.7	0.1	137	-	-	-	2.2	1.5	28
sedge (*Carex caespitosa*)	-	-	-	0.4	0.1	87	-	-	-	0.8	1.0	12
thistle (*Cirsium arvense*)	-	-	-	2.0	0.2	113	-	-	-	5.0	4.0	16
nettle (*Urtica cannabina*)	-	-	-		0.2	-	-	-	-	-	5.6	-
V, %	35	53	63	104	41	-	52	43	40	68	72	-

Note: - the given plant species has not been registered in the area; * # of testing ground

Based on the analysis results we have discovered that the ^{137}Cs and ^{90}Sr accumulation coefficients vary in both, different plant species under the identical physical-chemical soil properties and particular individual species at various testing grounds.

A value range of the ^{137}Cs accumulation coefficient is higher for some of plant species in the raw of testing grounds, than in the raw of various cultures coming from one certain research area. In turn, the A_c values range of ^{90}Sr radionuclide differentiates to a relatively lesser extent, than the ^{137}Cs accumulation coefficient, while a wider variation has been noted in the raw of different plant species. At the same time, accumulation of the ^{90}Sr radionuclide in particular species of plants varies inconsiderably at different research grounds. Therefore, we can assume that the ^{137}Cs radionuclide accumulation in plants is in a greater degree affected by soil characteristics in habitats, while the buildup of ^{90}Sr is influenced by the specific self-features of plants.

To determine statistically reliable A_c values for individual plant species of the meadow biogeocenosis in contaminated areas along the watercourses of tunnels 176 and 177 we have arranged 30 and 36 testing grounds, accordingly, where co-sampling (cross sampling) of soils and aboveground parts of plants was carried out (Figure 5). Tansy (*Tanacetum vulgare*), brier (*Rosa spinosissima*) and wild rye (*Leymus angustus*) in the area of tunnel 176, as well as tansy (*Tanacetum vulgare*), willow herb (*Chamaenerium angustifolium*) and thistle (*Cirsium arvense*) in the region of tunnel 177 were selected as test species for experiments. The area of sampling has not exceeded 1 m^2. Samples of soil were taken using the technique of "shots" to a depth of the most rooting layer down to the rock formation (20 cm).



a b

Figure 5. Layout of testing grounds in the area of tunnels 176 (a) and 177 (b)

Samples taken in the region of tunnel 176, were tested for the specific activity of ^{137}Cs and ^{90}Sr, while the A_c values of radionuclides ^{137}Cs, ^{90}Sr, $^{239+240}$Pu and ^{241}Am were determined in a sample from the tunnel 177 area. Data on radionuclide contents in the tested plant species and soils are represented in Table below (Table 3).

Table 3.

Specific activity ranges of radionuclides ^{137}Cs, ^{90}Sr, $^{239+240}$Pu and ^{241}Am determined in tested plants and soils in vicinity of tunnels 176 and 177

Object of research		Specific activity range, Bq/kg							
		^{137}Cs		^{90}Sr		$^{239+240}$Pu		^{241}Am	
		min	max	min	Max	min	max	min	max
176*	tansy	(0.12±0.02)*10^3	(8.7±0.1)*10^4	(12±2)*10^3	(2.5±0.3)*10^4	-	-	-	-
	brier	(0.03±0.01)*10^3	(1.2±0.1)*10^4	(4.4±0.8)*10^3	(2.7±0.4)*10^4	-	-	-	-
	wild rye	(0.03±0.01)*10^3	(5.4±0.1)*10^4	(3.9±0.8)*10^3	(2.1±0.3)*10^4	-	-	-	-
	Soil	(0.03±0.01)*10^3	(45.2±0.1)*10^4	(0.6±0.2)*10^3	(3.3±0.4)*10^4	-	-	-	-
177*	tansy	-**	101±3	(30±4)*10^3	(85±10)*10^3	2±0.5	51±4	-**	-**
	willow herb	-**	38±4	(7±1)*10^3	(150±20)*10^3	6±1	12±2	-**	2±1
	thistle	-**	540±4	(100±10)*10^3	(320±40)*10^3	40±10	90±10	-**	4±1
	Soil	16±3	11300±100	(1.8±0.4)*10^3	(130±15)*10^3	(2.9±0.04)*10^3	(12.2±0.1)*10^3	-**	680±10
Note: * - tunnel No. ** - below detection limit: ^{137}Cs - <0.1; ^{241}Am - <0.3									

Levels of radioactive contamination in soils and plants of the selected testing areas vary in a relatively broad range. The highest concentrations in the area of tunnel 177 are representative of ^{90}Sr radionuclide, while the ^{137}Cs content in some plants was below the detection limit, and values of the ^{241}Am specific activity in plants were too law as well and in many cases could not be defined. For the moment, the specific activity values of $^{239+240}$Pu radionuclide have been presented only for those research areas, where its high concentration in soils is well known.

Table below (Table 4) represents coefficients of radionuclides ^{137}Cs, ^{90}Sr and $^{239+240}$Pu accumulation in test species of plants. For lack of the specific activity quantitative data, the ^{241}Am accumulation coefficient values are presented barely in the form of an estimate.

Table 4.

Range of radionuclide accumulation coefficient (A_c) values in the vicinity of tunnels 176 and 177

Radionuclide	A_c					
	Tunnel # 176			Tunnel # 177		
	wild rye	brier	tansy	tansy	willow herb	thistle
^{37}Cs	1.30 (n=10) 0.05 - 7.50	2.60 (n=10) 0.01 - 13.20	2.80 (n=10) 0.01 - 16.30	0.02 (n=10) 0.002 – 0.06	0.20 (n=12) 0.001 – 1.00	0.10 (n=11) 0.008 – 0.60
^{90}Sr	3.30 (n=10) 0.90 - 9.50	2.00 (n=10) 0.40 - 4.50	1.80 (n=10) 0.80 - 5.70	0.80 (n=11) 0.50 – 1.40	1.30 (n=13) 0.50 – 4.00	2.47 (n=11) 1.80 – 3.20
$^{239+240}Pu$	-	-	-	0.01 (n=5) 0.0003 – 0.02	0.004 (n=3) 0.001 – 0.003	0.01 (n=2) 0.003 – 0.02
^{241}Am	-	-	-	<0.03 (n=10)	<0.13 (n=12)	<0.09 (n=9)
Note: in the numerator – arithmetic average; in the denominator – range of values, n – number of cases						

Based on the comparative analysis data, we can admit that, in spite of a relatively small area of testing grounds, identical climatological conditions and soil types, as well as the sampling of plants by species, the A_c values range of radionuclides ^{137}Cs and ^{90}Sr in the region of both tunnels is unexpectedly wide. It amounts to one order for ^{90}Sr radionuclide, while for ^{137}Cs it reaches three orders. Estimated coefficients of the ^{241}Am radionuclide accumulation in plants that grow in the area of tunnel 177, on an average, are not higher than 0.1, whereas the A_c of $^{239+240}Pu$ radionuclide ranges from 0.0001 to 0.01.

Comparative distribution of ^{137}Cs and ^{90}Sr coefficients of accumulation (A_c) in the investigated plants of the both tunnels area is demonstrated in the chart of their values frequency expressed in terms of decimal logarithms (Figure 6).

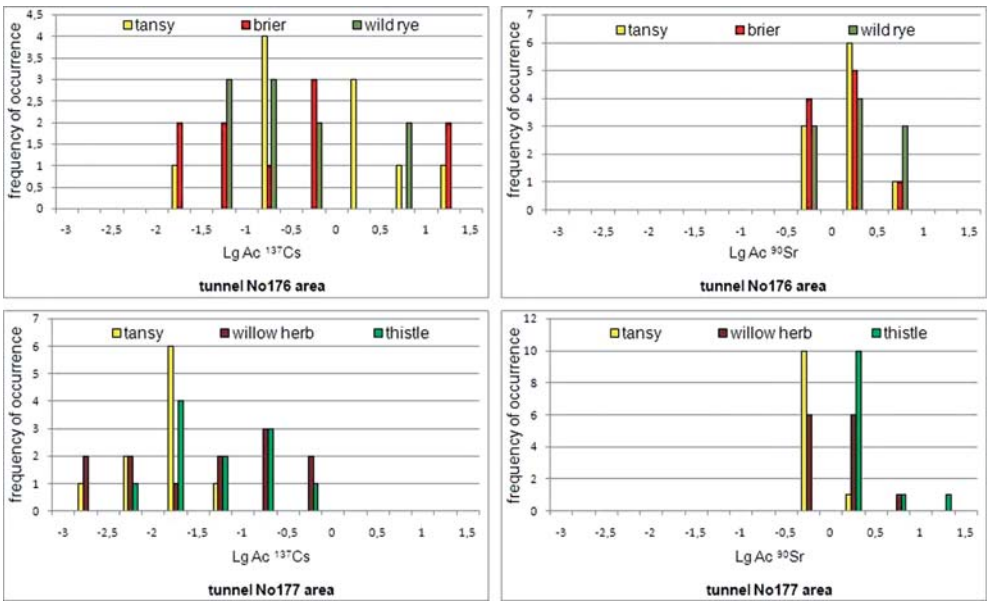


Figure 6. Distribution of ^{137}Cs and ^{90}Sr Lg A_c in the investigated plants in vicinities of tunnels 176 and 177.

The A_c distribution values vary both, by individual species of plants and by different radionuclides, herewith they are more distinct for ^{137}Cs, than for ^{90}Sr. For example, in the region of tunnel 176 the values rang of ^{137}Cs A_c in wild rye is lesser (two orders), than in tansy and brier (three orders each), when in the tunnel 177 area the value rang of ^{137}Cs A_c is lower in tansy (one order), than in thistle (two orders) and willow herb (three orders). In contrast to ^{137}Cs radionuclide, the average values of ^{90}Sr A_c, generally are closure to greater amounts, therefore, ^{90}Sr is the radionuclide that accumulates in plants in a greater degree. The data on A_c tansy in the area of tunnel 177 is a special case, as the average values of ^{90}Sr A_c in this plant do not exceed one unit. It also should be noted that a certain change of ^{137}Cs A_c values to the lesser amounts have been traced in all tested plants in the area of tunnel 177.

We have set 10 additional testing grounds of 1m × 1m each along the watercourse of tunnel 176 in order to enable assessment of the A_c values variability through several growing seasons. Composite soil samples were taken from a depth of the most rooting layer (20 cm) at each ground, and we have been carrying sampling from aboveground parts of plants for 2 years in succession. Test species included wild rye (*Leymus angustus*), cinquefoil (*Potentilla bifurca*), spiraea (*Spiraea hypericifolia*), speedwell (*Veronica spuria*) and sagebrush (*Artemisia latifolia*). Specific activity and accumulation coefficients of radionuclides ^{137}Cs and ^{90}Sr were determined in all samples and tabulated in Table below (Table 5).

Table 5.

Accumulation coefficient (A_c) values for radionuclides ^{137}Cs and ^{90}Sr; ratio of A_c values in 2008 to those in 2009

Ground No.	Plant	A_c				A_c ratio 2008 to 2009	
		^{137}Cs		^{90}Sr			
		2008	2009	2008	2009	^{137}Cs	^{90}Sr
I	wild rye (*Leymus angustus*)	0.09	0.12	1.06	1.13	0.8	0.9
	silverweed (*Potentilla bifurca*)	0.16	0.10	1.81	2.56	1.6	0.7
II	wild rye (*Leymus angustus*)	0.11	0.01	1.00	1.17	11.0	0.9
III	wild rye (*Leymus angustus*)	1.21	0.49	1.97	0.93	2.5	2.1
	silverweed (*Potentilla bifurca*)	0.09	0.14	3.55	4.08	0.6	0.9
	spiraea (*Spiraea hypericifolia*)	0.03	0.07	1.84	2.50	0.4	0.7
IV	wild rye (*Leymus angustus*)	0.32	0.05	1.86	2.22	6.4	0.8
V	wild rye (*Leymus angustus*)	0.15	0.06	4.39	3.23	2.5	1.4
	speedwell (*Veronica spuria*)	0.20	0.11	5.54	6.24	1.8	0.9
VI	wild rye (*Leymus angustus*)	0.13	0.16	1.70	0.89	0.8	1.9
	silverweed (*Potentilla bifurca*)	0.05	0.03	3.10	3.80	1.7	0.8
	sagebrush (*Artemisia latifolia*)	0.69	0.05	2.40	2.00	13.8	1.2
VII	wild rye (*Leymus angustus*)	0.34	0.41	1.60	0.77	0.8	2.1
	silverweed (*Potentilla bifurca*)	0.37	0.11	4.30	3.20	3.4	1.3
	spiraea (*Spiraea hypericifolia*)	0.42	0.16	1.80	1.50	2.6	1.2
	sagebrush (*Artemisia latifolia*)	0.25	0.08	1.20	1.90	3.1	0.6
VIII	wild rye (*Leymus angustus*)	0.83	0.76	1.36	1.36	1.1	1.0
	silverweed (*Potentilla bifurca*)	0.40	0.24	3.73	4.73	1.7	0.8
	spiraea (*Spiraea hypericifolia*)	0.20	0.09	1.27	1.55	2.2	0.8
IX	wild rye (*Leymus angustus*)	0.33	0.07	1.86	0.89	4.7	2.1
	silverweed (*Potentilla bifurca*)	0.18	0.20	4.68	4.65	0.9	1.0

Ground No.	Plant	A_c				A_c ratio 2008 to 2009	
		^{137}Cs		^{90}Sr			
		2008	2009	2008	2009	^{137}Cs	^{90}Sr
X	wild rye (*Leymus angustus*)	1.78	1.56	1.53	1.05	1.1	1.5
	silverweed (*Potentilla bifurca*)	0.47	0.35	4.24	5.06	1.3	0.8
Average ratio for A_c values obtained for the radionuclides in 2008 to those obtained in 2009.						2.9	1.2

The presented data reflect the A_c instability through various seasons not only for various plant species, but also for different testing grounds, where the accumulation of radionuclides ^{137}Cs and ^{90}Sr in some instance increase, while decrease in the others. The represented ratio between the radionuclide A_c values obtained in 2008 and A_c values of 2009 reflects variability of this parameter in regard of the ^{137}Cs and ^{90}Sr accumulation in the same species of plants. For example, the difference in A_c values of ^{137}Cs radionuclide through various seasons in some cases reaches one order, while the ^{90}Sr accumulation values on average change twice as much. The obtained findings speak for some presence of a significant influence of any particular conditions, as well as the climatic characteristics of each season on the seasonal redistribution of ^{137}Cs and ^{90}Sr in the soil and plant waste.

2.3. Assessment of influence of some physical and chemical properties of soils and species on radionuclide accumulation in plants

For lack of the quantitative data on radionuclides $^{239+240}Pu$ and ^{241}Am the influence of some physical and chemical properties of soils and forms of radionuclide species on accumulation of the latter in plants was evaluated solely in regard to ^{137}Cs and ^{90}Sr on the basis of data on 30 and 36 samples of soils and plants, taken in areas of tunnels 176 and 177, accordingly (Figure 5, a, b).

Table below (Table 6) represents the results of a correlation analysis of the ^{137}Cs and ^{90}Sr accumulation coefficients against some physical and chemical properties of soils and the content of radionuclide species there. Correlation factor values of the ^{137}Cs soluble form are not available due to its very low concentration.

Table 6.

Correlation of radionuclide accumulation coefficients (A_c), physical and chemical properties of soils and contents of radionuclide species

Compared items			Soil properties				Radionuclide species (% of total content in soil)		
			humus, %	pH	alphitite, %	salt amount, %	water-soluble	exchangeable	
							^{90}Sr	^{137}Cs	^{90}Sr
A_c ^{137}Cs	tunnel 176	tansy	-0.60	0.65	0.41	0.61	-	0.55	-
		brier	0.24	-0.25	0.76	0.73	-	0.70	-
		wild rye	-0.67	0.20	0.63	-0.21	-	0.38	-
	tunnel 177	tansy	0.23	-0.38	-0.06	-0.13	-	0.57	-
		willow herb	0.60	0.33	0.02	0.22	-	-*	-
		thistle	0.49	0.45	-0.05	-0.01	-	-*	-

Compared items			Soil properties				Radionuclide species (% of total content in soil)		
			humus, %	pH	alphitite, %	salt amount, %	water-soluble	exchangeable	
							^{90}Sr	^{137}Cs	^{90}Sr
A_c ^{90}Sr	tunnel 176	tansy	-0.32	0.79	0.68	0.41	0.70	-	0.06
		brier	-0.20	-0.14	0.79	0.49	0.86	-	-0.37
		wild rye	-0.43	0.09	0.73	0.20	0.65	-	-0.22
	tunnel 177	tansy	-0.04	-0.01	-0.57	-0.09	-0.26		0.74
		willow herb	0.19	0.19	0.35	-0.24	-*	-	-*
		thistle	-0.02	0.17	0.28	0.13	-*	-	-*
Note: * - no data									

Some direct correlation dependence has been observed between the ^{137}Cs accumulation coefficient and exchangeable form of this radionuclide species in soils. Statistically reliable positive factor values of correlation between the ^{90}Sr A_c and its water-soluble form of occurrence in soils nearby the tunnel 176, as well as its exchangeable form in the area of tunnel 177, are typical for all plant species under study.

Considering factors of correlation between the A_c and physical-chemical properties of soils under investigation, we can suggest a potential presence of a significant influence of alphitite (alphitite) on amounts of ^{137}Cs and ^{90}Sr radionuclides accumulation in the test plants near the tunnel 176. In other respects the obtained results are highly contradictive and do not reveal any regularities.

Lack of dependency of the radionuclides accumulation in plants upon physical and chemical properties of soils in this situation can explained by an insufficient range of the given factor variations within one of the biogeocenoses under consideration (in the area of tunnel 176 or 177). Nevertheless, we do not exclude a possibility that the cause and effect relationship between the A_c and some physicochemical properties of soils may be brought to light, while integrating the research data on tansy from the both tunnels area (Figure 7).

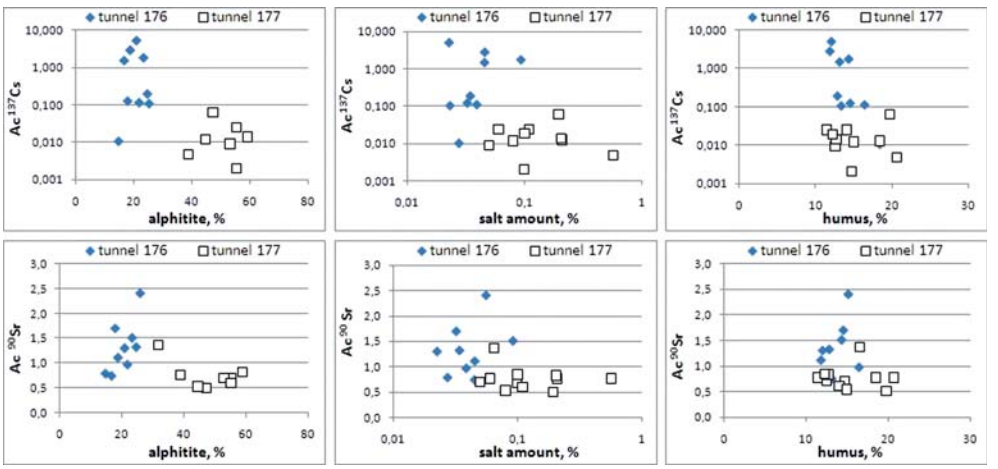


Figure 7. A_c as a function of some physical and chemical properties of meadow soils (for tansy)

The presented charts reflect variations in A_c values and physico-chemical soil properties for areas of the both tunnels. For instance, the soil in the tunnel 176 area is characterized by the lighter soil texture and is better washed of water-soluble salts, compare to the soil in the region of tunnel 177. At the same time, the humus contents in areas of the both tunnels are relatively even, while the coefficients of ^{137}Cs and ^{90}Sr accumulation in tansy are higher in the area of tunnel 176, than near the tunnel 177. In summary, we can assume that a relatively lesser accumulation of radionuclides in plants in the area of tunnel 177 may be aligned with the heavier texture and higher salt content in soils, in particular, in the area of this tunnel.

A similar analysis was conducted in regard of forms of radionuclide species in soils near the tunnels 176 and 177 (Figure 8).

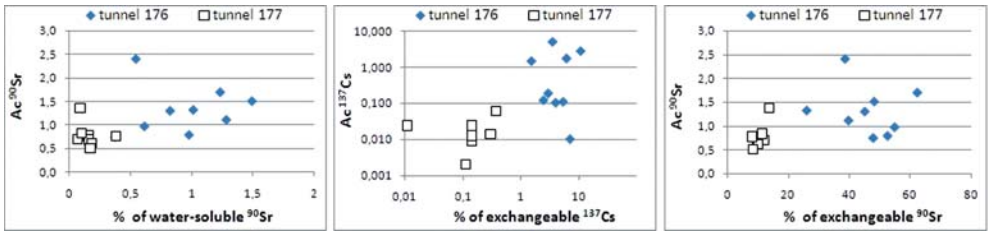


Figure 8. A_c dependence on radionuclide species (for tansy)

The obtained associations also generally confirm a distinct impact from the higher content in soil of radionuclide forms that are most accessible by plants (water-soluble and exchangeable forms) on the level of radionuclides accumulation in plants, in particular, in tansy in the area of tunnel 176.

2.4. Assessment of impact from some microelements and stable isotopes on radionuclide accumulation in plants

Accumulation of radionuclides in plants to a certain degree depends on contents in soil of some macroelements (elements-analogs) and stable isotopes of the studied radionuclides [12]. Possible influence of Na, K, Ca, Mg, Cs and Sr on accumulation of ^{137}Cs and ^{90}Sr radionuclides in the test species of plants have been considered on the ground of data for 30 plant and soil samples, taken in the area of tunnel 176 (in terms of tansy (*Tanacetum vulgare*)) (Figure 5, a). The charts below present the dependence of ^{137}Cs and ^{90}Sr accumulation coefficients upon the Na, K, Ca, Mg and Sr water-soluble form contents in meadow soils (A_c dependence on the Cs water-soluble form does not occur, due to a very low concentration) (Figure 9).

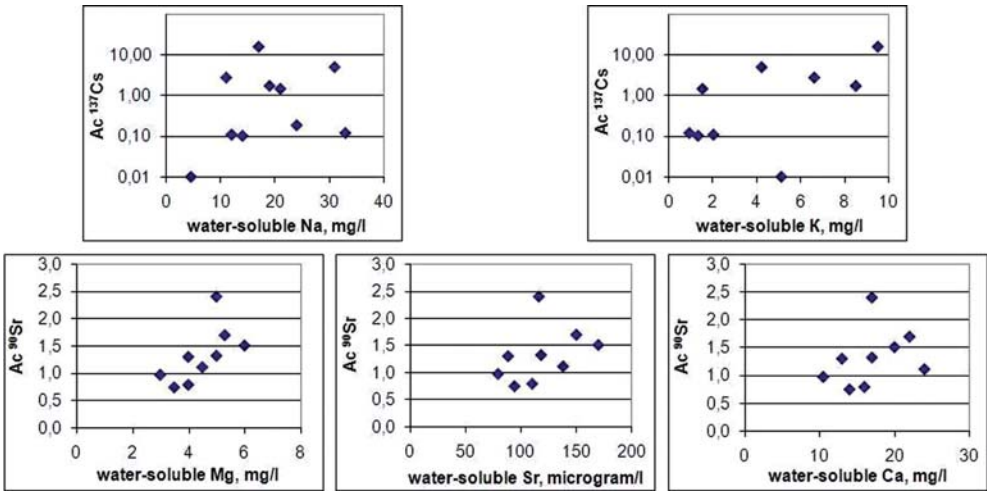


Figure 9. Dependence of the ^{137}Cs and ^{90}Sr accumulation coefficients on contents of some elements' water-soluble forms in meadow soils (in terms of tansy)

Some A_c direct dependency upon the Ca, Mg and Sr water-soluble form contents in the soil has been observed in relation to ^{90}Sr. The following reflects ratios of correlation between the ^{90}Sr accumulation coefficient and contents of some elements in this way: Ca - r = 0.31 ± 0.34 (t = 0.93), Mg – r = 0.64 ± 0.27 (t = 2.36), Sr – r = 0.39 ± 0.33 (t = 1.21). Dependence of the ^{137}Cs A_c on Na and K contents is not available.

The ^{137}Cs and ^{90}Sr accumulation coefficients dependency upon the Na, K, Ca, Mg, Cs and Sr exchangeable form contents in meadow soils is represented in the following charts (Figure 10).

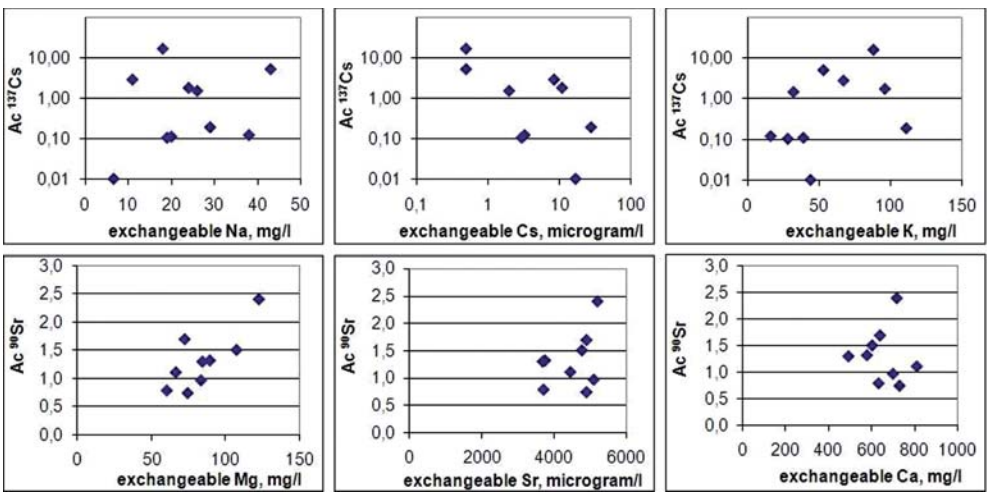
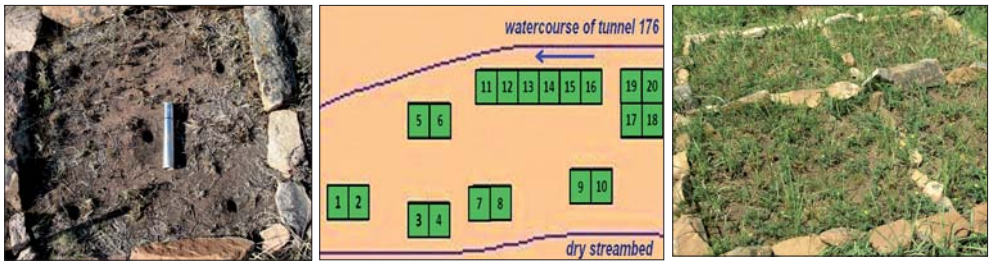


Figure 10. Dependency of the ^{137}Cs and ^{90}Sr A_c on some elements exchange form contents in meadow soils (in terms of tansy)

Diagrams of the ^{90}Sr A_c versus the exchangeable form of the considered elements indicate the same profile as in the results related to Mg and Sr soil-water extracts. Correlation factors of the ^{90}Sr A_c against the Mg contents were as follows: $r = 0.78 \pm 0.22$ ($t = 3.50$), and in case of Sr – $r = 0.38 \pm 0.33$ ($t = 1.15$). At the same time, the inverse relations have been observed between the ^{137}Cs accumulation coefficient and the exchangeable form of stable Cs content in the soil ($r = -0.43 \pm 0.32$ ($t = -1.34$)).

2.5. Analysis of dependence of the ^{137}Cs accumulation in plants on its stable isotope contents in soils

Based on the data for ^{137}Cs A_c dependence on its exchange stable isotope content in the soil (Figure 10), we have carried out a full-scale experiment in the wild. Twenty testing grounds of 1m x 1m each were arranged along the tunnel 176 watercourse in the area that is not bigger than 50m x 20m in size (Figure 11, a, b).



a b c

Figure 11. Testing grounds # 17, 18, 19, 20 (a), testing grounds layout (b), soil sampling (c)

At the end of May 2008 plant waste and aboveground height growth of the spring natural grasses have been removed from each ground. All grounds were divided into experimental (injected with stable cesium) and control ones, where surface irrigation was performed in a certain order as follows: 1) all 20 grounds were watered with 10 liters of clean water, 2) 10 experimental grounds were irrigated with 10 liters of water containing 0.3 g of cesium nitrate ($CsNO_3$), while 10 control grounds were watered with 10 liters of clean water, 3) all 20 grounds were irrigated with 10 liters of clean water. At the end of the vegetation season (late August, in 3 months after the experiment initiation) all testing grounds have been taken down. Cross-sampling of the aboveground parts of dominant plant species (wild rye (*Leymus angustus*), silverweed (*Potentilla bifurca*) (*Potentilla bifurca*), spiraea (*Spiraea hypericifolia*), speedwell (*Veronica spuria*), sagebrush (*Artemisia latifolia*), bedstraw (*Galium boreale*)) was carried out at each ground and a composite soil sample was taken from a depth of the most rooting layer of 20 cm thick, using an envelope technique (Figure 11, c). In one year time we have repeated sampling of the aboveground parts of the mentioned dominant plant species in all research areas with a view to study further eventual transformations.

We have determined the ^{137}Cs radionuclide content and calculated the A_c in all samples of soils and plants. Table below represents A_c values for the predominant species (Table 7).

Table 7.

Values of the accumulation coefficient for ^{137}Cs (A_c) in the test plant species

test ground No.	A_c ^{137}Cs											
	wild rye (*Leymus angustus*)				silverweed (*Potentilla bifurca*)				spiraea (*Spiraea hypericifolia*)			
	2008		2009		2008		2009		2008		2009	
	1*	2**	1	2	1	2	1	2	1	2	1	2
I	0.09	0.25	0.12	0.17	0.16	0.39	0.1	-	-	-	0.02	0.39
II	0.11	0.28	0.01	0.03	-	-	-	0.2	-	-	-	-
III	1.21	24.02	0.49	1.18	0.09	0.28	0.14	0.65	0.03	0.04	0.07	0.1
IV	0.32	0.05	0.05	0.03	-	-	-	-	-	-	-	-
V	0.15	4	0.06	0.85	-	1.43	-	1.13	0.09	0.41	-	-
VI	0.13	0.64	0.16	0.41	0.05	0.08	0.03	0.1	0.12	0.02	-	-
VII	0.34	0.87	0.41	0.12	0.37	0.5	0.11	0.11	0.42	0.19	0.16	0.29
VIII	0.83	1.8	0.76	1.56	0.4	0.26	0.24	0.33	-	-	0.09	-
IX	0.33	0.22	0.07	0.22	0.18	0.17	0.2	0.12	0.03	0.03	-	-
X	1.78	3.32	1.56	2.21	0.47	0.58	0.35	0.81	0.83	0.44	-	1.36
average	0.53	3.55	0.37	0.68	0.25	0.46	0.17	0.43	0.25	0.19	0.09	0.54
Note: - no this plant at this testing ground; * no Cs spiked; ** Cs spiked												

The initial experimental results have not confirmed an advanced assumption (hypothesis) that the ^{137}Cs radionuclide intake by plants may decline after injecting its stable isotope into the soil. Moreover, values of the ^{137}Cs accumulation coefficient at the end of a vegetation season in most of instances appeared to be much higher in areas, where the stable Cs was injected, than in the non-treated areas. This, in turn, might be associated both, with forms of nutrient consumption by plants and their adaptive behaviour in response to a stress factor. Some reduction in A_c values was observed by the end of a year period.

SUMMARY

In accordance with results of studying the peculiarities of radionuclides redistribution and the dynamics of their content in aboveground parts of plants, the major concentrations of ^{137}Cs and ^{90}Sr were found in leaves, while the highest contents of $^{239+240}Pu$ were detected in flowers. The ^{90}Sr radionuclide concentration in plants dramatically increases at the beginning of blossoming period and builds up through the whole vegetative season. Its maximum accumulation falls on autumn months.

We have determined that the accumulation of radionuclides in the investigated plant species vary over a wide range under the meadow biogeocenosis conditions. The range of ^{90}Sr A_c values reaches one order, while the ^{137}Cs range is up to three, where the ^{137}Cs content values variety is more expressed than the ^{90}Sr accumulation amounts differential. Estimated ^{241}Am A_c values, in an average, are not higher than the deciles, and the $^{239+240}Pu$ A_c remains within the limits from 0.0001 to 0.01. We have also traced the instability of ^{137}Cs and ^{90}Sr radionuclides accumulation in the aboveground parts of plants over the two growing seasons.

It was determined that the highest coefficient values of ^{137}Cs and ^{90}Sr accumulation in meadow plants are typical for the soils that have lighter texture and higher content of the most easier accessible for plants forms of radionuclide species (water-soluble and exchangeable ones). Also, we have detected a considerable impact from the Ca, Mg and Sr contents in soil on the ^{90}Sr accumulation coefficient, as well as some inverse relation between the ^{137}Cs A_c and the exchangeable Cs content (in terms of tansy).

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"ДЕГЕЛЕН" АЛАҢЫНЫҢ ШТОЛЬНЯДАН ШЫҚҚАН АҒЫН СУЛАРДЫҢ АУМАҒЫНДАҒЫ ӨСІМДІКТЕРДІҢ ТЕХНОГЕНДІ РАДИОНУКЛИДТЕРДІ ЖИНАҚТАУ ЕРЕКШЕЛІГІ

Н.В. Ларионова, С.Н. Лукашенко, А.М. Қабдырақова, А.Е. Құндызбаева, А.В. Паницкий, Ж.А. Байғазинов

Бұл мақалада "Дегелен" алаңының (жайылымдық биогеоценоз жағдайында) штольнядан шыққан ағын сулардың аумағындағы өсімдіктердің техногенді радионуклидтерді жинақтау ерекшелігін зерттеудің ортақтастырылған нәтижелері ұсынылған. Өсімдіктердің жер бетіндегі бөлігіндегі радионуклидтердің құрамының қарқыны мен таралу сипаты (ағзалар бойынша) анықталды, сонымен қатар вегетацияның негізгі фазалары анықталды. Өсімдіктің жер беткі бөлігіндегі топырағынан ^{241}Am бағалаушы және ^{137}Cs, ^{90}Sr, $^{239+240}$Pu радионуклидтерінің жинақтаушы параметрлері анықталды. Вегетациялық 2 кезең бойында өсімдіктердің ^{137}Cs және ^{90}Sr жинақтауында тұрақсыздығы байқалды. Топырақтың кейбір физико-химиялық қасиеттерінің әсеріне, радионуклидтерді, олардың тұрақты изотоптарының және өсімдіктердің радионуклидтерді жинақтауына макроэлементерді анықтау түріне бағалау жұмыстары жасалды. Тұрақты Cs топыраққа енгізу жолымен ^{137}Cs радионуклидтерінің жинақтау параметрлерін төмендету мүмкіндігі қарастырылды.

Түйін сөздер: радиоэкология, радиоактивті ластану, радионуклидтер, тұрақты изотоптар, радионуклидтерді анықтау түрлері, макроэлементтер, топырақтар, топырақтардың физико-химиялық қасиеттері, өсімдіктер, вегетация фазасы, жинақтау шамасы (Жш).

ОСОБЕННОСТИ НАКОПЛЕНИЯ ТЕХНОГЕННЫХ РАДИОНУКЛИДОВ РАСТЕНИЯМИ В РАЙОНЕ ШТОЛЬНЕВЫХ ВОДОТОКОВ ПЛОЩАДКИ "ДЕГЕЛЕН"

Н.В. Ларионова, С.Н. Лукашенко, А.М. Кабдыракова, А.Е. Кундузбаева, А.В. Паницкий, Ж.А. Байгазинов

В статье представлены обобщенные результаты исследования особенностей накопления техногенных радионуклидов растениями в районе штольневых водотоков площадки "Дегелен" (в условиях лугового биогеоценоза). Выявлен характер перераспределения (по органам) и динамики содержания радионуклидов в надземной части растений, в том числе от основных фаз вегетации. Установлены параметры накопления радионуклидов ^{137}Cs, ^{90}Sr, $^{239+240}$Pu и оценочно ^{241}Am из почвы в надземной части растений. Прослежена нестабильность накопления ^{137}Cs и ^{90}Sr растениями в течение 2-х вегетационных периодов. Дана оценка влияния некоторых физико-химических свойств почв, форм нахождения радионуклидов, их стабильных изотопов и макроэлементов на накопление радионуклидов растениями. Рассмотрена возможность снижения параметров накопления радионуклида ^{137}Cs в растениях путем внесения в почву стабильного Cs.

Ключевые слова: радиоэкология, радиоактивное загрязнение, радионуклиды, стабильные изотопы, формы нахождения радионуклидов, макроэлементы, почвы, физико-химические свойства почв, растения, фазы вегетации, коэффициенты накопления (Кн).

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DEVELOPMENT AND APPLICATION OF THE METHOD FOR ASSESSMENT OF CONTAMINATION WITH TRITIUM IN GROUNDWATER BY ITS CONTENT IN VEGETATION

N.V. Larionova, S.N. Lukashenko, O.N. Lyakhova, S.B. Subbotin, A.O. Aidarkhanov

The article describes the main stages of development and application of a method for assessing the groundwater contamination with ^{3}H by its content in vegetation cover. There has been revealed the dependence of ^{3}H concentration in the plants on its content in groundwater. The most suitable, in terms of accumulative bioindication, plants have been identified. There was assessed the ^{3}H contamination of groundwater that flow beyond Degelen mountain massif by its content in the vegetation cover.

Key words: radioecology, radioactive contamination, radionuclide, tritium, vegetation, plants, groundwater, bioindication.

INTRODUCTION

The groundwater areas contaminated with a radionuclide ^{3}H are one of the Semipalatinsk test site problems. As of today, the ^{3}H concentration in groundwater outside Degelen mountain massif (former "Degelen" testing ground) comes up to tens of kiloBecquerel per kilogram, while in some areas near Shagan River it amounts up to hundreds kiloBecquerel per kilogram. This particular situation may become more complicated due to risk of inclusion of the latter in a unified hydrodynamic system, and thus become a region-scale problem.

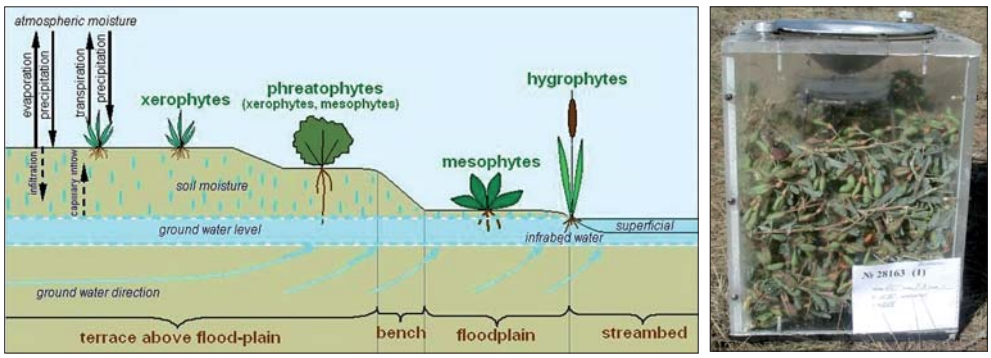
Due to its priority, the above matter requires development of a method for assessing the ^{3}H concentration in groundwater without expensive drilling operations. In the given work, plants have been served as an indicator of ^{3}H concentration in groundwater. A similar method previously applied to forecast a salinity of subsurface water [1], was based on selection of special deep-rooted plants species, which use groundwater as an available source of moisture.

1. MATERIALS AND METHODS

The studies were performed in the area of Shagan River and Degelen mountain massif.

The vegetation cover was investigated by using some methods of geobotanical description [2] by means of identification of major vegetation types and definition of species composition of the plants; choice of species of plants to be investigated was based on their *classification* in *respect* to moisture. Accordingly, based on the above characteristics of the vegetation cover, among hygrophytes having the highest demand for water and thus, often growing near the surface watercourses, reed (*Phragmites australis*) was chosen to be a target of research. To mesophytes that are settled for a moderate moisture and grown in the areas where groundwater occurs near the surface, the following plants may be referred: wild rye (*Leymus angustus*), sorrel (*Rumex confertus*), and also samples of meadow herbs. The following dry steppe plants (xerophytes), being adapted to minimal evapourative water loss

and reflection of light, and also having a shallow but extended root system, were sampled: wormwood (*Artemisia terrae-albae*), spiraea (*Spiraea hypericifolia*) and alfalfa (*Medicago falcata*). To special group of plants – phreatophytes, being the deep-rooted plants reaching the groundwater level, and growing along the watercourses on the areas of relatively shallow occurrence, were referred the following: cottonwood (*Populus nigra*), osier (*Salix triandra*), cheegrass (*Achnaterum splendens*), salt tree (*Halimodendron halodendron*), licorice (*Glycyrrhiza uralensis*) (Figure 1, a). Investigated samples of herbaceous plants were presented by their aerial part, while shrubs and trees – by their current annual increment.



a b

Figure 1. Objects of research (a), unit for evapourating free water from the plants (b)

The ^{3}H content was determined in a plant free water, derived from the condensate by using special unit equipped with a sealed container for plants samples to be loaded in, having a cooled surface and a container to collect moisture evapourated from the plants (Figure 1, b).

To conduct a benchmarking study of the ^{3}H concentration level in plants being investigated and its groundwater content, the water wells were drilled in some research sites, from which groundwater samples were drawn [3].

Measurements of ^{3}H radionuclide specific activity in the prepared samples were made with the help of liquid scintillation spectrometry using TRI-CARB 2900 TR (LS-spectrometer) [4].

1. RESULTS AND DISCUSSION

2.1. Development of methods for ^{3}H identification in groundwater, based on the ^{3}H concentration level in plants using the principle of accumulative bioindication

To reveal the relationship between ^{3}H concentration in plants and in groundwater contaminated with ^{3}H a section line of five research sites along the watercourse for drawing samples of plants and groundwater from the drilled wells (Figure 2) was established at one of the expected sites (length ~ 60 m) with high ^{3}H contents groundwater discharge area on Shagan River, chosen based on findings concerning ^{3}H concentration level in infrabed water in the territory of the "old" river bed, located a shade higher the mainstream.

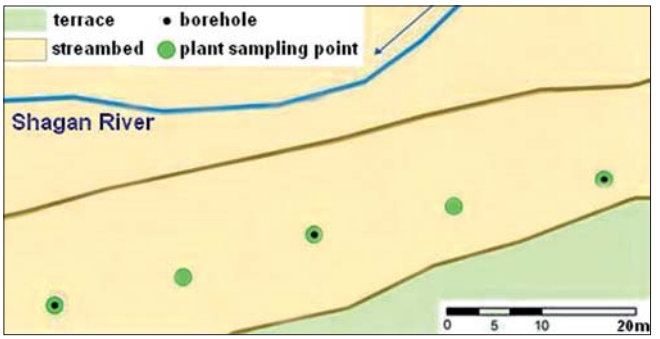
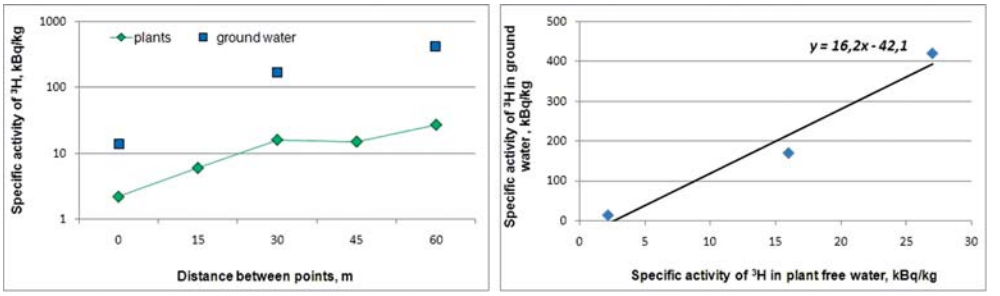


Figure 2. Allocation of sampling points

Samples of aerial parts of the plant under study: wild rye (*Leymus angustus*), were drawn at each research site. Along with that, the groundwater samples were drawn from 3 points of the drilled wells. The results of the ^{3}H specific activity in the vegetation cover and groundwater are presented in the diagrams (Figure 3, a, b).



a b

Figure 3. ^{3}H specific activity in the plants and groundwater (a), dependence of ^{3}H concentration in the plants on its concentration in the groundwater samples (b).

It has been revealed that within the study area a range of the ^{3}H specific activity values in plants is equal to 1 order of magnitude, and in groundwater – 2. However, even with this difference in values, a symbate change of the ^{3}H concentration level in plants with its concentration in groundwater has been observed. Consequently, the ^{3}H concentration in plants may be considered a kind of indicator of the ^{3}H concentration level in groundwater.

2.2. Revealing the dependence of ^{3}H concentration in plants on its concentration in groundwater

The large-scale studies on revealing the dependence of ^{3}H concentration in groundwater on its concentration in plants were conducted outside Degelen mountain massif in the Baitles river valley. The research sites to take samples of plants and groundwater (if any) from the drilled wells were established along the mainstream. The following phreatophytes: cheegrass (*Achnaterum splendens*) and salt tree (*Halimodendron halodendron*) which grow on the areas of relatively shallow groundwater were the major plant species sampled for investigation. Additionally, in some points, due to a lack of these particular species, samples of steppe

shrubs spiraea (*Spiraea hypericifolia*) and goat's-wheat (*Atraphaxis frutescens*) were drawn; and on a site of outcrop of water mixed meadow grasses samples were drawn.

Findings presented in the figure (Figure 4) shows that within the territory under investigation the increased ^{3}H concentration level in plants registered in some sites (up to tens of kBq/kg) often exceeds the ^{3}H concentration in groundwater.

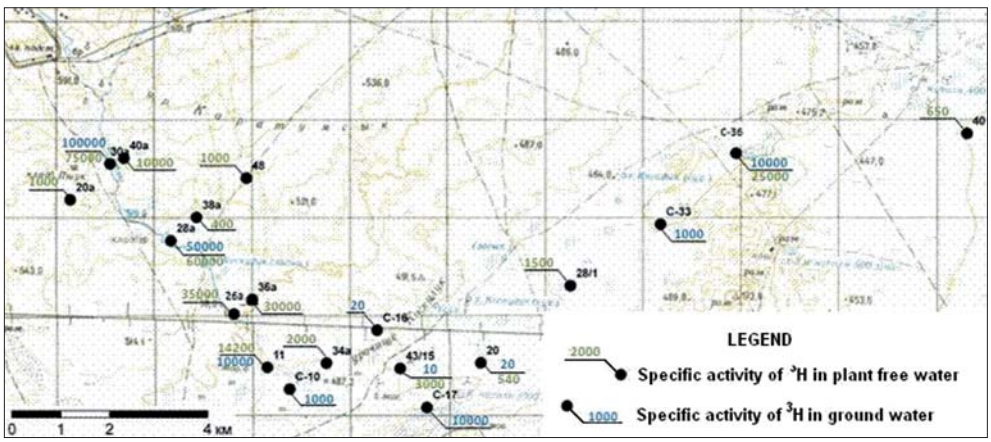
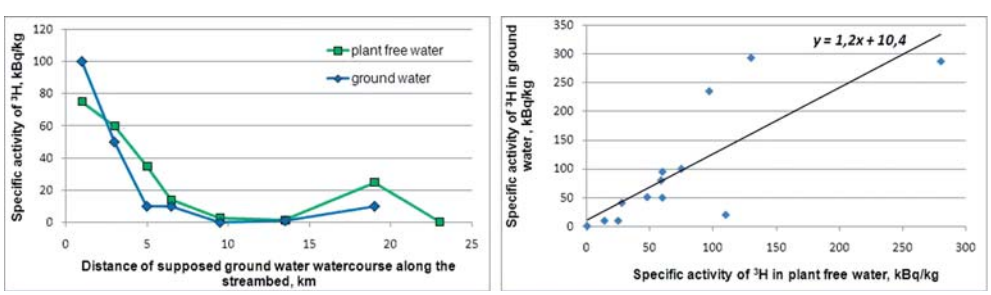


Figure 4. Map of the ^{3}H areal extent in plants and groundwater in the Baitles river valley

However, the increased ^{3}H specific activity in free water of plants, in general, coincides with zones of increased ^{3}H specific activity in groundwater. Maximum ^{3}H concentrations were seen along the Baitles river bed with their gradual decrease as its flowing away from Degelen mountain massif (Figure 5, a). Some increase in the ^{3}H concentration in plants observed in point 36, which can occur as a result of additional source of ^{3}H in groundwater and, consequently, in plants within the concerned area, and also because of the Baitles groundwater flow pattern.



a b

Figure 5. Distribution of the ^{3}H specific activity in plants and groundwater in the Baitles river bed as its flowing away from Degelen mountain massif; the dependence of the ^{3}H concentration in free water of plants on its concentration in groundwater (b)

We were able to reveal the dependence of the ^{3}H concentration in plants on its content in groundwater (Figure 5, b) based on our studies conducted within the Baitles river area, and

also through integrating additional data of the contiguous sampling of plants and groundwater within the area of Degelen mountain massif (zone of side drift #177 [5]) and outside.

2.3 Choice of the most significant species of plants in terms of their accumulative bioindication

To reveal the most significant in terms of their accumulative bioindication species of plants in areas of known high ^{3}H concentration in groundwater (along the main rivulets beds within Degelen mountain massif), 15 research sites were laid down. At each site, the contiguous sampling of aerial parts of the following plants: cheegrass (*Achnaterum splendens*) and), wild rye (*Leymus angustus*), spiraea (*Spiraea hypericifolia*), salt tree (*Halimodendron halodendron*), reed (*Phragmites australis*), cottonwood (*Populus nigra*), licorice (*Glycyrrhiza uralensis*), osier (*Salix triandra*), wormwood (*Artemisia terrae-albae*), alfalfa (*Medicago falcata*), sorrel (*Rumex confertus*), sedge (*Carex melanostachya*) has been made. To conduct a benchmarking study, the ratio of ^{3}H concentration in the studied plants *normalized for its maximal concentration* for each research site was calculated based on data of ^{3}H specific activity in free water in the plants (Table 1).

Table 1.

The ratio of ^{3}H concentration in free water of the plants *maximum normalized* **for each research site**

Research sites	The ratio of ^{3}H concentration in free water of the studied plants											
	spiraea	alfalfa	sorrel	worm wood	sedge	wild rye	salt tree	licorice	chee-grass	reed	osier	cotton-wood
	*n=1	n=1	n=1	n=2	n=3	n=3	n=4	n=4	n=8	n=3	n=5	n=1
1	-	-	-	-	-	1.00	-	-	0.96	-	-	-
2	0.08	-	-	-	-	-	1.00	-	-	-	-	-
3	-	-	-	-	-	-	-	-	0.86	1.00	-	-
4	-	-	-	-	-	-	0.40	-	-	-	-	1.00
5	-	-	-	-	-	-	-	1.00	0.98	-	-	-
6	-	-	-	-	-	-	-	0.88	-	-	1.00	-
7	-	-	-	1.00	-	0.59	-	-	0.91	-	-	-
8	-	-	-	-	-	-	0.87	-	1.00	-	-	-
9	-	-	-	-	0.67	-	-	-	-	-	1.00	-
10	-	-	-	-	0.78	-	-	-	-	0.96	1.00	-
11	-	0.28	-	-	-	0.60	-	0.46	1.00	-	-	-
12	-	-	0.53	-	0.49	-	-	-	-	-	1.00	-
13	-	-	-	-	-	-	-	1.00	0.84	-	-	-
14	-	-	-	0.10	-	-	1.00	-	0.30	-	-	-
15	-	-	-	-	-	-	-	-	-	0.92	1.00	-
average ratio values	0.08	0.28	0.53	0.55	0.65	0.73	0.82	0.84	0.86	0.96	1.00	1.00

- no such species of plants was found in the site
* – number of cases

The resulted average ratio values enable to distribute the studied plant species by their ability to accumulate the radionuclide ^{3}H (Figure 6).

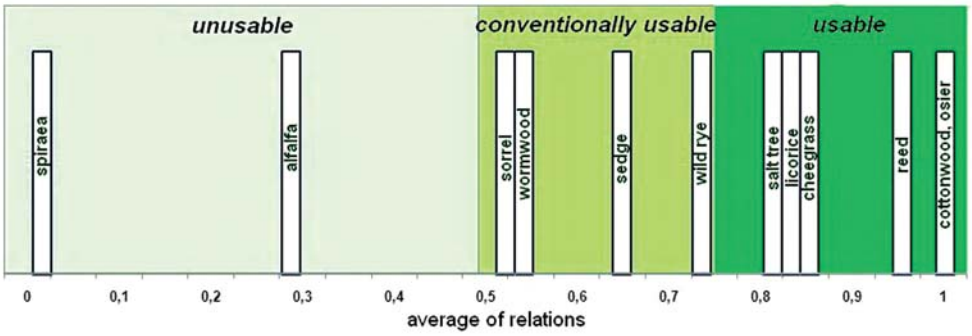


Figure 6. Average ^{3}H concentration ratio in various species of the plants

Relying on the figure above, all the examined species of the plants may be divided into three tentative groups by their suitability for bioindication of groundwater contaminated with radionuclide ^{3}H: reed (*Phragmites australis*) (hydrophyte, growing directly in the water), cottonwood (*Populus nigra*), osier (*Salix triandra*), cheegrass (*Achnaterum splendens*), licorice (*Glycyrrhiza uralensis*), salt tree (*Halimodendron halodendron*) (phreatophytes) –usable; wild rye (*Leymus angustus*), sedge (*Carex melanostachya*), sorrel (*Rumex confertus*), wormwood (*Artemisia terrae-albae*) (plants, often growing in the areas of high humidity)– conventionally usable; spiraea (*Spiraea hypericifolia*) and alfalfa (*Medicago falcata*) – unusable.

2.4. Assessment of groundwater contamination with ^{3}H by its concentration in vegetation

To assess the groundwater contamination with ^{3}H by its concentration in the vegetation cover along the dried beds of the following rivulets: Karabulak, Uzynbulak, Baitles, Toktakushik, Aktybay and one unnamed small river (further - Bezymyanniy), that is areas of intended discharge of groundwater contaminated with ^{3}H radionuclides outside Degelen mountain massif, over a distance on an average of 500 m - 2 km, depending on the length of each, the research sites for drawing samples of certain species of plants were established.

Results of analysis of the ^{3}H specific activity in free water of plants sampled along the beds of rivulets: Karabulak, Uzynbulak, Baitles, Toktakushik, Aktybay and Bezymyanniy one are given in the table (Table 2).

Table 2.

^{3}H concentration in free water of the plants

Name of rivulets	Range of values of ^{3}H specific activity in free water of the plants, kBq/kg		Number of samples	Relative length of the bed (up to end-zone flow), km
	Min	max		
Uzynbulak	1.30 ± 0.13	60 ± 6	n = 24	10.2
Karabulak	0.35 ±0.03	60 ± 6	n = 25	16

Name of rivulets	Range of values of ^{3}H specific activity in free water of the plants, kBq/kg		Number of samples	Relative length of the bed (up to end-zone flow), km
	Min	max		
Toktakushik	2.30 ±0.23	85 ± 8.5	n = 12	5.5
Baitles	0.09 ±0.01	60 ± 6	n = 14	19.7
Aktybay	0.40 ± 0.04	51 ± 5.1	n = 7	6
Bezymyanniy	0.15 ±0.02	7.8 ±0.8	n = 7	5

As one can see from the specific activity ranges, the ^{3}H concentration in free water of the plants sampled in all beds of small rivers averages tens of kBq/kg. The highest ^{3}H concentration of 85 kBq/kg was registered in the Toktakushik river bed. The lowest concentrations registered in the area of Uzynbulak river and Toktakushik were more than thousands of Bq/kg; Karabulak, Aktybay and Bezymyanniy rivers beds its value was hundreds of Bq/kg, and Baitles river – 90 Bq/kg.

The diagram below (Figure 7) shows ^{3}H concentration level in free water of plants sampled along the rivulet beds from the border and further as moving away from Degelen mountain massif, maximum normalized for each rivulet.

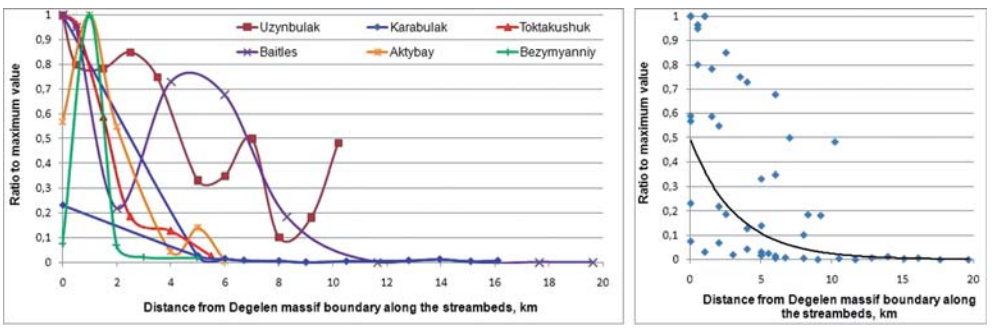


Figure 7. Distribution of ^{3}H in free water of the plants sampled along the main rivulet beds

The highest ^{3}H concentrations are geographically confined to the border of Degelen mountain massif and, at large, they become lower as moving away from it. A slight increase in the ^{3}H concentration was seen in Uzynbulak river bed at a distance of 9-10 km from the border of Degelen mountain massif; and the lowest values of ^{3}H specific activity in free water of the plants presented in the table (Table 2), indicate its high concentration in the end-zone flow of this rivulet. A complicated structure of the curves showing each rivulet can be associated with both the hydrological features of the area under study and the choice of one or another plant in any single case, because accumulation ability of ^{3}H for various plant species varies in a rather wide range**.**

The diagram (Figure 8) shows the ^{3}H concentration distribution in free water of the plants sampled in the sections laid athwart the beds of Karabulak and Uzynbulak rivers, *maximum normalized* for each line section.

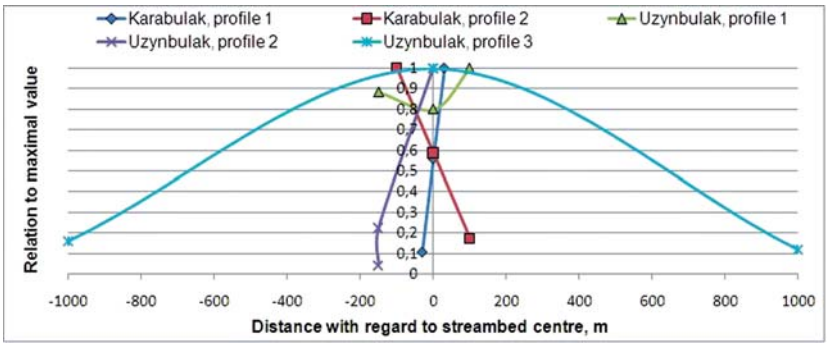


Figure 8. ^{3}H concentration distribution in free water of the plants sampled in the line sections laid athwart the rivulets beds

In some specified sense, ambiguous character of the resulted curves confirms the hypothesis about the impact of choice of any given species of the plants on distribution of the ^{3}H concentration level in free water of the plants, sampled in the line sections laid perpendicular to the bed.

The given data and the linear dependence presented above (Figure 5, b) enable to complete an indirect assessment of the radionuclide ^{3}H distribution in groundwater sampled within the studied area. The figure helps to forecast a character of groundwater contaminated with ^{3}H by its concentration in free water of the plants, and provides experimental values of ^{3}H specific activity identified in groundwater sampled from the drilled wells (Figure 9).

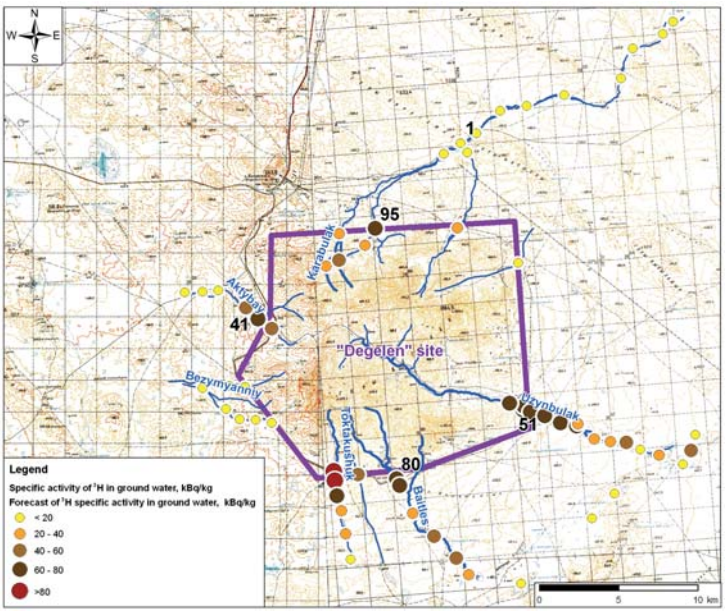


Figure 9. Forecasting the character of groundwater contamination with ^{3}H by its concentration in free water of the plants sampled in the study area

According to data on ^{3}H distribution in free water of plants sampled in the study area, the Toktakushik river bed is the most expectable maximum ^{3}H contaminated groundwater area. In general, the estimated ^{3}H specific activity values found in groundwater do not contradict the experimentally obtained ones. The ^{3}H radionuclide concentration in groundwater amounts to tens of kBq/kg, however, the intervention level when this radionuclide arrives together with water in accordance with Radiation Safety Norms (NRB-99) is 7.7 kBq/kg. From the quantitative point of view, the recommended forecast can be considered relatively objective if ^{3}H concentration level in groundwater would be no less than 20 kBq/kg.

CONCLUSIONS

In the present research we revealed the dependence of the ^{3}H concentration in plants on its concentration in groundwater. From the standpoint of their accumulative bioindication the following plants: reed (*Phragmites australis*), being a hygrophyte, and also phreatophytes - cottonwood (*Populus nigra*), osier (*Salix triandra*), cheegrass (*Achnaterum splendens*), licorice (*Glycyrrhiza uralensis*), salt tree (*Halimodendron halodendron*) were considered the most suitable ones. An assessment of ^{3}H contamination of groundwater flowing away from Degelen mountain massif was made based on its concentration level in the vegetation cover.

Taken as a whole, one can say that plants as indicators of ^{3}H concentration in groundwater can be used as a cost-effective and technically simple practical method for both prospective assessment of ^{3}H concentration in groundwater of potentially hazardous sites and monitoring the sanitary protection zones of the nuclear fuel cycle facilities.

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ӨСІМДІК ЖАМЫЛҒЫСЫНДАҒЫ ҚҰРАМЫ БОЙЫНША ТОПЫРАҚ АСТЫ СУЛАРЫНЫҢ ТРИТИЙМЕН ЛАСТАНУЫН БАҒАЛАУ ӘДІСІН ӘЗІРЛЕУ ЖӘНЕ ПАЙДАЛАНУ

Н.В. Ларионова, С.Н. Лукашенко, О.Н. Ляхова, С.Б. Субботин, А.О. Айдарханов

Бұл мақалада өсімдік жамылғысындағы құрамы бойынша топырақ асты суларының ^{3}H ластануын бағалау әдісін әзірлеу және қолданудың негізгі кезеңдері қарастырылды. Топырақ асты суларындағы құрамының өсімдіктерде ^{3}H шоғырлануына тәуелділігі белгіленді. Өсімдіктердің жинақталу биоиндикациясы тұрғысынан қарағандағы жарамды дегендері анықталды. Дегелең таулы массивінен тыс жерлерге ^{3}H радионуклидімен ластанған топырақ асты суларының өсімдік жамылғысындағы құрамы бойынша (бұрынғы "Дегелең" сынақ алаңы) ластануын бағалау жұмыстары орындалды.

Түйін сөздер: радиоэкология, радиоактивті ластану, радионуклид, тритий, өсімдік жамылғысы, өсімдіктер, топырақ асты сулары, биоиндикация.

РАЗРАБОТКА И ПРИМЕНЕНИЕ МЕТОДА ОЦЕНКИ ЗАГРЯЗНЕНИЯ ТРИТИЕМ ГРУНТОВЫХ ВОД ПО ЕГО СОДЕРЖАНИЮ В РАСТИТЕЛЬНОМ ПОКРОВЕ

Н.В. Ларионова, С.Н. Лукашенко, О.Н. Ляхова, С.Б. Субботин, А.О. Айдарханов

В статье рассмотрены основные этапы разработки и применения метода оценки загрязнения ^{3}H грунтовых вод по его содержанию в растительном покрове. Выявлена зависимость концентрации ^{3}H в растениях от его содержания в грунтовых водах. Определены наиболее пригодные, с точки зрения аккумулятивной биоиндикации, растения. Выполнена оценка загрязнения ^{3}H грунтовых вод, выходящих за пределы горного массива Дегелен (бывшей испытательной площадки "Дегелен"), по его содержанию в растительном покрове.

Ключевые слова: радиоэкология, радиоактивное загрязнение, радионуклид, тритий, растительный покров, растения, грунтовые воды, биоиндикация.

УДК 504.4.054:577.4: 546.11.02.3

MECHANISMS FOR FORMATION OF AIR BASIN CONTAMINATION WITH TRITIUM WITHIN THE "DEGELEN" MASSIF

O.N. Lyakhova, S.N. Lukashenko, N.V. Larionova

The issue of tritium contamination occupies a special place in the radioactive contamination of the environment. In this paper we assess the level and distribution of tritium in the atmospheric air of ecosystems with water seepage at tunnels № 176 and № 177, located on "Degelen" site. There has been presented general nature of tritium distribution in the atmosphere relative to surface of a watercourse which has been contaminated with tritium. The basic mechanisms were studied for tritium distribution in the air of studied ecosystems, namely, the distribution of tritium in the systems: water – atmosphere, tunnel air - atmosphere, soil water - atmosphere, vegetation - atmosphere. An analytical calculation of tritium concentration in the atmosphere by the concentration of tritium in water has been performed. There has experimentally obtained the dependence for predictive assessment of tritium concentrations in air as a function of tritium concentration in one of the inlet sources such as water, tunnel air, soil water, vegetation. The paper also describes the general nature of tritium distribution in the air in the area "Degelen".

Key words: Degelen site, tritium, atmospheric air, tritium contamination, tritium distribution.

INTRODUCTION

A special approach to the study of levels and nature of tritium distribution in the environment objects is required taking into consideration the specific nature of tritium contamination of Degelen site located at the Semipalatinsk test site (STS). The nature of tritium contamination of the atmospheric air is of significance.

Tritium in nature. In nature, tritium is formed due to cosmic radiation in the uppermost levels of atmosphere at the rate of 1,200 atoms per second per 1 m^2 of earth surface. The sources of tritium synthesis in the atmosphere are nuclear reactions induced by cosmic radiation: fast cosmic neutrons split nitrogen to two fragments, ^{12}C and tritium, and high-energy protons and mesons can generate tritium in reactions with any nuclei in the atmosphere [1]:

$$^{14}N+n \rightarrow {}^{12}C+T \quad (1)$$

Tritium generated this way undergoes to water molecules via oxidation and isotope exchange reactions, and then tritium as a component of rain water falls off on the ground. According to the current assessments, the equilibrium activity of cosmogenic tritium in the environment (hydrosphere and atmosphere) is $(1.11\text{-}1.30)\cdot 10^9$ GBq (3.0-3.5 kg). About 90% of natural tritium is present in the hydrosphere (mainly as HTO), 10% in the stratosphere and 0.1% in the troposphere [2]. For thousands of years, the concentration of tritium in nature has been almost constant: its continuous generation in the atmosphere has been indemnified by the natural decay.

Background tritium concentration in water is H:T = $1:10^{18}$. Such concentration of tritium atoms has a special name, "tritium unit" (TU). One liter of water contains on the average $1.1\cdot 10^{-1}$ Bq of tritium and 1 liter of air - $5.7\cdot 10^{-4}$ Bq [3].

Artificial tritium. Since 1945 (the beginning of nuclear tests), the situation has drastically changed and the content of tritium in the rain water has increased for thousand times.

The H-bomb explosion of 1 megaton (Mt) yield causes 0.7 to 2 kg release of tritium. The total yield of air explosions in the 1945-1962 period was 406 Mt and that of surface explosions - 104 Mt. At the same time, the total amount of tritium released to the biosphere due to the tests was hundreds kilograms.

Thus, the formation of artificial tritium is due to mainly nuclear tests and the activities related to the nuclear fuel cycle.

During an explosion, the synthesis reaction produces ~$7*10^{14}$ Bq/kt and the fission reaction ~$4*10^{10}$ Bq/kt. In case of underground explosion these figures increase due to the nuclear reactions on boron and lithium existing in hard rocks [4]:

$$^{10}B_5 + n \rightarrow 2^4He_2 + T \quad (2)$$

$$^{6}Li_3 + n \rightarrow {}^{4}He_2 + T \quad (3)$$

Most likely, artificial tritium generated at explosions can exist as three basic compounds: tritiated (HTO), tritiated hydrogen (HT) and tritiated methane. Tritium can be present in all natural environments: atmospheric air, water and vegetation [3].

"Degelen" site. During the tests in tunnels at Degelen massif, 209 nuclear underground explosions were made in 180 tunnels resulting in the formation of considerable amount of tritium. The yield of nuclear explosions in tunnels in the surface and groundwater of Degelen massif ranged from 0.001 to 125 kg. According to the earlier test data, the tritium concentration values in the surface and groundwater of Degelen massif are of the order of n*10^5 Bq/kg [5]. The obtained values exceed considerably the tritium concentration in the surface water due to the natural content in nature.

Since geologically Degelen massif is a big granite batholite (granite rocks are spread there for 75-80%), one of the ways for tritium generation inside the tunnels at Degelen site is, possibly, the above-mentioned reactions (2, 3) during underground explosions. Besides the main elements this rock comprises such microelements as Sn, W, Mo, Li, Be, B, Rb, Bi, Ta and Au. The content of Li in hard granite rocks varies from 1 to 30 mg/kg and that of B is much lower, being about 0.001 mg/kg [6, 7]. Tritium formed by the reactions on lithium and boron, undoubtedly, contributes to the total amount of tritium generated at Degelen site; however, assessment of this contribution would be rather difficult. Based on the assumption that the tritium concentration in tunnel watercourses remains constant in time, it can be suggested that inside the tunnels tritium accumulated during nuclear explosions is subjected to gradual outwashing (leaching out) off the crystalline structure of the melt surface of inner walls.

It is also possible that during nuclear tests inside tunnels, gaseous tritium as tritiated hydrogen (HT) is also formed. Gaseous tritium released to the atmosphere is less toxic and mobile than HTO or T_2O and in a quite a short period of time transforms to tritium oxide (HTO) actively substituting stable hydrogen in the water molecules of water vapour [8].

Tritium formed inside the tunnels is washed out by rainfalls and groundwater coming out to the day surface as tritiated water (HTO).

In a quite a short period of time the whole tritium formed, in one way or another, transforms to tritium oxide (HTO) or, more simply, to tritiated water. Hence, it would be

more reasonable to study the level and nature of tritium distribution by investigating the tritium presence in the objects of ecological interest such as water, air, vegetation, etc., studying tritiated water contained in these objects.

The high mobility of tritium in the environment and the data on tritium concentration in the water bodies of Degelen site have been a ground for conducting a detailed study to assess the level of tritium contamination of the air basin of Degelen massif and mechanisms of its formation.

1. METHODS AND OBJECTS OF STUDY

The main objective of the study was the assessment of level and nature of tritium distribution in the atmospheric air and mechanisms of tritium contamination within Degelen massif.

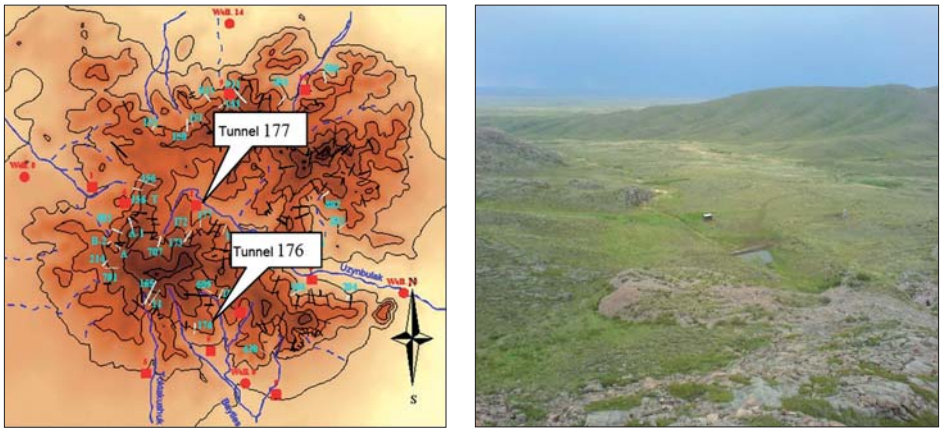
Taking into consideration the peculiarities of explosions carried out at Degelen site, the objects chosen for the study were those the concentration of tritium in which can affect directly the tritium contamination of the atmosphere in this area. These are water, vegetation, and soil water as well as tunnel air.

1.1. Brief description of objects of study

A detailed study has been carried out taking as an example the ecosystems of tunnel No 176 and No 177 watercourses located in the tributaries of the Baitles and the Uzynbulak streams. The basic characteristic of the selected study areas is the existence of tunnel with seepage as open water flowing out to the tunnel portal surface. The channel length is about 400–600, gradually drying up as it moves away from the portal. At a distance of 700–750m there is no permanent water flow in the channel. The annual average discharge of water stream is more than 500 l/min. The faults of East-West and northern directions facilitate the groundwater inflow to the tunnels up to 60 l/min. Hydrologically, Degelen massif is a transit zone of rainfalls which by seeping through the system of conjugated cracks form interstitial water.

The average annual specific activity of tritium in water is 350 and 550 kBq/kg in tunnels No 176 and No 177, respectively [9]. The channel width in both cases does not exceed 1 m. The stream channel in tunnel No 176 is notable for the presence of pond due to the dam built on the stream, abundant vegetation cover on the right side and hard rock brow downstream on the left side. The watercourse channel of tunnel No 177 is notable for the presence of a technological dump in the point where the tunnel watercourse exits to the surface and an abundant vegetation cover on the right and left sides at the beginning of channel; further downstream the vegetation is sparse; hard rock brow on the right side. There is no nearby tunnel with water inflow.

The Figure below shows the location of tunnels where the above-mentioned study has been carried out (Figure 1,a) and the general view of study areas (Figure 1,b).



a b

Figure 1. Location of study areas on Degelen massif site (a), general view of study area (b).

1.2. Collection and preparation of samples

Sampling of atmospheric water vapour

The study of the level of tritium in the atmospheric air has been carried out by taking water vapour samples by the method of cryogenic freezing and liquid scintillation beta-spectrometric analysis of these samples.

The cryogenic freezing method is based on the ability of water vapour present in the air to condense on the surface of radiator plates cooled by liquid nitrogen to below dew-point temperature. Since the temperature of radiator fins is much lower than 0°C, water vapour contact with the plate surface results in crystallization.

The cryogenic plant includes the Dewar vessel filled with liquid nitrogen, cold supply system, a radiator connected a cold supply pipe in permanent contact with liquid nitrogen and a system of liquid nitrogen refilling to the Dewar. The liquid nitrogen refill system is designed to deliver the cryogenic liquid to the Dewar vessel because it permanently boils off. The refill is performed through a filling funnel inserted into the vessel inlet. The general view of the plant for collection of water vapour samples is shown in the figure below (Figure 2, a).

a b

Figure 2. The view of plant for collection of samples of air and free water of plants

Water vapour of atmospheric air was sampled by the open method. A cryogenic plant was installed on a selected site, and freezing out at 50 mm height from the ground surface was provided.

The water vapour samples from soil and tunnels were placed in a vessel making it possible to isolate the entry of atmospheric air to the collected sample. To achieve this, the cryogenic plant was covered with an isolating cylindrical vessel of 100 l in volume. To isolate from the atmosphere, the vessel edges were buried into the earth that was then well compacted. A vacuum pump of 2 l/min power was used to create about 25-30 Pa vacuum inside the vessel to be monitored by a liquid manometer.

Water vapour samples from the tunnel air were taken with a cryogenic plant with the isolating vessel installed on the surface of portal bank so as to position it roughly at the level of the central axis of tunnel. Based on the practical experience, average time of taking one sample should be about 1.5–2 hours to provide the volume of water vapour sample (atmospheric, soil and tunnel air) sufficient for laboratory analyses (about 5 ml).

The following parameters were set for the air sampling: ambient temperature and relative air humidity, which were measured by means of electronic hydrometer [10].

The values of these parameters were used later on for the recalculation of specific activity of tritium in the sample (Bq/kg) to the volumetric activity (Bq/m^3).

Vegetation sampling

Vegetation was selected for the study based on geobotanical description singling out basic vegetation types, assessment of projective cover degree and floristic composition [11] and their classification according to the moisture. Basic studied vegetation species were the wild rye (*Leymus angustus*) and mixed species of meadow herbs the samples taken from which relate to the aboveground part. In some cases, simultaneously with the description of vegetation cover, the vegetative cover productivity was determined by the method of mowing on 3 sites, 1 m^2 each. Under these conditions, herbage plants were cut at 1–3 cm height and tall grasses at 4–6 cm; the current annual increment of subshrubs was cut or plucked. To determine the productivity, the dry-weight excised plants were left to dry to the air-dry weight (breakable) and weighed.

The content of tritium was determined in free water of plants obtained from condensate by a special apparatus constituting in a form of a sealed vessel with cooled surface and a vessel for the collection of moisture evapourated by the vegetation samples (Figure 2, b).

Tritium assessment by liquid scintillation analysis

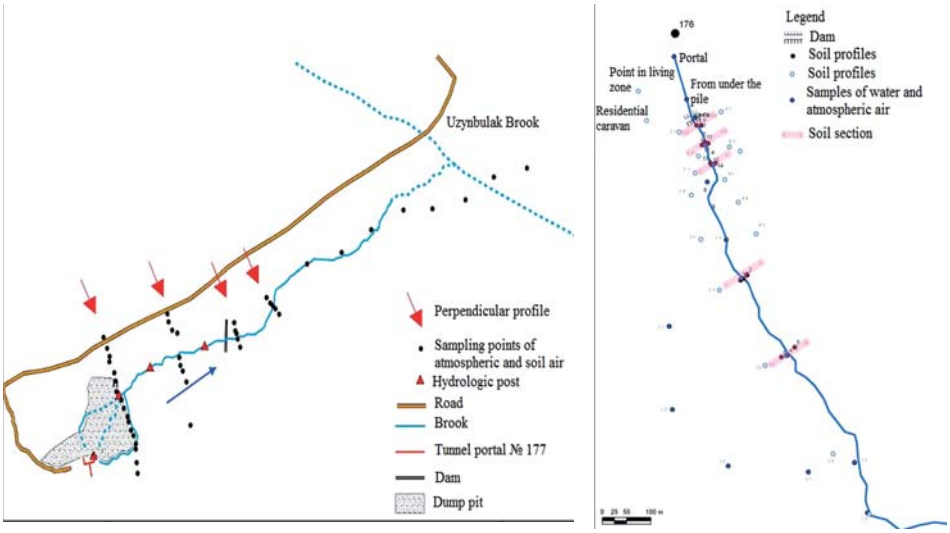
The preparation of samples for measuring the specific activity of tritium complied with the certified procedure [12]. The samples were filtered to remove mechanical impurities; then a 5 ml sample was chosen and placed in a standard geometry (a plastic vial of 20 ml volume) adding Ultima Gold LLT scintillation cocktail at 5:15 ratio (sample-scintillator ratio) specially designed for tritium measurements in natural samples (the registration efficiency for tritium in the 0-18 keV range is about 45-50 %). TriCarb 2900 TR liquid scintillation spectrometer was used to determine the volumetric activity in the samples. The energy range for evaluation of tritium was set to be within 0 to 15 keV. The measurement time for each sample was about 120 min.

2. RESULTS AND DISCUSSION

2.1. Evaluation of level and nature of tritium distribution in the atmospheric air in the vicinity of tunnels at Degelen massif

Taking into consideration the lithium level in tunnel watercourses in the area under study, it is the tunnel stream that can be a basic source of tritium released to the atmospheric air. The investigation of tritium concentration in the air basin in the tunnel stream area was conducted in two directions: along the channel and by the profiles perpendicular to the channel, gradually moving away from it.

The locations on the shore land along the tunnel stream channel at a distance of not more than 50 cm from the water edge and those perpendicular to the mainstream (perpendicular profiles) were chosen to collect water vapour samples of air. The locations under study are shown in the figure below (Figure 3).



a b

Figure 3. Locations under study on tunnels No 177 (a) and No 176 (b)

Distance between the locations along the stream was about 50 m. Distance between perpendicular profiles was 50–100 m and between the points under study in perpendicular profiles – 5–15 m equally right and left of the watercourse.

2.1.1. Study of tritium distribution nature in the atmospheric air along the surface stream channel

To conduct the study along the stream channel, samples of atmospheric air were taken in the locations maximum close to the stream channel, beginning from the point of stream exit to the day surface. The profile length along the channel was 400 m. The results are presented in the figure below (Figure 4).

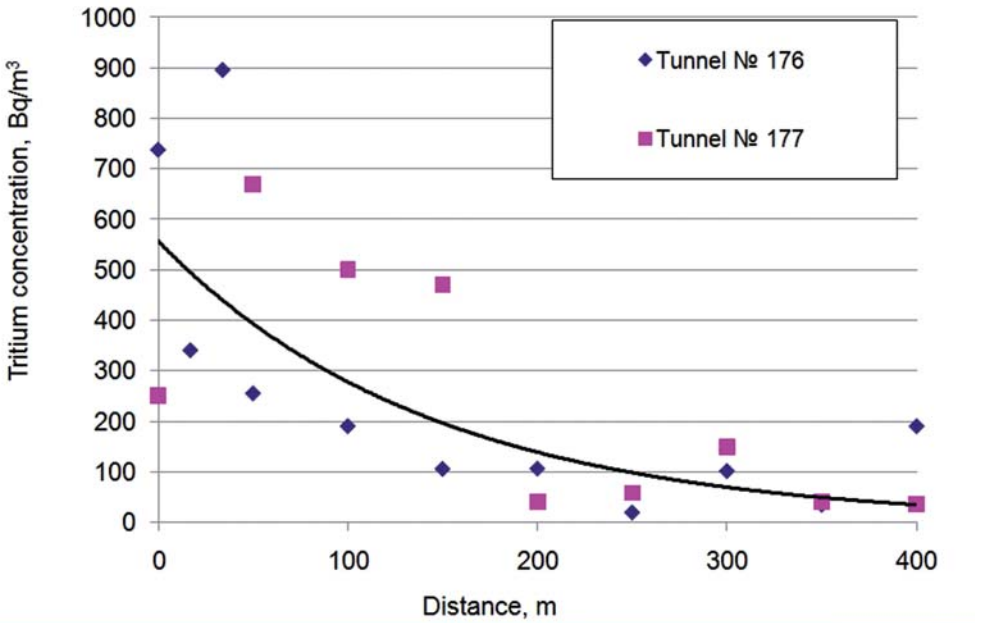


Figure 4. Variation of ^{3}H concentration in the atmospheric air along the surface stream channel

Analysis of data shows that the distribution of tritium in atmospheric air along the surface stream channel is almost the same for both tunnels.

Maximum values on both sites recorded in the points of stream exit to the day surface are 1,400 and 700 Bq/m^3 in the area of tunnels No 176 and No 177, respectively. As the stream length changes, the tritium concentration values in the air basin considerably decrease and are of the order of 50 Bq/m^3 in the area of tunnel No 177 and 100 Bq/m^3 in the area of tunnel No 176 at a distance of 400 m from the study start point.

This pattern displays well the general nature of tritium propagation in the air along a tunnel stream. It should be clearly understood that generally such nature of tritium propagation in the air along the tunnel stream is typical only for these particular sites. For other tunnel sites the general nature of tritium distribution in the atmosphere along the stream would most likely remain the same. With different landscape pattern and hydrogeological regime (possibility of additional water seepage to the channel from the outside, inflows of tritium-contaminated groundwater, etc.) the above nature of tritium distribution in the atmosphere can change: become not so smooth and have jumps (sharp drops) in the concentration.

2.1.2. Study of tritium concentration away from the stream channel

Perpendicular profiles with the air sampling locations uniformly remote from the channel center to the right and left have been set to study the nature of tritium distribution from the surface water source in the stream channel of tunnel No 177 (Figure 3, a). Obtained results are presented in the figure below (Figure 5).

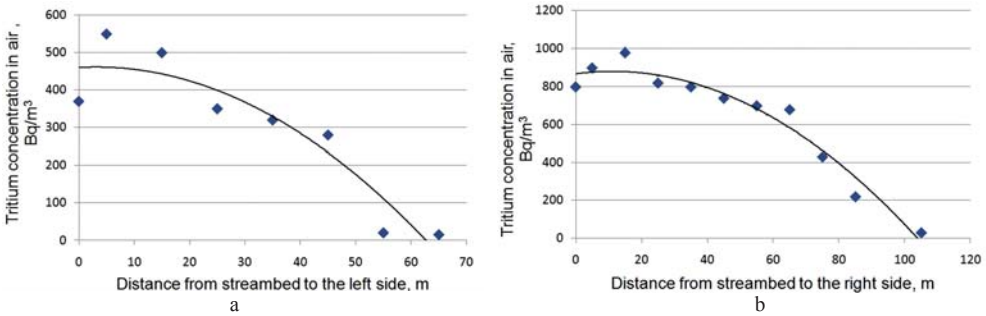


Figure 5. Change of ^{3}H in the atmospheric air moving away from the surface stream channel

Our experiment shows that as the distance from the watercourse increases, the tritium concentrations in the atmosphere decrease for an order of magnitude and more, ranging from 600 Bq/m^3 near the channel to 30–50 Bq/m^3 at 100m away from the channel.

Maximal tritium concentration in the air on the right-hand side of the channel is practically two times higher than that obtained for atmospheric air on the left side, i.e. 1,000 and 550 Bq/m^3, respectively. Such distribution is conditioned by the study area landscape: the right side of the channel is a trough flooded heavily by water in springs what, probably, acts as an additional source of tritium release at this site. However, the left side, on the contrary, leads to a hill near a rock terrain; the soil is dry, stony, and virtually no extra source of moisture from the outside exists

Similar studies have been carried out for the four profiles located perpendicular to the stream channel of tunnel No 177. The figure depicts the general character of tritium distribution at a distance from the water source extending along the axial line (Figure 6). The lines in the figure represent the trends plotted for the values of tritium concentration in the atmosphere for each profile.

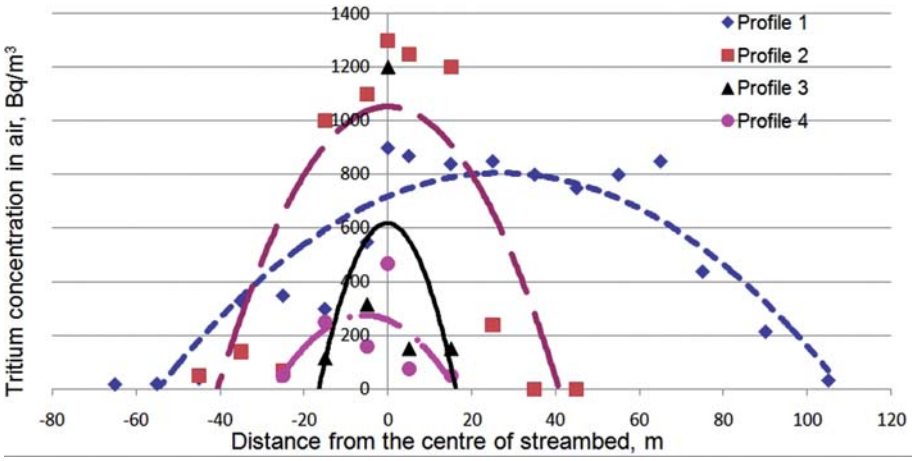


Figure 6. Character of ^{3}H concentration distribution in atmospheric air moving away from the open-channel flow

The highest tritium concentrations in atmospheric air of the ecosystems under study are typical for the tunnel stream locations with high tritium concentrations in the air. Tritium concentration in the atmosphere in the areas of tunnel watercourses significantly decreases at larger distances both along the tunnel stream channel and away from it.

The general pattern of tritium distribution in the air basin on the site of tunnels with water seepage constitutes an isosurface with an arched form of section (Figure 7).

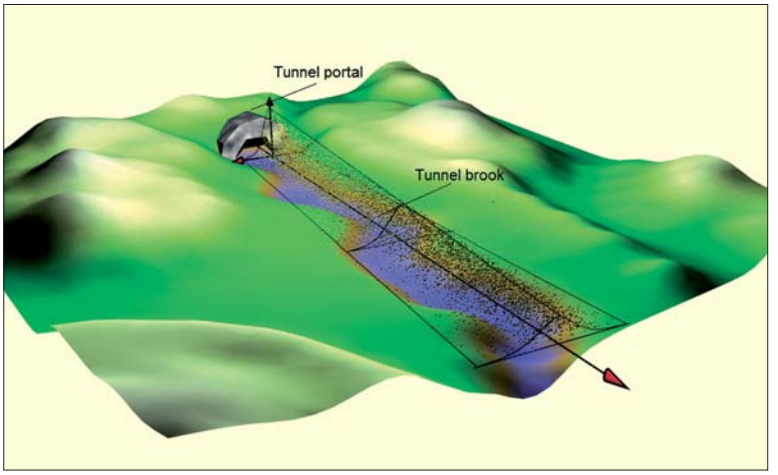


Figure 7. Nature of tritium distribution in the air in vicinity of tunnels with water effects

Obtained pattern of tritium distribution in the atmosphere relative to the surface stream depicts only the general nature as applied to this terrain. Change of any topographic, hydrogeological or landscape conditions at this territory can slightly change the nature of tritium distribution in the air, although the general pattern will, probably, remain, i.e. a tendency towards a decrease in the tritium concentration away from the surface stream.

2.2. Study of basic mechanisms and nature of tritium distribution in atmospheric air of studied ecosystems

2.2.1. Nature of tritium distribution in water-atmospheric air systems

General research methodology

The study of the dependence of tritium concentration in the atmosphere on the tritium concentration in the water was carried out on a single study site to determine the contribution of tritium-contaminated surface stream to the tritium contamination of atmospheric air. The contiguous sampling of water vapours of air and water was conducted at the shore at a minimum possible distance of about 30-50 cm from the water sampling location. Simultaneously, analytical estimation of the dependence of tritium concentration in the atmosphere on tritium concentration in water based on the experimental data was conducted.

Research results

The research results are presented in the figure below (Figure 8).

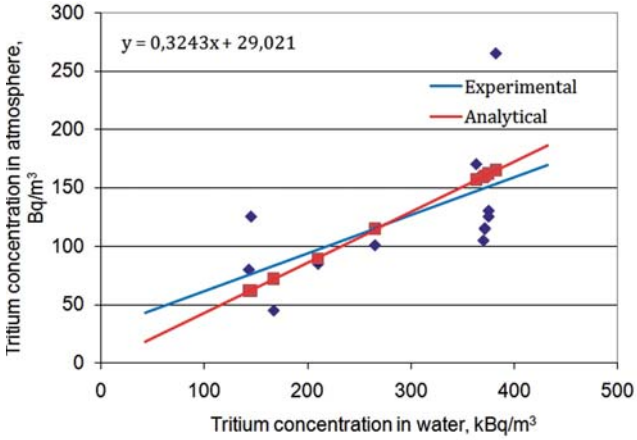


Figure 8. ^{3}H concentration in the atmospheric air versus ^{3}H concentration in water

Our experiments show that tritium content in atmospheric air changes with tritium content in water virtually in direct proportion and can be approximated by the following linear relationship. :

$$C_{T\ in\ atmosphere}\ [Bq/m^3] = 0.32*C_{T\ in\ water}\ [kBq/kg] + 30 \quad (4)$$

According to the obtained relationship, the formula includes a free term, +30, which stands for certain background tritium concentration in the atmosphere (contribution from other release sources) at a tritium concentration in the water equal to "0".

Analytical assessment of tritium concentration in atmosphere near a surface stream has been conducted to confirm the obtained relationship and to make it possible to perform the predictive assessment of tritium content in atmosphere by the computational method.

Analytical assessment of tritium concentration in atmosphere has been conducted based on the assumption that the basic source of tritium release to the air in locations most close to a stream is the stream itself contaminated by tritium. The value of tritium concentration in the atmosphere depends on the intensity of water evapouration from the stream surface and the rate and nature of its propagation in the air [13, 14].

As the samples were taken in the immediate vicinity of the watercourses (~0.5 m from the water edge) and at low wind speeds, any significant diffusion of water vapours in wind-transverse directions at such a distance from the source of contamination would not occur. Therefore, it was not reasonable to use Gaussian model of impurities proliferation in the air depending on the coefficients related to the atmospheric stability. In this paper it is assumed that the water vapours diffuse uniformly in the near-surface air volume within the time during which wind passes the distance from one stream bank to another and, hence, concentration of tritium released from the stream water is described by the following formula:

$$C=E*TFC*A_{water}*(1/H)*(L/(v*\sin(\alpha)), \text{ where} \quad (5)$$

C – the concentration of tritium in the air, Bq/^{3};

E – the water evapouration rate;

TFC – the temperature adjusting factor;

A_{water} – the activity of tritium in water, Bq/kg (using practical data);

V – the wind speed relative to the soil level, m/s;
A – the angle between wind directions and fluid motion direction, radian;
H – the height (equal to the height of cryogenic plant), m; and
L – the width of stream, m.

Intensity of evapouration (evapouration rate) is generally a function of saturated vapour pressure, water and air temperature, heat of evapouration, speed of wind, water, etc. and is described in literature by semiempirical formulas. In this work the methodology worked out by the Environmental Protection Agency of the USA (EPA U.S.) is used as having an official status of federal technical manual. In accordance with this methodology the evapouration of water is determined by the following formula [15]:

$$E=10.4*u^{0.78}*M^{0.667}*P_f/(R*T_f), \text{ where} \quad (6)$$

E – the evapouration rate, kg/f*m^2;
U – the air speed relative to the water surface, m/s;
M – the molar weight of fluid, 18 g/mole;
P_f – the pressure of saturated vapour of fluid at a specified fluid temperature, kPa;
R – the universal gas constant, 82.05 atm*cm^3/(mole*K); and
T_f – fluid temperature, K.

As this formula has been obtained on the assumption that the temperatures of water and environment are equal, for the conditions when these components have typically different temperatures (like in this study) it is recommended to multiply the evapouration rate by the adjusting temperature factor [16]:

$$TFC=P_f*T_{air}/(P_{air}*T_f), \text{ where} \quad (7)$$

P_f – the pressure of saturated vapour at T_f temperature, kPa;
T_f – the fluid temperature, K;
P_{air} – the pressure saturated vapour at T_{air} temperature, kPa; and
T_{air} – the environment temperature, K.

Calculation of tritium concentration is based on the average values of the range of the following parameters: T_f, T_{air}, v, H, L, A_{water} determined experimentally (Table 1).

Table 1.

Initial data for performing the analytical calculation

Parameter	T_f, K	T_{air}, K	v, m/s	H, m	L, M
Average	288	300	1	0.7	1
Minimum	286	293	0.5	0.6	0.8
Maximum	290	303	1.3	0.8	1.2

Based on the analysis, the analytical curve of dependence of tritium concentration in the atmosphere on the tritium concentration in the water has been plotted (Figure 8). Tritium concentrations determined by experimental and analytical methods differ by not more than 20 %.

To assess the effect of basic parameters to a various degree influencing the value of tritium concentration in the atmosphere, the analysis of variation of tritium concentration in the air on changing these parameters has been performed in the range between their minimum and maximum values recorded during the experiment (Table 1).

The analysis is based on the simulation model employing the Monte-Carlo method randomly using the basic parameters influencing the concentration of tritium in the air, such as the fluid temperature (T_f), air temperature (T_{air}), wind speed (v_{wind}), stream width (L). The simulation made us possible to build a model to describe a range of tritium concentration values in the air (Figure 9).

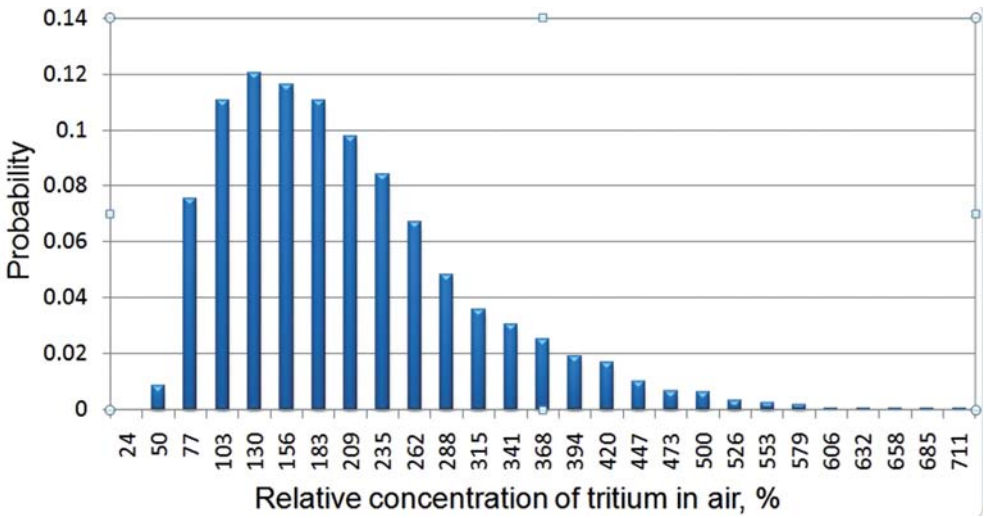


Figure 9. The range of model parameters determined experimentally

Another model has been constructed to assess the change of tritium concentration in the air under the anomalous natural conditions. The spread of tritium concentration values in the air at a more significant difference in the maximum and minimum values of basic affecting parameters has been studied. The study is based on the assumption that the maximum values of specified parameters (Table 1) differ from the specified mean value by 50 % (Table 2).

Table 2.

Initial data for performing the analytical assessment

Parameter		T_f, K	T_{air}, K	v, m/s	H, M	L, M
Value	Mean	288	300	1	0.7	1
	Minimum	280	286	0.5	0.35	0.5
	Maximum	295	313	1.5	1.5	1.5

The frequency profile for the model parameters varying within 50% range is shown in the figure below, (Figure 10).

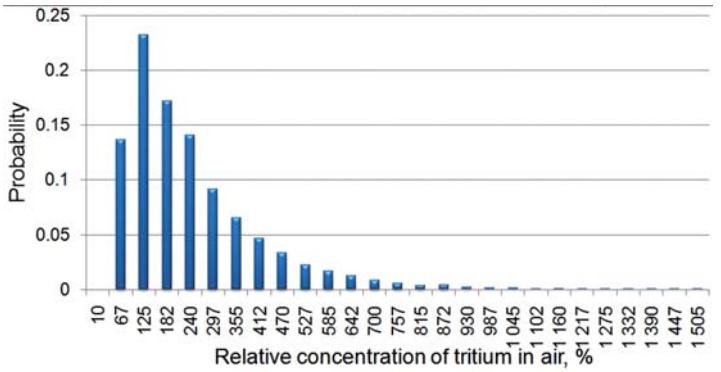


Figure 10. Range of model parameters varying within 50%

Probability distribution analysis of tritium concentrations in the air, with the specified tritium concentration in the air determined by independent variation of the estimation model parameters between their minimum and maximum values, shows that at 80 % confidence the tritium concentration values in the air can vary from 75 to 340 % relative to a mean value (or from 63 to 279 Bq/m^3).

It therefore follows that the dependence of tritium concentration in air on tritium concentration in water can be expressed as:

$$C_{T\ in\ at.\ air}\ [Bq/m^3] = (0.26 \div 1.03) * C_{T\ in\ water}\ [kBq/kg] \qquad (8)$$

Assessment of model sensitivity to the basic parameters has been made to determine the effect of individual significant parameters on the tritium concentration value near a surface stream (Figure 11). Three most significant parameters have been chosen for such assessment: air temperature, water temperature and wind speed.

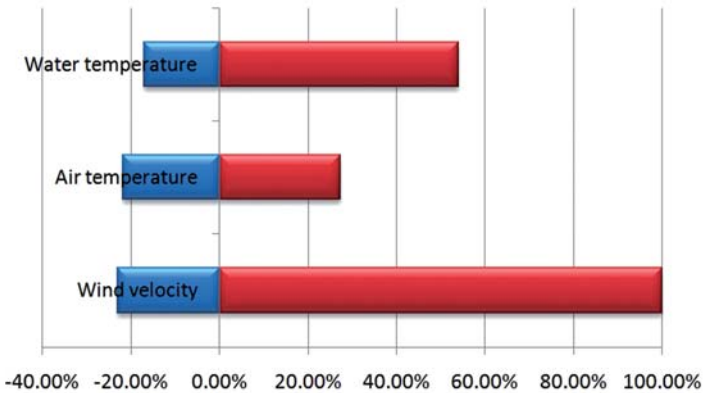


Figure 11. Effect of individual parameters on the concentration of tritium in the air

Sensitivity analysis shows that tritium concentration in air is mostly affected by the air temperature and wind speed. Uncertainty in the value of water temperature affects tritium concentration in the air much less, although this parameter cannot be totally neglected.

According to the studies and analytical assessment, tritium concentration in atmospheric air near a surface stream contaminated with tritium changes proportionally to the tritium concentration in water depending on a number of parameters such as wind speed, air temperature and water temperature.

According to the probability distribution analysis, tritium concentration in the atmosphere according to the above parameters can vary within 75 - 340 % (from 63 to 279 Bq/m^3) range taking into account the maximum spread of variable values.

Assessment of tritium concentration in air by tritium concentration in water using the experimental and theoretic formulas is consistent. By the predictive assessment with the formula obtained from the experimental data (4) it is found that at the same specified tritium concentration in water (≈270 Bq/m^3) used for the assessment of the spread of estimated values, tritium concentration in air is of the order of 120 Bq/m^3 what fits the above mentioned range.

2.2.2. Nature of tritium distribution in the tunnel air-atmospheric air system

General research methodology

The main objective was to assess the significance of contribution from tritium released to the atmospheric air from tunnels.

The first stage was to study the nature of tritium distribution in the air basin inside a tunnel. To achieve this, four tunnels without water seepage were chosen for air sampling, beginning from the tunnel portal and further in-depth at 10 to 60m spacing depending on the work conditions.

The second stage was to study the level of tritium adjacent to the portal of "suspended" tunnels without any visible water seepage. Since the tunnel sprung-hole after the "suspension" was not perfectly air-tight, and the motion of air along the tunnel through the crushing zone to the day surface and back was possible due to the gas convection processes, tritium could migrate with the air flows from the tunnel to the atmosphere. Water vapour samples from the tunnel air were taken directly on the tunnel portal and at a distance of 50 m from each tunnel to assess the contribution of tunnel air to the tritium contamination of the air.

Results of investigation

Tritium concentrations in the air inside the tunnels are shown in the figure below, (Figure 12).

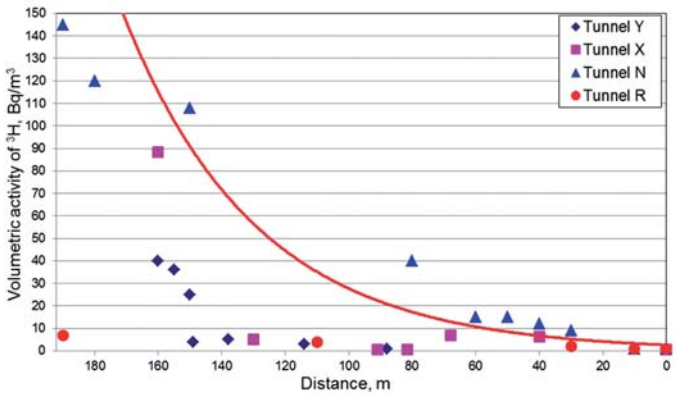


Figure 12. The nature of tritium distribution with the air flows inside tunnels

Our analysis shows that tritium contamination in the air of the tunnels under study ranges from 140 to 1 Bq/m^3 in the tunnel depth and on the portal respectively (according to the results of studying four tunnels). Maximal concentration of tritium inside a tunnel corresponds to the level nearest to the end box were the explosion was conducted, and the major amount of tritium formed due to the explosion should be right there. Approaching the tunnel exit, the concentration of tritium considerably drops due to both moving away from the explosion epicenter and the diffusion mixing of tunnel air layers with the atmospheric air.

At the second stage of research, the contribution of in-tunnel air tritium to the tritium contamination of air on the tunnel portal has been assessed (Figure 13).

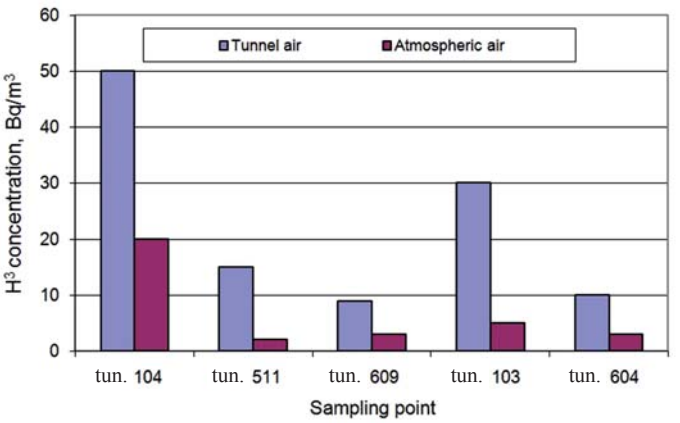


Figure 13. Concentration of ^{3}H in the air on the tunnel portal

We have shown the possibility for tritium to release from the "suspended" tunnels to the surface with the air flows. Concentration of tritium on the portals is up to 50 Bq/m^3, which is essential taking into consideration that the background tritium concentrations in the air within Degelen massif are of the order of 1–2 Bq/m^3. However, further investigations have shown that the tritium concentration in the atmosphere at a distance of 50 m from the tunnel portal is 5–15 % of the tritium concentration in the tunnel air, which is virtually a background level and cannot contribute considerably to the tritium contamination of the atmosphere.

Tritium concentration in the tunnel air does not therefore contribute much to the tritium contamination of the atmospheric air in the area of tunnel location. The tunnel air is not regarded as a significant source of tritium inflow to the atmosphere.

2.2.3. Nature of tritium distribution in the soil air-atmospheric air system

General methodology of investigation

As it is known that part of underground water is consumed by the evapouration from the ground surface [17], it can be assumed that under the present conditions tritium can release to the atmosphere with the evapourated moisture.

The graph below (Figure 14) shows that the evapouration of underground water begins at 2.5 m depth where, considering the transpiration by vegetation, the amount of evapourated water is 100 mm. Maximum values up to 700 mm per year are achieved by this process at the depth up to 20 cm. This graph has been constructed using mean values, not registering

such parameters as the permeability and porosity of soil the value of which varies depending on the soil type and affects to a considerable degree the amount of moisture evapourated from the ground surface.

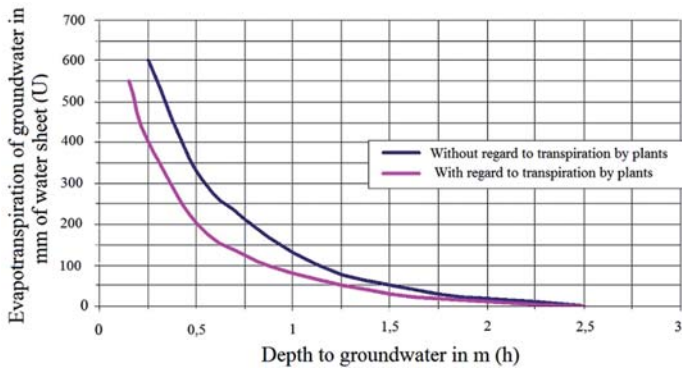


Figure 14. Nature of moisture evapouration from the ground surface depending on the depth of groundwater occurrence

The objective of research was to determine the tritium concentration in the soil air and to assess the contribution of soil air tritium to the tritium contamination of the atmosphere.

For the experiments, dry channels of major streams of Degelen massif have been chosen where, according to the hydrogeological maps, underground watercourses existed and, consequently, the release of tritium to the atmosphere with the soil air is possible. According to the research outcomes, concentration of tritium in the groundwater on Degelen site is about 300 kBq/kg [18]. Water vapour sampling from the soil and atmospheric air has been carried contiguously, that is simultaneously, at a minimum distance from each other (to 50 cm) in the same point of investigation. The sampling grid is shown in the figure below (Figure 15).

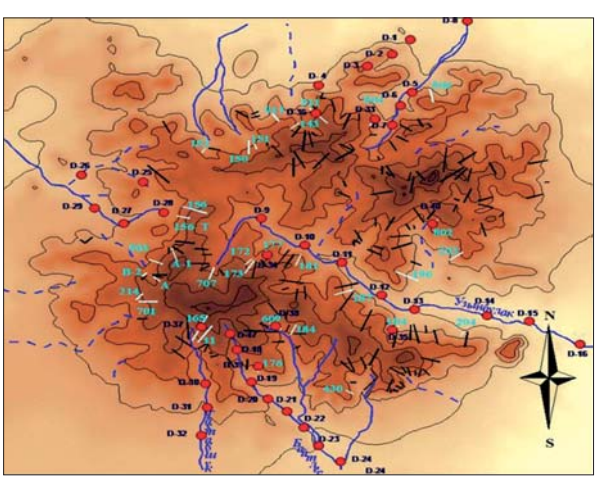


Figure 15. Locations for the collection of water vapour samples from the atmospheric and soil air on Degelen massif site.

Results of research

The results are presented at the figure below (Figure 16).

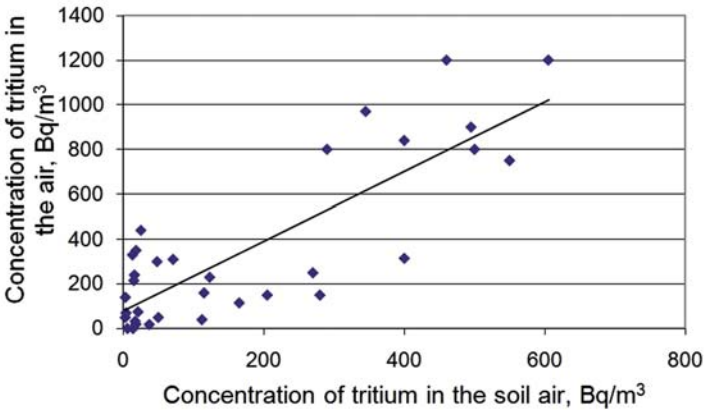


Figure 16. The relationship of ^{3}H concentration in the atmospheric and soil air

Tritium concentration values in the soil air vary over a wide range. During the data analysis in a number of cases it has been noted that the tritium concentration in the soil water exceeds or is comparable to the tritium concentration in the atmosphere.

The obtained relationship has been approximated and described by the following function:

$$C_{T\ in\ at.\ air}\ [Bq/m^3] = 1.6*C_{T\ in\ soil\ air}\ [Bq/m^3] + 30 \qquad (9)$$

The background concentration of lithium in the atmosphere, given the concentration of lithium in the soil air equal to zero, is described by a free member of this equation, which is 30 Bq/m^3 for the experiment performed.

The research has shown that on the tritium-contaminated sites of groundwater occurrence (at not more than 2.5–3 m depth) there proceeds the release of tritium to the atmosphere due to the processes of emanation from the ground surface.

Having the data on the tritium concentration in the soil water, one can estimate the ^{3}H concentration delivered to the atmospheric air and thus evaluate tritium contamination of the atmosphere on a selected study site.

Taking into consideration the specific features of Degelen site such as the high tritium concentration in the groundwater and low depth of occurrence, the revealed nature of tritium distribution fits only to this territory or territories with similar groundwater characteristics and, perhaps, will not occur on other study sites.

2.2.4. Nature of tritium distribution in the vegetation-atmospheric air system

General research methodology

During the investigations of tritium contamination in the tunnel streams on Degelen massif the concentration of tritium in the water was found to be up to 300–400 kBq/kg [19]. It has been assumed that at so high concentrations in the water the shore vegetation must have high concentration of tritium and, in its turn, act as a source of tritium releasing to the air due to the vegetation transpiration processes.

To check and confirm this, an experiment to investigate the tritium concentration in the free water of wild rye (*Leymus angustus*) has been first conducted at the tunnel stream shore (tunnel No 176). Samples from the vegetation have been taken on a profile perpendicular to the main channel of tunnel watercourse with 10m spacing between the sites under study. The results are presented in the figure below (Figure 17).

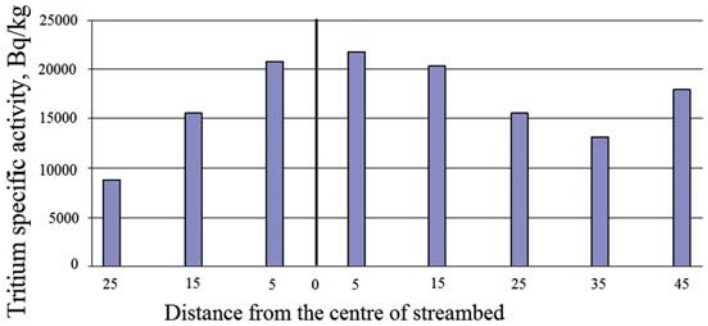


Figure 17. Investigation of tritium concentration in the free water of plants

As a result of investigation, the tritium concentration values have been found to range from 80,000 to $2.2*10^5$ Bq/kg. Based on the obtained data, a number of experiments have been conducted to identify a possible relationship between the concentration of tritium in the atmospheric air, concentration of tritium in the vegetation and productivity of vegetation cover.

With this end in view, four profiles have been established on the site of tunnel No 177 perpendicular to the tunnel watercourse. Each profile included several study sites, 5 to 18 depending on the surface topography. The distance between the study sites was 5–15 m. The collection of samples from the air and vegetation (composite samples) was performed in a contiguous manner: 1 sq. m area was allocated on each study site, and vapour samples from the air were taken in the center of it. Further, the vegetation on the site was mowed down to assess the productivity of vegetation cover.

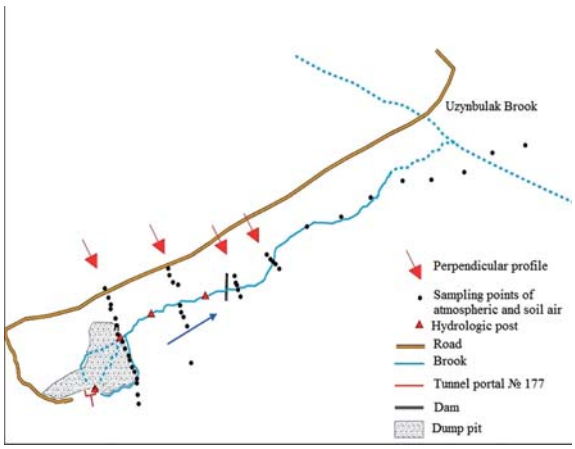


Figure 18. Atmospheric air and vegetation sampling grid (along the perpendicular profiles) on the site of tunnel No 177

Results of research

The analysis of obtained data has shown the tritium concentration values in the free water of vegetation on the selected sites to vary from 3 to 250 kBq/kg. The tritium concentration values in the air on the same study sites vary from 80 to 1,200 Bq/m^3.

Based on the obtained results, the diagrams of the dependence of tritium concentration in the atmospheric air on its concentration in the vegetation (Figure 19, a) and variation of tritium concentration in the atmosphere with the productivity of tritium-contaminated vegetation cover (Figure 19, b) have been constructed.

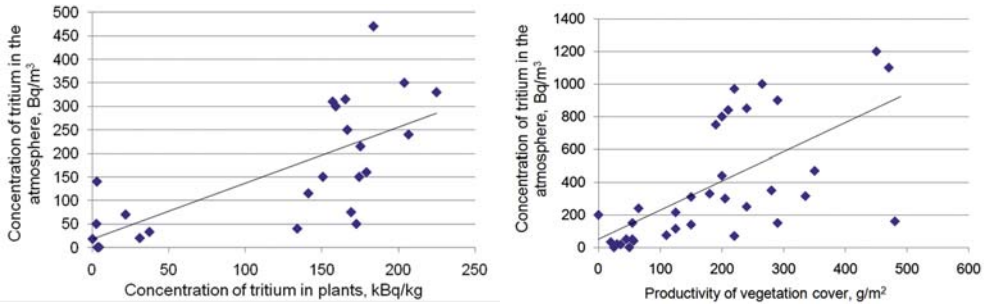


Figure 19. ^{3}H concentration in the atmospheric air as a function of ^{3}H concentration in the vegetation (a) and of the productivity of vegetation cover contaminated with ^{3}H (b)

The data on these diagrams have been approximated by a linear function. Using the obtained relationships it is possible to predict the tritium concentration in the atmospheric air based on its content in the vegetation via the following empirical equation:

$$C_{T\ in\ at.\ air}\ [Bq/m^3] = 1{,}2 * C_{T\ in\ vegetation}\ [kBq/kg] + 50 \qquad (10)$$

For the predictive assessment, the productivity of vegetation cover on the study site should be taken into account.

The tritium contamination of atmospheric air on the tritium-contaminated vegetation sites is, probably, caused by the vegetation transpiration processes. Obtained experimental relationships have confirmed the contribution of vegetation to the tritium contamination of the atmosphere.

In accordance with the above format, the formula includes a free member, +50, which represents the background tritium concentration at the site due to other sources of tritium release to the atmosphere.

CONCLUSIONS

Investigation of tritium levels in the air of Degelen massif on the sites of tunnels with water seepage shows that tritium contamination of the atmosphere at the tunnel portals is due to several basic sources. These are the soil air on the sites of shallow groundwater, surface water and vegetation. The basic sites contaminated with tritium are along the stream channels due to the carry-over of tritium by watercourses from the tunnels.

Tritium concentration in the air decreases considerably as the distance from the tritium release source increases due to the diffusion mixing of air beds, and decreases for an order and more compared to the initial value at distances about 50-70 m.

Tritium concentrations recorded for the air on the tunnel portals are in some cases higher than 1,000 Bq/m^3 what does not exceed but is close to the margin of activity concentration in the air for the population, being 1,900 Bq/m^3 [20].

It should be noted that the obtained relationships can vary depending on the conditions and on site, and some sources of tritium release to the atmosphere (for instance, water) may not be present. In this paper we presented the level and nature of tritium distribution in the air basin relative to Degelen site conditions, namely, the location of tunnels with water seepage.

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"ДЕГЕЛЕҢ" ТАУЛЫ МАССИВІ ШЕГІНДЕГІ АУА БАССЕЙНІНІҢ ТИРТИЙМЕН ЛАСТАНУЫН ҚАЛЫПТАСТЫРУ МЕХАНИЗМДЕРІ

О.Н. Ляхова, С.Н. Лукашенко, Н.В. Ларионова

Тритиймен ластану мәселесі табиғи орталардың радиоактивті ластануында ерекше орын алады. Аталған жұмыста "Дегелең" алаңында орналасқан су шығулары бар №176 және №177 штольнялар үлгісіндегі экожүйенің атмосфералық ауасында тритийдің таралу деңгейіне және сипатына бағалау жүргізілген. Тритиймен ластанған беткі су ағынына қатысты тритийдің атмосферада таралуының жалпы сипаттамасы келтірілген. Зерттелетін экожүйенің атмосфералық

ауасында тритий таралуының негізгі механизмдері, нақтырақ, тритийдің келесі экожүйелердегі таралу сипаттамасы: су - атмосфера, штольнялық жел – атмосфера, топырақ ылғалы – атмосфера, өсімдік - атмосфера зерттелген. Тритийдің судағы шоғырлануы бойынша тритийдің атмосферадағы шоғырлануына талдамалық есеп жүргізілді. Болжамдық бағалау жүргізу үшін түсу көзінің біріндегі – су, штольнялық ауа, топырақ ылғалы, өсімдіктегі - тритий шоғырлануынан тритийдің ауадағы шоғырлануының бағыныштылығы тәжірибелік жолмен алынды. "Дегелең" алаңы аумағындағы ауа ортасында тритий таралуының жалпы сипаттамасы келтірілген.

Түйін сөздер: Дегелең алаңы, тритий, атмосфералық ауа, тритиймен ластану, тритий таралуының сипаттамасы.

МЕХАНИЗМЫ ФОРМИРОВАНИЯ ЗАГРЯЗНЕННОСТИ ТРИТИЕМ ВОЗДУШНОГО БАССЕЙНА В ПРЕДЕЛАХ ГОРНОГО МАССИВА "ДЕГЕЛЕН"

О.Н. Ляхова, С.Н. Лукашенко, Н.В. Ларионова

Вопрос тритиевого загрязнения занимает особое место в радиоактивном загрязнении природных сред. В данной работе проведена оценка уровня и характера распределения трития в атмосферном воздухе экосистем штолен с водопроявлением на примере штолен № 176 и № 177, расположенных на площадке "Дегелен". Представлен общий характер распределения трития в атмосфере относительно поверхностного водотока, загрязненного тритием. Изучены основные механизмы распределения трития в атмосферном воздухе изучаемых экосистем, а именно, характер распределения трития в системах: вода – атмосфера, штольневый воздух – атмосфера, почвенная влага – атмосфера, растительность – атмосфера. Проведен аналитический расчет концентрации трития в атмосфере по концентрации трития в воде. Экспериментальным путем получены зависимости для проведения прогнозной оценки концентрации трития в воздухе от концентрации трития в одном из источников поступления – вода, штольневый воздух, почвенная влага, растительность. Представлен общий характер распределения трития в воздушной среде на территории площадки "Дегелен".

Ключевые слова: площадка Дегелен, тритий, атмосферный воздух, тритиевое загрязнение, характер распределения трития.

УДК 577.391:577.4:504.74:636.32/.38:539.16

EXPERIMENTAL STUDIES OF ARTIFICIAL RADIONUCLIDES TRANSFER IN ORGANS AND TISSUES OF SHEEP AT STS

Zh.A. Baigazinov, A.V. Panitskiy, S.N. Lukashenko, S.A. Keller, S.S. Karataev, A.R. Ivanova

The paper reports on the field studies of the peculiarities of artificial radionuclides transfer in organs and tissues of sheep for different conditions and periods of radionuclides intake. A quantitative assessment was made for intake of T, ^{137}Cs, ^{90}Sr, $^{239+240}$Pu and ^{241}Am with components of the environment: air, water and vegetation based on their actual contents at "Degelen" experimental site. It was determined that the bioavailability of ^{137}Cs and ^{90}Sr to organs and tissues of sheep intaken with tunnel water is higher than their bioavailability for intake with forage plants. Bioavailability of T intaken with food is higher than that of T intaken with water. We determined that regardless of the time and type of intake, the distribution of ^{137}Cs, ^{90}Sr and T in the organs and tissues has a dependency that makes it possible to assess the content of these radionuclides in various organs by one of them.

Key words: Semipalatinsk Test Site (STS), transfer factor (R), sheep, tissues and organs, radionuclide bioavailability, tritium (T), cesium-137 (^{137}Cs), strontium 90 (^{90}Sr), plutonium-239 +240 ($^{239+240}$Pu), americium-241 (^{241}Am).

INTRODUCTION

Unsupervised agricultural activities in the territory of the Semipalatinsk test site (hereinafter STS) is one of the most important problems, as the transfer of radionuclides through the trophic chains is related to their uptake into the human body. It should be noted that for livestock production that meets the requirements of radiation safety, knowledge about the patterns of intake and distribution of artificial radionuclides in the organs and tissues of animals have higher priority value, since animal husbandry is the main activity for this region.

Peculiarities of ^{90}Sr transfer to animal body. Patterns of accumulation and distribution of ^{90}Sr in the bodies of laboratory [1, 2] and farm [3, 4, 5] animals have been studied quite well. All the authors note that ^{90}Sr, in any way of its intake into the body, is accumulated in the skeleton, and competes with calcium in the processes of bone formation.

In order to explain the absorption of ^{90}Sr we proposed membrane hypothesis, implying the possibility of diffusion through the pores of the membranes and its transfer from the contents of the gastrointestinal tract into the blood [6, 7]. One should note that absorption of ^{90}Sr depends on age of an animal; in young animals the incorporation of ^{90}Sr in bone tissue is faster than in adults. No significant differences in the accumulation of ^{90}Sr in the skeleton of females and males of different groups have been found [8].

Rate of ^{90}Sr incorporation in bone tissue varies for different parts of one skeleton. However, to calculate the activity of the whole skeleton, we can confine to analysis of one - three ribs, taken as a whole, or scapula [9].

In the transfer of calcium (hereinafter- Ca) and ^{90}Sr through biological chains there is a preference for calcium relative to ^{90}Sr, that is expressed in coefficient of discrimination. Exchange of ^{90}Sr and calcium in the body is subject to general patterns and in the transfer from food to the skeleton the relation between them remains, which is available in food with certain correction for discrimination of ^{90}Sr relative to Ca [10, 11].

Protective effect of calcium against the deposits of ^{90}Sr has been studied sufficiently well [12, 13]. Also, a protective effect against the deposits of ^{90}Sr can be considerably enhanced by enriching the diet with magnesium, phosphorus, and fat [14, 15]. Activities to accelerate the removal of ^{90}Sr from the body should be carried out soon after intake of the isotope in the body [16].

Peculiarities of ^{137}Cs transfer to animal body. Data on the nature of ^{137}Cs metabolism in the body of sheep are adequately presented in experimental studies on oral priming of animals with solutions of ^{137}Cs [17, 18] and in studies of radionuclides transfer in sheep from global fallouts and fallouts after the Chernobyl accident [19, 20, 21].

Cs radionuclides are readily absorbed and incorporated into the process of metabolism in mammals. Absorption of ^{137}Cs in the body of sheep depends on the age of animals, forms of its intake and stable isotope ratio in the diet [22, 23, 24].

Physical-chemical properties of ^{137}Cs allocate this element is predominantly in muscular tissue, relatively evenly distributing all over and other soft bodies [25, 26]. As a result of experiments with separation of muscular tissue into proteins, lipids, glycogen and water-soluble fraction we determined that the ^{137}Cs is gradually incorporated in the bioorganic elements, which may affect period of removal [27].

It should be noted that contrary to the opinion that ^{137}Cs is always accompanies potassium, some researches disagree with this statement. Data obtained by L.G. Shahidzhanyan [28, 29], rejects the theory that the distribution of ^{137}Cs is due to hematoencephalic and placental barriers to the element Cs. Further findings on this issue have not been found.

Peculiarities of T transfer in animal body. T, as an isotope of hydrogen, can substitute it in any molecule inside the cell. Tritium water enters the cell without segregation by simple diffusion. Distribution of T among the molecules in cells is proportional to the hydrogen content in the molecules.

There was studied the influence of the form of T compound (organic and inorganic) on the kinetics of exchange in single and chronic mechanical intake into the body of laboratory animals [30, 31, 32]. It is shown that T in the organic compounds is retained in much larger quantities and got out more slowly than inorganic compounds.

The pattern of T distribution in the single and chronic intake is determined by compound form introduced [33]. Effective half-life organically bound T in piglets that intook T with milk (sows were fed with tritiated water) was 59 days for the brain, 28 days for muscles, 22 days for kidneys, spleen and stomach sweetbread, 17 days for the liver and intestine [34]. Also it was found that the biological half-life of T in the DNA was 318 days [35].

In mammals, at the chronic consumption of T balance with the exchange of its hydrogen is quickly reached. Upon termination of T access the activity of tissue decreases more slowly than the water activity of the body, so in the new balance conditions, the activity of tissues exceeds that of water [36].

Part of the organically bound T, intaken by the body, goes into tritium water of the cells, the other part - the synthesis of proteins and fats. Proteins and fats pass into the milk and forming organs and tissues of the fetus [37, 38].

Experiments on rabbits [39], which were fed with tritium water and alfalfa within 2 weeks showed that the specific activity of aqueous and organic fractions of tissues of all animals was the same as food and water, which indicates the absence of biological concentration of T.

Accumulation of the organically bound T in the body depends on the period of intake and physical impacts. The content of organically bound T in organs and tissues can be up to 25% with respect to tritiated water [40].

Features of the transfer of $^{239+240}$Pu and ^{241}Am in the body of animals. Pu in the blood 90–95 % is coupled with the globulin proteins or lipoprotein complex, and only 15% goes to the ultrafiltrate [41]. On the contrary, the relationship of Am^{3} + with serum proteins is weak, nonspecific, and, apparently, plays an important role in the capture of a polymeric Pu, Am by liver cells. It is also known that the mechanism of intestinal absorption of Pu depends on the age of the animal, so Pu absorption in adult animals is 10 times lower than in neonates [42].

Accumulation ^{239}Pu, ^{241}Am in the muscle of cattle from received the daily amount per 1 kg of muscle ranges within $(1–4)\times10^{-4}$ %, sheep - $(0.3\text{-}1.0) \times 10^{-3}$%, and chickens $(2\text{-}7) \times 10^{-3}$%. No data on transfer coefficient of Pu and Am elements in cow's milk are available [43].

In 1986 and 1987 there were studied the transfer of Pu and Am isotopes in the tissue of sheep on natural pastures [44]. The experiment found that in the gastrointestinal tract to digest 0.01% of Pu and 0,005% of Am of the total number of intaken with feed for animals. The main depot is the liver and skeleton. ^{239}Pu initially is distributed on the surface of bones, but after 1-6 months penetrates into the bone stock and binds in the nuclei of cells [45].

It is known that transfer coefficient (hereinafter Kn) for animals that have been subjected to Pu action prenatally, is 4 times higher for thighbone and 2 times higher than for all other tissues compared with old animals that have begun to graze on contaminated pastures [46].

When inhalation of soluble compounds of ^{239}Pu the main depositing organs are the lungs and the skeleton [47]. The value of deposits of ^{239}Pu in the skeleton does not exceed 9-30%, and in the liver – 4.1-7% of the original content in the lungs after inhalation intake. When it is intaken through the intact skin of pigs the citrate plutonium is absorbed 6 days in an amount of 0.347% of the damage to the skin, 0.183% of which is absorbed within the first days. Distribution of the resorbed fraction of emitter at cutaneous application differs a little from the distribution after internal injection. In both cases, more than 81% retains in the skeleton and not more than 18.5% - in the liver [48].

The analysis showed that so far the study of the issue has been the subject of many studies, the main results of which are included in the IAEA database [49], some of them are listed in the table (Table 1). As the table shows, transfer of Sr and Cs has been studied well enough, but the transfer coefficients variation occurs in a wide range. Very few researchers studied transfer of plutonium and americium; there are no data on tritium. These facts justify the urgency and the need to study this issue in terms of the former STS, where similar studies have been conducted neither.

Table 1.

Transfer coefficients of radionuclides in muscular tissue of sheep

Radionuclide	min	max	Total works	Radionuclide	min	max	Total works
Sr	3.0×10^{-4}	4.0×10^{-3}	25	Pu	2.0×10^{-5}	$8{,}5\times10^{-5}$	2
Cs	5.3×10^{-2}	1.3	41	T	No data		
Am	4.8×10^{-4}		1	–	–	–	–

1. BASIC RESEARCH METHODOLOGY

1.1. Research venue

Studies were carried out at "Degelen" experimental site, located in the south-western part of the former STS. To conduct the study we selected tunnels (#176 and #177) with water seepage, formed by groundwater flowing from the sprung holes of the tunnels [50], thereby forming a radioactive contamination of the environmental components of soil, water, vegetation and air. Sites were selected as one of the tunnels with water seepage most contaminated ^{137}Cs, ^{90}Sr, $^{239+240}$Pu, ^{241}Am and T.

1.2. Subject of research

As the experimental animals we used fat tail sheep of the Kazakh coarse-wooled breed ("Edilbaevskaya"). All the animals were similar in clinical condition and exterior-statical data.

Selection of the subject and research venue is justified by the fact that sheep farming is the main farm activities in the region and that near the "Degelen" site there is the largest number of households engaged in animal husbandry at the STS [51] (Figure 1).

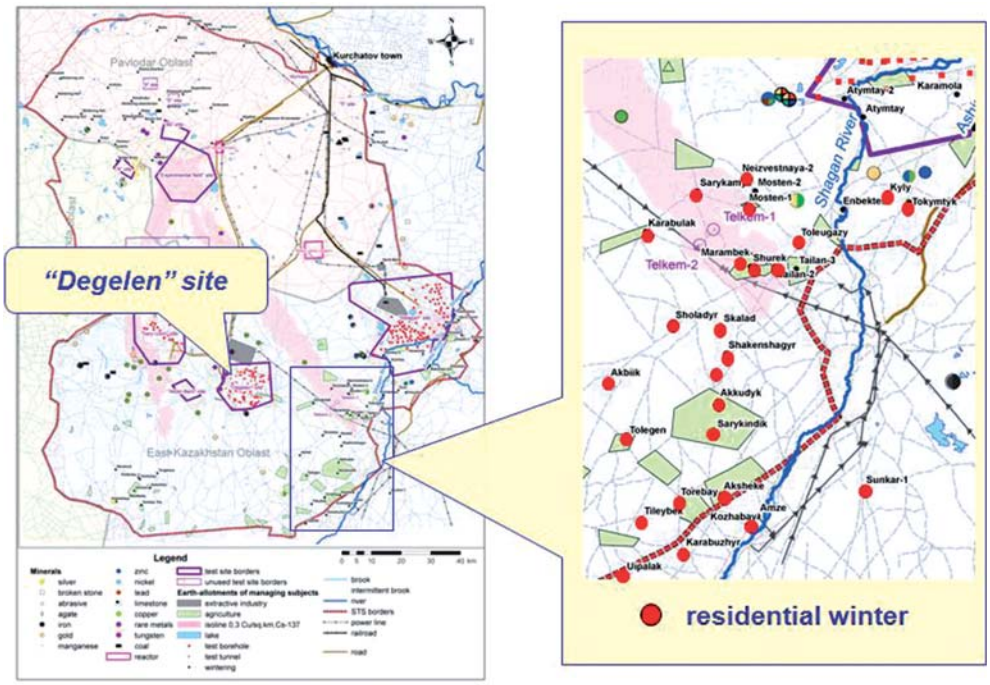

Figure 1. Residential wintering sites located in the south-western part of STS

1.3. Research procedure

The studies were performed under various conditions of animal husbandry:

- under different timing of intake with meadow vegetation (hereinafter - fodder) and water at selected sites in the area of tunnel #176 under grazing conditions;

- under different timing of intake with fodder in stable keeping in the area of tunnel #177;
- under different timing of intake with water in stable keeping in the area of tunnel #177.

1.3.1. Pastures management

In 2007, grazing was carried out on radiation-contaminated areas in vicinity of the tunnel #176, for 20, 30, 60, 70, 130 and 150 days (Group B. 1.). During each period one animal was pastured. The selected area for animals grazing was divided into eight sections with size of 20×20 m in the central part of which watercourse runs from the tunnel #176 with width of ~ 0.5 m. The sections were outlined after the preliminary geo-botanical description and dosimetric measurements (the flux density of β-radiation, dose rate of γ-radiation on the soil surface and at a height of 1 m). Animals were grazed from 7.00 am to 21.00 pm with unlimited access to tunnel water.

1.3.2. Stabling

In 2008, studies were conducted under conditions of stabling around the tunnel # 177. Animals were placed in a yard (4×4 m) with ground run (10×10 m) to ensure full exercise. Animals were divided into two groups according to the conditions of radionuclides intake into the body with tunnel water and contaminated fodder. The first group (group B.2.) was fed radioactively contaminated fodder and clean water; the animals of the second group (group B.3.) were fed with "clean" fodder (steppe forbs) and radioactively-contaminated water. Period of experiments for two groups - 7, 14, 28, 56 and 112 days. We used meadow vegetation of contaminated areas for feeding. Radioactively-contaminated water for drinking was selected from a watercourse at the portal.

In the formulation of experimental work with animals we used crude procedures for scientific experiments based on the principles of group-periods and pair-analogues [52]. To assess the overall physical condition of the animals their weekly clinical examination was performed. No feeding with concentrated fodder and other feeds was carried out.

According to the experiment procedure we carried out butchering of two control and experimental sixteen animals with sampling the organs (heart, liver, lung, kidney, testis, spleen), blood and muscle and bone tissues.

2. RESULTS AND DISCUSSION

2.1. Study of the nature of radionuclides intake into animals at radioactively-contaminated areas

2.1.1. Assessment of radionuclide content in the diet of animals grazed in the area of the tunnel #176

2.1.1.1. Assessment of radionuclide content in the plants, selected from the grazing site.

To assess the radionuclide content in the feed we took mixed samples of plants (three replications) from each grazing site. Each mixed sample consisted of 25 spot samples (Figure

2). For each site we calculated the average values of radionuclide content per kg of fodder (Table 2).

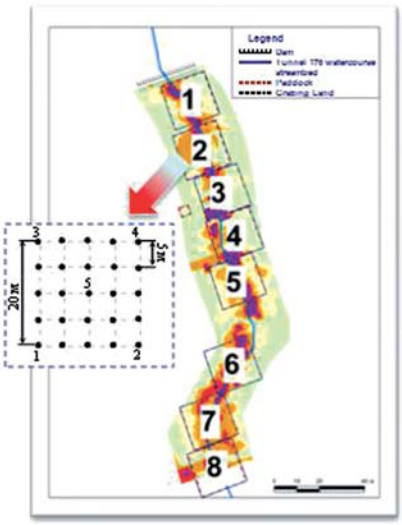

Figure 2. The grid for taking of the integral vegetation sample at pasturing sites (central points denote the animal tether point)

Table 2.

Average values of radionuclide content in mixed samples of plants taken from of grazing sites

Site #	Specific activity, Bq/kg	
	^{137}Cs	^{90}Sr
1	9.5×10^{4}	6.3×10^{4}
2	6.1×10^{4}	6.7×10^{4}
3	2.2×10^{4}	9.1×10^{4}
4	4.2×10^{4}	8.7×10^{4}
5	2.0×10^{4}	2.6×10^{4}
6	1.9×10^{4}	1.2×10^{4}
7	2.2×10^{4}	1.4×10^{4}
8	6.6×10^{3}	1.5×10^{4}
Acceptable level according to [53]	7.4×10^{1}	1.1×10^{2}
Note: error of the average value does not exceed 30%		

We see from this table that the plants selected from areas of grazing are heterogeneous in content of radionuclides.

2.1.1.2. Assessment of radionuclide content in tunnel water

To assess the radionuclide content of water intended for watering the animals throughout the experiment water samples were taken. Content of ^{137}Cs in water samples did not exceed 72 Bq/kg. Only once the value reached 175 Bq/kg. Content of ^{90}Sr varies within

105-189 Bq/kg. Values of T specific activity are within one order and vary within 1×10^5–4.5×10^5 Bq/kg. Level of water contamination intended for watering the experimental animals significantly exceeds the level of intervention for water drinking [54], which is set by NRB-99 and is: for ^{137}Cs - 11Bq/kg, ^{90}Sr - 5 Bq/kg, T – 7.7 kBq/kg.

2.1.1.3. Assessment of radionuclides content in air sampled from animals grazing site

Studies conducted earlier in the area of tunnel #176 [55] found that content of ^{137}Cs and ^{90}Sr is insignificant in the air. Average value of concentration of ^{137}Cs and ^{90}Sr is <0.01 Bq/m^3 and <0.002 Bq/m^3, respectively.

To monitor the intake of T with air in animal body of the experimental group we sampled the air vapour in the area of pastures and in the area of paddocks. Based on the fact that the lung ventilation of the tested animals is 12-13 l/min [56], we calculated intake of T in animal bodies of the experimental group during the entire study period. In the calculation it was taken into account that animal spent an average 15 hours a day in the pasture and 9 hours in the paddock. Average value of T concentration in the air in the grazing and paddock areas were 390 (24 measurements) and 110 (12 measurements) Bq/m^3 respectively.

2.1.1.4. Assessment of daily intakes of radionuclides by animals grazed in the area of the tunnel #176

Each experimental animal drank an average 1.0 ± 0.2 liter of water per day. According to the norms of feeding farm animals [57], animals of this age and weight consume an average 1.5 ± 0.3 kg of fodder. On this basis, we calculated the intake of radionuclides from food and water in the body of each animal over the entire period and calculated the average concentrations of daily intake. It should be noted that in animals kept on pastures, contribution of radionuclides entering the body composed of particles of contaminated soil was not taken into account. Results of determination of daily intake are presented in the table (Table 3). The results showed that the fodder takes main role in the intake of ^{137}Cs and ^{90}Sr in animals. Water takes the main role in intake of T.

Table 3.

Average daily intake of radionuclides by animals grazing on the radioactively contaminated area around the tunnel #176, with unlimited access to tunnel water

Animal code	Number of days	^{90}Sr, Bq			^{137}Cs, Bq			T, Bq			
		with fodder	with water	Total	with fodder	with water	Total	with fodder	with water	with air	Total
B.1.1.	20	2.4×10^4	1.5×10^2	2.4×10^4	3.2×10^4	4.9×10^1	3.2×10^4	2.1×10^5	3.4×10^5	5.3×10^3	5.6×10^5
B.1.2.	31	4.7×10^4	1.5×10^2	4.7×10^4	1.3×10^5	4.9×10^1	1.3×10^5	2.1×10^5	3.4×10^5	5.3×10^3	5.6×10^5
B.1.3.	62	5.7×10^4	1.5×10^2	5.7×10^4	1.8×10^5	4.9×10^1	1.8×10^5	2.1×10^5	3.4×10^5	5.3×10^3	5.6×10^5
B.1.4.	68	6.8×10^4	1.5×10^2	6.8×10^4	1.1×10^5	4.9×10^1	1.1×10^5	2.1×10^5	3.4×10^5	5.3×10^3	5.6×10^5
B.1.5.	130	5.5×10^4	1.5×10^2	5.5×10^4	6.9×10^4	4.9×10^1	6.9×10^4	2.1×10^5	3.4×10^5	5.3×10^3	5.6×10^5
B.1.6.	151	1.1×10^5	1.5×10^2	1.1×10^5	1.1×10^5	4.9×10^1	1.1×10^5	2.1×10^5	3.4×10^5	5.3×10^3	5.6×10^5

Contribution of air, water and fodder into the daily intake of radionuclides by sheep is shown in the figure (Figure 3).

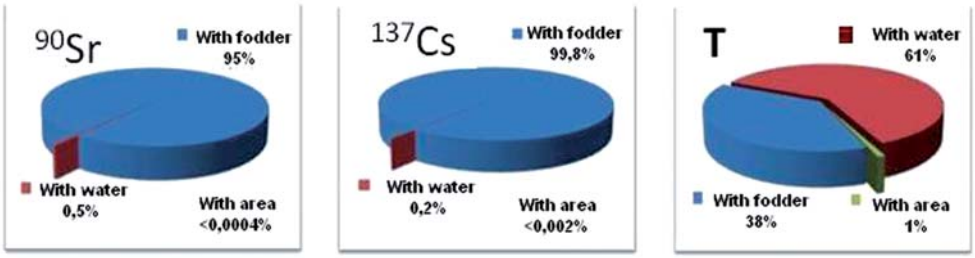

Figure 3. Contribution of air, water and food in daily intake of ^{137}Cs, ^{90}Sr and T

2.1.2. Assessment of radionuclide content in feed and water for animals kept in the area of tunnel #177

2.1.2.1. Assessment of radionuclide content in the plant fodder

To assess the radionuclide concentrations in the plant fodder for each week every day 300 g of vegetation was taken from the daily mass of radioactively-contaminated feed, from which 2.1 kg of vegetation sample was formed (300 g × 7 days) for radionuclide analysis. In summary, the results were obtained on the content of radionuclides in the plant feed for each weekly period during the experiment. Concentrations of ^{137}Cs in the plant feed for the entire period of the experiment varied in the range of <11–305 Bq/kg, only last week a value reached 5.8 × 10^3 Bq/kg, ^{90}Sr within 4.8×10^4 – 1.1×10^5 Bq/kg, T - in the range of 1.3 × 10^5-2.6 × 10^5 Bq/l, $^{239+240}$Pu in the range of 1.2-47 Bq/kg, ^{241}Am within <1.2-25 Bq/kg.

2.1.2.2. Evaluation of radionuclides in organism of animals with plant food

Daily, during the experiment we registered amount of plant feed contaminated with radionuclides, eaten by each experimental animal of group B.2. Based on these data and data on radionuclide concentrations in vegetable feed there was calculated the average daily intake of radionuclides from the plant feed into the body of each experimental animal of this group. The calculation was performed taking into account the dynamics of radionuclide content in water. Calculation of average daily intakes of radionuclides with vegetable food into the body of experimental animals is presented in the table (Table 4).

Table 4.

Average daily intakes of radionuclides with vegetable food into the body of experimental animals of the Group B.2.

Animal code	Number of days	Average daily intakes, Bq				
		^{137}Cs	^{90}Sr	T	$^{239+240}$Pu	^{241}Am
B.2.1.	7	3.7×10^2	7.8×10^4	1.9×10^5	4.7×10^1	1.2×10^1
B.2.2.	14	3.0×10^2	1.0×10^5	2.3×10^5	7.8	1.4×10^1
B.2.3.	28	1.0×10^2	1.2×10^5	3.1×10^5	9.7	2.0×10^1
B.2.4.	56	4.2×10^2	2.0×10^5	4.4×10^5	5.8	2.8×10^1
B.2.5.	112	1.6×10^3	1.7×10^5	3.7×10^5	1.1×10^1	2.4×10^1

2.1.2.3. Assessment of radionuclide content in water for animals

To assess the radionuclide concentrations in water on a weekly basis water samples were taken. Values of ^{137}Cs concentrations in the water during the entire period of the experiment ranged within <0.18-28 Bq/l, ^{90}Sr - within $3.7\times102 - 4.3\times10^3$ Bq/l, T - within $300\times10^3 - 310\times10^3$ Bq/l, $^{239+240}Pu$ - within <0.06–4.3 Bq/l, ^{241}Am - within <0.06–1.2 Bq/l.

2.1.2.4. Assessment of radionuclides intake by animals with water

During the experiment we recorded daily amount of water drunk by each experimental animal of the group B.3. Based on these data and data on radionuclide concentrations in water intended for watering, there was calculated the average daily intake of radionuclides into the body of each experimental animal of this group. The calculation was performed taking into account the dynamics of radionuclide content in water. Calculated average daily intakes of radionuclides from water for experimental animals of the group are presented in the table (Table 5).

Table 5.

Average daily intakes of radionuclides with water into the body of experimental animals of the Group B.3.

Animal code	Number of days	Average daily intakes, Bq				
		^{137}Cs	^{90}Sr	T	$^{239+240}Pu$	^{241}Am
B.3.1.	7	1.1×10^2	3.2×10^3	1.2×10^6	1.7×10^1	4.8
B.3.2.	14	9.9×10^1	4.2×10^3	1.6×10^6	1.0×10^1	8.3
B.3.3.	28	6.5	7.5×10^3	1.4×10^6	6.9×10^{-1}	4.4
B.3.4.	56	3.6	5.5×10^3	1.2×10^6	5.4×10^{-1}	2.4
B.3.5.	112	2.3×10^1	5.1×10^3	1.3×10^6	3.0	2.6

2.2. Investigation of the laws of transfer and distribution of artificial radionuclides in the organs and tissues of sheep

According to the study procedure, animals after being taken out from the experiment were left for 12 hours without food and water and then sacrificed by exsanguinations method (a total of 18 sheep).

To assess the content of radionuclides ^{90}Sr, ^{137}Cs, T, $^{239+240}Pu$, ^{241}Am in biological samples we determined specific (Bq/kg) and absolute (Bq) activities of organs and tissues. Absolute activity was determined on the basis of specific activity of both mass of organs and tissues. In determining the weight of muscle and bone tissue we based on the calculation of slaughter yield of 45%. Yield of pure meat was 60%, bone tissue - 30% of slaughter yield [58].

As a parameter for radionuclides transfer level from the environment into animal products, we used transfer coefficient K_n, which is by definition of the IAEA is calculated as the ratio of specific activity of an organ or tissue (Bq/kg) to the total amount of radionuclides intaken during the day in the animal organism (Bq/d):

$$K_п = \frac{A_п}{A_р} \quad (2), \text{ where}$$

$A_п$ - concentration of a radionuclide in an organ or tissue (Bq/kg);
A_p - amount of total daily intake of a radionuclide, Bq/day.

2.2.1. Peculiarities of ^{90}Sr transfer in the body of sheep

2.2.1.1. The content of ^{90}Sr in the bodies of experimental animals

The following table (Table 6) shows the specific activity of ^{90}Sr in tissues and organs of sheep from the Group B.1., in the bodies of which ^{90}Sr came with food and water. Specific activity of ^{90}Sr in tissues and organs of sheep from Group B.2. and B.3. in the bodies of which ^{90}Sr came with food and water, respectively, is presented in tables (Table 7, Table 8).

Table 6.

Specific activity of ^{90}Sr in tissues and organs of sheep at different periods of grazing at radioactively contaminated sites, Bq/kg

Organs and tissues	Number of days for animal husbandry, days					
	20	31	62	68	130	151
	Average daily intake with food and water, Bq/kg					
	2.4×10^4	4.7×10^4	5.7×10^4	6.8×10^4	5.5×10^4	1.1×10^5
	Specific activity, Bq/kg					
Heart	4.9±2.1	19±2.2	22.1±3.5	32.2±1.9	22.6±2.4	20.1±4.1
Kidney	10.6±1.7	31.4±2.8	47.7±8.2	18.5±2.1	24.8±23	30.2±2.9
Lung	11.3±1.6	59.9±4.3	28.7±6.3	31.3±2.3	37.8±2.8	29.3±3.6
Liver	5.8±1.4	20.3±4.7	13.9±3.7	16.1±2.2	18.6±2.7	15.7±1.8
Muscular tissue	6.0±1.7	11.1±1.7	14±5.3	16.9±2.9	11.4±1.8	22.4±4.1
Bone tissue	2000±87	11900±63	16100±57	35600±84	18100±58	51400±303
Wool	915 ± 18	284 ± 13	–	294 ± 8	–	488 ± 12

Table 7.

Specific activity of ^{90}Sr in tissues and organs of sheep at different periods of its intake with food, Bq/kg

Organs and tissues	Number of days for animal husbandry, days				
	7	14	28	56	112
	Average daily intake with food and water, Bq/kg				
	7.8×10^4	9.9×10^4	1.2×10^5	2.0×10^5	1.7×10^5
	Specific activity, Bq/kg				
Heart	12.0 ± 3.3	17.2 ± 3.5	33.6 ± 2.9	75.1±2.4	29.7±2.3
Kidney	30.6 ± 2.7	24.7 ± 3.8	66.1 ± 3.2	140.6±3.0	59.9±3.6
Lung	22.4 ± 2.5	31.9 ± 3.3	118.9 ± 4.1	160.6±3.5	52.0±2.6
Liver	13.0 ± 2.4	8.2 ± 4.0	44.6 ± 3.0	72.8±2.4	32.3±2.8

Organs and tissues	Number of days for animal husbandry, days				
	7	14	28	56	112
	Average daily intake with food and water, Bq/kg				
	7.8×10^4	9.9×10^4	1.2×10^5	2.0×10^5	1.7×10^5
	Specific activity, Bq/kg				
Muscular tissue	7.9 ± 0.1	10.9 ± 0.2	32.7 ± 0.5	47.2±2.8	29.4±2.2
Bone tissue	2200 ± 52	4600 ± 79	3600 ± 72	19300±149	18300±160
Testes	21.8 ± 3.0	27.7 ± 3.3	54.7 ± 3.2	233.5±4.0	48.8±4.1
Spleen	12.7 ± 2.7	25.2 ± 3.9	39.7 ± 3.8	82.2±2.1	25.9±2.3
Wool	193 ± 7	81.9 ± 5.8	691 ± 16	2520 ± 21	852 ± 13

Table 8.

Specific activity of ^{90}Sr in tissues and organs of sheep at different periods of its intake with water, Bq/kg

Organs and tissues	Number of days for animal husbandry, days				
	7	14	28	56	112
	Average daily intake with food and water, Bq/kg				
	3.2×10^3	4.2×10^3	7.5×10^3	5.5×10^3	5.1×10^3
	Specific activity, Bq/kg				
Heart	< 5.7	< 5.0	< 3.0	5.4±1.5	14.8±3.6
Kidney	< 3.7	< 4.3	6.7 ± 1.8	9.6±2.6	19.1±3.0
Lung	< 5.0	< 5.0	8.5 ± 1.5	7.0±2.1	18.1±1.9
Liver	< 3.8	< 4.6	3.3 ± 1.7	<3.4	6.8±1.9
Muscular tissue	1.5 ± 0.1	2.1 ± 0.1	< 2.4	<3.9	<5.8
Bone tissue	600 ± 55	700 ± 42	1300 ± 47	2200±100	7900±94
Testes	< 4.8	< 5.0	13.0 ± 3.8	8.2±1.4	10.8±1.9
Spleen	< 5.3	< 6.7	5.1 ± 2.1	10.2±2.0	22.9±3.9
Wool	162 ± 7	91.4 ± 6.4	192 ± 7	584 ± 19	928 ± 19

2.2.1.2. Distribution of ^{90}Sr in the body of sheep

As expected, the research showed that major concentrations of ^{90}Sr were found in bone tissue. It should be noted that wool is also the main depot for ^{90}Sr, as well as bone tissue. Activities of other organs in relation to bone tissues were 2-3 orders of magnitude smaller.

Analysis allows us to say that the distribution of ^{90}Sr specific activity of soft organs has a slightly noticeable peculiarity: highest concentrations are found in the lungs, further decreases as follows: kidney ≥testis ≥ liver ≥ spleen ≥ heart ≥ muscular tissue. A similar distribution pattern of ^{90}Sr specific activity in tissues and organs occurs in the majority of experimental animals and does not depend on the type and timing of the radionuclide intake.

At long-term husbandry in the radioactively contaminated territories of STS, in the organs (lungs, liver, kidney, heart, testis, spleen), based on their weight and the weight of the muscular tissue of sheep carcasses, less than ~ 0.004% of the total amount of ^{90}Sr received in a day detained.

It was determined that regardless the time and type of intake, distribution of ^{90}Sr in the bodies with relation to specific activity in the wool tissue, has a certain relationship that allows predicting of ^{90}Sr in the soft organs by the specific activity in wool. Relative concentrations of ^{90}Sr in the organs and wool tissue are presented in the table (Table 9) (specific activity of wool is taken as a unit). There is a relationship between the distribution of bone tissue and wool of sheep. However, talking about a more exact relationship between the content of ^{90}Sr in bone and wool is much more difficult. Content of ^{90}Sr in bone tissue may be 5, sometimes 70 times greater than that in the wool.

Similar assessment of ^{90}Sr in the organs can also be done using other organs (e.g, liver). If we take the specific activity of liver as a unit, the average relative concentrations of ^{90}Sr in the heart, kidneys, lungs, muscle, testis and spleen of sheep will have a value of 1.3±0.13; 2.1±0.14; 2.2±0.10; 0.9±0.07; 2.4±0.37; 1.7±0.33, respectively. There is no relationship of ^{90}Sr distribution between bone tissue and liver.

Table 9.

Ratio of specific activity of ^{90}Sr in organs to the specific activity of ^{90}Sr in wool

Organs and tissues	Average	min	Max	Q-ty of values
Heart	0.05	0.01	0.21	11
Kidneys	0.05	0.02	0.10	10
Lung	0.07	0.01	0.17	11
Liver	0.04	0.01	0.07	10
Muscular tissue	0.03	0.01	0.05	10
Testes	0.06	0.01	0.11	7
Spleen	0.04	0.02	0.07	7
Bone tissue	21	4	73	13

2.2.1.3. Peculiarities of ^{90}Sr transfer in various conditions and periods

The figure (Figure 4) shows the transfer coefficients of ^{90}Sr in the muscular tissue of sheep from Groups B.1. and B.2. that were fed with plants from different parts of "Degelen" site. It is seen from the graphs that the balance is achieved immediately after intake, the process of accumulation is absent, dispersion of K_n is low. Similar dynamics are also set for the lungs, spleen and testes.

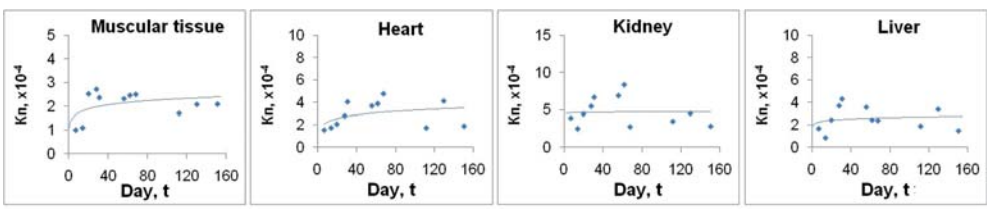

Figure 4. ^{90}Sr transfer dynamics into organs and tissues of sheep (Groups B.1. and B.2.)

Despite the fact that we did not determine the dynamics of change in K_n of ^{90}Sr in "soft" organs of animals, since equilibrium is reached in 7 days, we noted a direct relation of its content in the organs to the average daily intake (Figure 5).

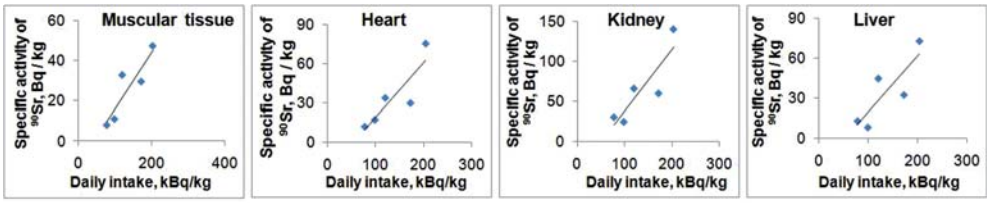

Figure 5. Ratio of ^{90}Sr specific activity in organs to its daily intake with contaminated food (Group B.2.)

One of the main objectives of the study was to determine the possible differences of radionuclides transfer in the organs and tissues of animals from Group B.1. and B.2. depending on the income of vegetable food with different radioactively contaminated sites (areas of the tunnel #176 and #177). Although animals from Groups B.1. had contaminated water besides the contaminated food in the diet, we believe that this factor had no significant effect, as the fraction of polluted water in the diet was less than 0,1% of average daily intake. This allows us to compare the obtained values of ^{90}Sr transfer in tissues and organs of sheep from Group B.1. and B.2.

The results of data analysis showed that the transfer of ^{90}Sr in the organs and tissues of animals kept at various tunnels, has no significant difference - the values of K_n in equilibrium are identical or similar to the overall average. Comparing the values of ^{90}Sr K_n to organs of sheep with food at various radioactively contaminated sites are presented in the table (Table 10).

Table 10.

Average values of ^{90}Sr K_n in organs and tissues of sheep when intaken with food at different sites

Organs and tissues	Average values of K_n in equilibrium, $\times 10^{-4}$		Average
	when intaken with food and water (Group B.1.)	when intaken with food (Group B.2.)	
Heart	3.5	2.3	2.9
Kidneys	4.9	4.4	4.7
Lung	6.1	5.4	5.8
Liver	2.7	2.3	2.5
Muscular tissue	2.3	1.8	2.1
Testes	–	4.9	4.9
Spleen	–	2.6	2.6
Note: error of the average value does not exceed 30%			

Distribution of ^{90}Sr in tissues and organs of sheep after 112 days of intake with food and water is presented in the table (Table 11). The sum of ^{90}Sr specific activity intaken by

"soft" organs of the animals from Group B.2. is ~ 0.16%, and as for the animal of Group B.3. is ~ 1.8%. This shows that ^{90}Sr transfers much more with water.

Table 11.

Distribution of ^{90}Sr in tissues and organs of sheep after 112 days of intake with food and water, expressed as a percentage of daily intake

Organs and tissues	Amount of ^{90}Sr in organs and tissues, %	
	when intaken with food (Group B.2.)	when intaken with water (Group B.3.)
Heart	0.017	0.289
Kidneys	0.035	0.372
Lung	0.030	0.353
Liver	0.019	0.133
Muscular tissue	0.017	– *
Testes	0.028	0.211
Spleen	0.015	0.446
Total specific activity	~ 0.16	**~ 1.80**

Note: measurement error of ^{90}Sr is no more than 20%
* - sample activity was below the detection limit

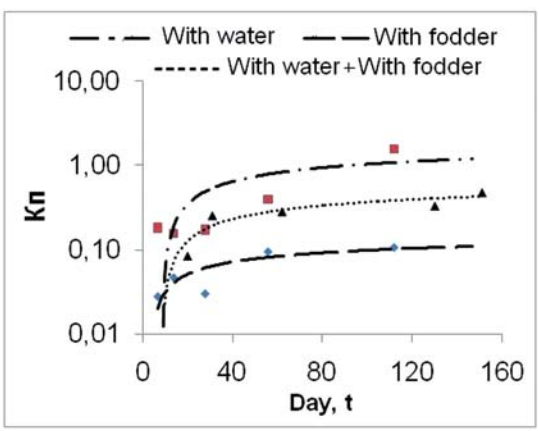

Figure 6. Transfer coefficient of ^{90}Sr in bone tissue of sheep at various conditions and periods of intake

Bioavailability of ^{90}Sr for bone tissue of sheep intaken with tunnel water is approximately one order of magnitude higher than ^{90}Sr, intaken with food. When ^{90}Sr inflows into the body of animals with water and food together, K_n of ^{90}Sr into bone tissue is close to the average individual values (Figure 6). It is also noteworthy that no balanced state of ^{90}Sr in the bone tissue of sheep has been found according to the conditions of the time intervals corresponding to periods of grazing season in these areas.

2.2.1.4. Calculation of allowable concentration of ^{90}Sr in the daily diet

It should be noted that husbandry at the most contaminated with ^{90}Sr areas at the former STS, even with prolonged daily intake into the body of sheep (up to 7×10^4 Bq/d), does not result in excess concentrations of radionuclide in muscular tissue with respect to sanitary and hygienic standards (50 Bq/kg) [59].

2.2.2. Peculiarities of ^{137}Cs transfer into the body of sheep

2.2.2.1. Content of ^{137}Cs in the body of experimental animals

The table below (Table 12) shows the specific activity of ^{137}Cs in organs and tissues of sheep from Group B.1., in the bodies of which ^{137}Cs came with food and water. Specific activity of ^{137}Cs in organs and tissues of sheep from Group B.2. and B.3. in the bodies of which ^{137}Cs came with food and water, respectively, is presented in tables (Table 13, Table 14).

Table 12.

Specific activity of ^{137}Cs in organs and tissues of sheep grazing at different periods at the radioactively-contaminated sites, Bq/kg

Organs and tissues	Number of husbandry days, days					
	20	31	62	68	130	151
	Average daily intake with food and water, Bq/kg					
	3.2×10^4	1.3×10^5	1.8×10^5	1.1×10^5	6.9×10^4	1.1×10^5
	Specific activity, Bq/kg					
Heart	4,600±100	5,300±100	1,100±110	3,800±200	5,000±220	1,800±140
Kidney	8,400±300	8,600±280	2,000±120	7,600±350	6,600±250	4,100±230
Lung	3,700±190	4,600±200	1,000±40	3,200±210	4,300±220	1,400±120
Liver	4,040±190	5,200±230	1,000±100	3,500±200	5,000±230	1,300±120
Muscular tissue .	7,600 ± 290	15,500±160	4,400±210	9,400±300	9,800±130	2,000±140
Bone tissue	4,700±290	9,100±330	1,200±130	8,100±740	2,500±200	2,500±250
Wool	14 ± 1	–	–	26 ± 1	70 ± 10	190 ± 2

Table 13.

Specific activity of ^{137}Cs in organs and tissues of sheep at different timing of its intake with food, Bq/kg

Organs and tissues	Number of husbandry days, days				
	7	14	28	56	112
	Average daily intake with food, Bq/kg				
	3.66×10^2	2.94×10^2	1.02×10^2	4.17×10^2	1.60×10^3
	Specific activity, Bq/kg				
Heart	4.2 ± 1.0	10.2 ± 1.3	14.4 ± 2.0	11.6±1.7	326.7±6.3
Kidney	8.2 ± 1.4	18.2 ± 2.1	14.9 ± 2.0	31.2±3.4	562.3±11.0
Lung	2.1 ± 0.8	6.9 ± 1.1	6.2 ± 1.2	8.4±1.2	191.5±4.5
Liver	4.6 ± 1.1	9.0 ± 1.1	15.2 ± 1.2	12.4±1.3	352.7±5.8

Organs and tissues	Number of husbandry days, days				
	7	14	28	56	112
	Average daily intake with food, Bq/kg				
	3.66×10^2	2.94×10^2	1.02×10^2	4.17×10^2	1.60×10^3
	Specific activity, Bq/kg				
Muscular tissue .	3.71 ± 0.16	6.40 ± 0.21	8.63 ± 0.23	39.7±2.0	220.3±4.8
Bone tissue	3.68 ± 0.57	28.4 ± 1.3	12.0 ± 2.0	11.76±0.58	116.6±4.1
Testes	4.1 ± 1.1	5.7 ± 1.1	7.1 ± 1.1	11.9±1.9	301.3±5.6
Spleen	9.0 ± 2.1	5.1 ± 2.2	7.1 ± 2.0	13.5±2.9	307.1±5.6
Wool	2.77 ± 0.19	< 0.19	< 0.48	0.68 ± 0.13	< 0.45

Table 14.

Specific activity of ^{137}Cs in organs and tissues of sheep at different timing of its intake with water, Bq/kg

Organs and tissues	Number of husbandry days, days				
	7	14	28	56	112
	Average daily intake with water, Bq/kg				
	1.11×10^2	9.90×10^1	6.45	3.56	2.32×10^1
	Specific activity, Bq/kg				
Heart	7.3 ± 1.1	2.7 ± 1.1	< 1.5	<1.6	10.3±1.5
Kidney	10.3 ± 2.4	6.2 ± 1.4	< 1.8	<2.0	20.7±2.5
Lung	6.6 ± 1.2	3.9 ± 1.0	< 1.1	<1.1	5.0±0.9
Liver	6.1 ± 1.0	3.4 ± 0.9	< 1.1	<1.0	6.6±1.1
Muscular tissue .	6.29 ± 0.18	4.88 ± 0.16	0.71± 0.09	<0.9	3.9±0.9
Bone tissue	2.78 ± 0.49	29.2 ± 2.3	< 1.1	<1.7	<2.8
Testes	7.5 ± 1.2	1.9 ± 0.9	< 0.9	<0.9	5.5±1.0
Spleen	6.2 ± 2.0	< 4.0	< 2.0	<2.6	<3.8
Wool	1.26 ± 0.32	< 0.24	< 1.02	< 0.41	0.12±0.05

2.2.2.2. Distribution of ^{137}Cs in the body of sheep

Distribution of ^{137}Cs specific activity between organs and tissues varies depending on the periods of grazing the animals in the radioactively contaminated territories. When there is a small amount of ^{137}Cs intake (up to 400 Bq per day), the radionuclide is uniformly distributed in organs and tissues. However, if the concentration increases, the distribution of ^{137}Cs in organs becomes uneven. So, when animals are grazed 30 days, the highest activity ^{137}Cs is in the kidneys, but when grazed for 60 days or more, the highest specific activity of ^{137}Cs is in the muscular tissue.

At long-term intake the specific activity of ^{137}Cs in different parts of virtually all animals decreases as follows: muscle> kidney> bone ≥ heart> liver ≥ lungs> testis> spleen. Submitted distribution pattern of ^{137}Cs in organs and tissues of sheep is similar to data obtained in the zone of Chernobyl accident [60].

Studies carried out under the leadership of V.P. Burov [61] say that the concentration of ^{137}Cs in the bone tissue is approximately 5-10 times lower than that in the muscular tissue. In our study no significant difference of ^{137}Cs specific activity of in bone and muscular tissues was found. Concentration of ^{137}Cs in the bone tissue is approximately 2 times lower than in the muscular tissue, sometimes the values are almost identical.

It was determined that regardless of the time and type of intake, distribution of ^{137}Cs in organs with relation to specific activity in wool has poor pattern. Ratio of ^{137}Cs specific activity in organs to the specific activity in wool is presented in the table (Table 15).

Table 15.

Ratio of ^{137}Cs specific activity in organs to ^{137}Cs specific activity in wool

Organs and tissues	Average	min	max	Q-ty of values
Heart	75	16	190	5
Kidney	130	29	290	5
Lung	60	12	160	5
Liver	80	15	190	5
Muscular tissue	140	50	380	5
Bone tissue	70	17	180	5

Regardless of the time and type of intake, distribution of ^{137}Cs in organs and tissues has a certain interdependence that enables the prediction of this radionuclide in one organ by the content in another. For example, if we take the specific activity of liver as a unit, average relative concentration of ^{137}Cs in the heart, kidneys, lungs, bones, testes and spleen of sheep will be 1.08±0.05; 1.99±0.16; 0.82±0.06; 1.28±0.25; 0.80±0.09; 0.99±0.22, respectively. No dependence of ^{137}Cs distribution between the muscle and liver has been revealed.

2.2.2.3. Peculiarities of ^{137}Cs transfer under different conditions and periods of intake

The figure below (Figure 7) shows K_n of ^{137}Cs into muscular tissue, heart, kidney and liver of sheep of Groups B.1. and B.2. that were fed with plants from different tunnels of "Degelen" site. The experiments showed that under conditions of prolonged daily intake ^{137}Cs into the body of sheep with different components of the environment, the concentration of ^{137}Cs in organs increases to a certain value. In case of prolonged intake (20 days or more), despite the ongoing inflow of the isotope, we observed a dynamic balance between accumulation and removal of it; as a result of such balance no further increase in ^{137}Cs in organs occurs. A similar dynamics of changes in K_n with time is also determined for lungs, spleen and testes.

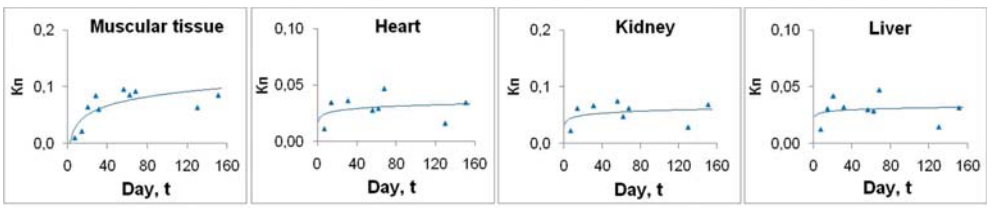

Figure 7. ^{137}Cs transition dynamics to organs and tissues of sheep (Groups B.1. and B.2.)

There are no obvious differences in K_n of ^{137}Cs into the muscular tissue of sheep when the animals are bred near tunnels #176 and #177. Some differences of K_n are explained by tolerable error when intake is being registered. Although animals from Groups B.1. had in their diet contaminated water besides contaminated food and, we believe that this factor had no significant effect, as the fraction of contaminated water in the diet was less than 0.01% of average daily intake. This allows comparing the obtained values of ^{137}Cs transfer in organs and tissues of sheep from Groups B.1. and B.2.

Comparisons of the values of ^{137}Cs K_n in organs of sheep when it comes with food at different radioactively contaminated sites are presented in the table (Table 16). It should be noted that, despite significant differences in daily intake the concentration of ^{137}Cs in the body of animals (5×10^4 times) when husbandry at various tunnels, K_n turned to be almost the same or similar.

Table 16.

Average values of ^{137}Cs K_n in organs and tissues of sheep when taken with feed at different sites

Organs and tissues	Average values of K_n at balanced state, $\times10^{-2}$		
	when intaken with food and water (Group B.1.)	when intaken with food (Group B.2.)	Average
Heart	3.7	6.8	5.2
Kidneys	6.7	9.4	8.1
Lung	3.0	3.5	3.3
Liver	3.3	7.0	5.1
Muscular tissue	7.5	6.7	7.1
Bone tissue	4.7	8.1	6.4
Testes	–	3.9	3.9
Spleen	–	4.0	4.0
Note: error of the average value does not exceed 30%			

Results presented in the table (Table 17) show that the transfer of ^{137}Cs into organs of sheep when intaken with water can be up to two times more than when intaken with food.

Table 17.

Distribution of ^{137}Cs in tissues and organs of sheep when intake with food and water, expressed as a percentage of daily intake

Organs and tissues	Amount of ^{137}Cs in organs and tissues*, %	
	when intaken with food (Group B.2.)	when intaken with water (Group B.3.)
Heart	8.3	17.9
Kidneys	14.5	34.9
Lung	4.9	10.5
Liver	8.8	12.5
Muscular tissue	5.6	9.1

Organs and tissues	Amount of ^{137}Cs in organs and tissues*, %	
	when intaken with food (Group B.2.)	when intaken with water (Group B.3.)
Bone tissue	6.0	17.9
Testes	7.3	10.8
Spleen	7.8	5.6
Total specific activity	~63	**~119**
Note: measurement error of ^{137}Cs is no more 30% * - average of 3 animals		

2.2.2.4. Calculation of allowable concentration of ^{137}Cs in the daily diet.

Data obtained on the dynamics of ^{137}Cs concentration in organs of sheep when kept at the radioactively contaminated territories of STS allow regulating the allowable levels of feeding with radioactive food that provides livestock, corresponding to sanitary rules and norms.

To assess the allowable level of ^{137}Cs content in the daily diet of sheep we can take requirements for foods according to which the specific activity of ^{137}Cs in muscular tissue should not exceed 160 Bq/kg. Based on the fact that the maximum K_n value of ^{137}Cs in muscular tissue equals to – 0.17, then the allowable level of ^{137}Cs in the daily diet of sheep will be about ~900 Bq.

2.2.3. Peculiarities of T transfer into the body of sheep

2.2.3.1. T content in the body of experimental animals.

The table below (Table 18) shows the specific activity of T in tissues and organs of sheep from Group B.1., into the bodies of which T came with food and water. Specific activity of T in tissues and organs of sheep from Group B.2. and B3, into the bodies of which T came with food and water, respectively, is presented in tables (Table 19, Table 20).

Table 18.

Specific activity of T in the free water of sheep at different periods of pasture at radioactively contaminated areas

Organs and tissues	Number of husbandry days, days					
	20	31	62	68	130	151
	Average daily intake with food and water, Bq/kg					
	5.6×10^5	5.6×10^5	5.6×10^5	5.6×10^5	5.6×10^5	5.6×10^5
	Specific activity, Bq/kg					
Heart	111 ± 10	200 ± 20	155 ± 14	135 ± 11	148 ± 13	92 ± 10
Kidney	131 ± 11	130 ± 11	153 ± 14	168 ± 15	136 ± 12	108 ± 10
Lung	102 ± 10	146 ± 12	173 ± 15	164 ± 15	176 ± 16	102 ± 10
Liver	116 ± 10	141 ± 12	179 ± 17	145 ± 15	173 ± 16	96 ± 10
Muscular tissue	114 ± 10	139 ± 12	189 ± 17	131 ± 12	164 ± 16	146 ± 13
Bone tissue	62 ± 6.0	28 ± 3.0	33 ± 3.0	33 ± 3.0	28 ± 3.0	27 ± 3.0

Table 19.

Specific activity of T in the free water of sheep organs at different periods of intake with food

Organs and tissues	Number of husbandry days, days				
	7	14	28	56	112
	Average daily intake with food, Bq				
	1.90×10^5	2.25×10^5	3.10×10^5	4.36×10^5	3.72×10^5
	Specific activity, kBq/kg				
Heart	67 ± 6.2	26 ± 0.8	43 ± 4.0	50 ± 4.1	70 ± 6.3
Kidney	40 ± 4.0	30 ± 1.8	39 ± 3.0	65 ± 5.2	61 ± 5.1
Lung	21 ± 0.8	21 ± 1.7	52 ± 4.1	63 ± 5.2	54 ± 4.1
Liver	16 ± 1.7	24 ± 1.8	52 ± 4.0	59 ± 5.1	80 ± 7.3
Muscular tissue	18 ± 1.7	21 ± 1.7	43 ± 3.0	62 ± 5.2	56 ± 4.0
Bone tissue	5 ± 0.5	11 ± 1.2	9 ± 8.5	6 ± 0.7	7 ± 0.6
Testes	20 ± 2.1	19 ± 1.7	n/d	65 ± 5.1	72 ± 6.0
Spleen	40 ± 4.0	23 ± 1.8	47 ± 3.0	60 ± 5.2	122 ± 10

Table 20.

Specific activity of T in free water of sheep organs at different periods of intake with water

Organs and tissues	Number of husbandry days, days				
	7	14	28	56	112
	Average daily intake with water, Bq/kg				
	1.24×10^6	1.64×10^6	1.41×10^6	1.21×10^6	1.31×10^6
	Specific activity, kBq/kg				
Heart	75 ± 6.0	117 ± 10	144 ± 14	140 ± 17	126 ± 10
Kidney	93 ± 8.0	99 ± 10	162 ± 20	123 ± 11	144 ± 13
Lung	101 ± 10	109 ± 10	163 ± 20	123 ± 11	101 ± 10
Liver	77 ± 6.0	101 ± 10	141 ± 14	120 ± 11	113 ± 10
Muscular tissue	78 ± 7.0	93 ± 8.0	147 ± 14	125 ± 11	152 ± 14
Bone tissue	3 ± 0.4	10 ± 1.0	16 ± 1.0	9 ± 0.8	6 ± 0.7
Testes	89 ± 7.4	106 ± 10	159 ± 20	140 ± 13	138 ± 12
Spleen	77 ± 6.3	n/d	160 ± 20	128 ± 11	66 ± 5.0

2.2.3.2. Distribution of T in the body of sheep

Studies have shown that, if there is a prolonged intake of T with food and water, there is a uniform distribution of it between the organs (lungs, liver, kidney, heart, testis, spleen, muscular tissue) of a sheep. However, distribution of T between the muscle and bone tissue varies and depends on the form of intake. So, when animals have intaken with plant free water, concentration of T in the bone tissue is up to 6 times less than in the muscular tissue, and T comes with water the concentration of T in the bone tissue is 18 times less than in the muscular tissue .

It is obvious that the uniform distribution of T in tissues and organs of sheep allows making a forecast of its content in organs and tissues of sheep with up to 90% of accuracy, knowing only activity of one organ (e.g. liver). Ratio of specific activity of T in tissues and organs to its specific activity in liver is presented in the table (Table 21).

Table 21.

Average value of the ratio of T specific activity in organs and tissues to its specific activity in liver

Organs and tissues	Heart	Kidney	Lung	Muscular tissue	Bone tissue	Testes	Spleen
Avearage value	1.01±0.04	1.05±0.04	1.04±0.04	1.02±0.05	0.20±0.04	1.08±0.05	1.19±0.19

2.2.3.3. Peculiarities of T transfer under different conditions and periods

It was determined that the intake of T into organs with plant free water and water is more intense, and very quickly there comes a dynamic equilibrium between the amount taken T and amount derived. After that, an increase of T in the organs does not occur (Figure 8).

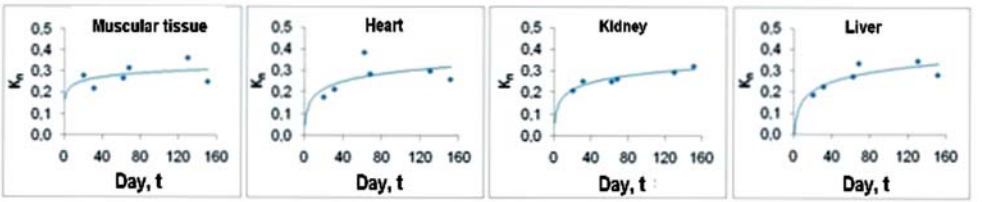

Figure 8. ^{137}Cs transfer dynamics in organs of sheep (Group B.1.)

Of particular interest is transfer of T from plants free water, as the bioavailability of T, coming with plant free water, is higher than T taken water. Similar results were obtained in laboratory animals, which for a long time were given food contaminated with T or triturated water [62]. Perhaps this is due to the fact that the body there is an intensive accumulation of organically bound tritium, intaken in a considerable amount with forage (Table 22).

Table 22.

Distribution of T in organs and tissues of sheep at intake with food and water, expressed as a percentage of daily intake

Organs and tissues	Amount of T in organs and tissues*, %	
	when intake with food* (Group B.2.)	when intake with water* (Group B.3.)
Heart	18.1	8.9
Kidney	15.5	9.2
Lungs	13.2	8.8
Liver	14.1	8.2
Muscular tissue	12.4	8.8

Organs and tissues	Amount of T in organs and tissues*, %	
	when intake with food* (Group B.2.)	when intake with water* (Group B.3.)
Bone tissue	2.8	0.6
Testes	13.2	9.4
Spleen	18.5	8.3
Total specific activity	~108	**~62**
Note: measurement error of T is not more than 20% * - Average values of 5 animals.		

2.2.3.4. Allowable concentrations of the radionuclide in the daily diet

It is known that when tritium water inflows in the animal organism, only a small portion (1%) is converted to organically bound T [63], and therefore we believe that our findings in the study of tritium water intake into the body of sheep can make a forecast of expected concentrations of T, as well as regulate the allowable time of watering with radioactive water. Intake of T with daily diet of sheep should not exceed 4×10^4 Bq, to avoid raising intake limit of T into the human body with the production of sheep (meat, offal). Calculation was performed on the basis of the annual intake limit of inorganically bound T with food for the population [64].

2.2.4. Peculiarities of $^{239+240}$Pu and ^{241}Am into the body of sheep

Content of radionuclides in the bodies of experimental animals. The following table (Table 23) presents the specific activity of $^{239+240}$Pu in organs and tissues of sheep at different periods of its intake with food and water.

Table 23.

Specific activity of $^{239+240}$Pu in organs and tissues of sheep at different periods of its intake with food and water

Organs and tissues	Average daily intake with food, Bq/kg					Average daily intake with water, Bq/kg				
	4.68×10^1	7.79	9.7	5.8	1.0×10^1	1.7×10^1	9.8	6.9×10^{-1}	5.4×10^{-1}	3.0
	Number of husbandry days , days					Number of husbandry days , days				
	7	14	28	56	112	7	14	28	56	112
	Activity of organs, Bq/kg									
Heart	< 0.1	< 0.1	< 0.1	<0.080	<0.080	< 0.1	< 0.1	< 0.073	<0.078	<0.079
Kidney	< 0.1	< 0.1	< 0.1	<0.076	<0.080	< 0.1	< 0.1	< 0.072	<0.075	<0.072
Lung	< 0.1	< 0.1	< 0.1	<0.076	<0.079	< 0.1	< 0.1	< 0.068	<0.076	<0.074
Liver	< 0.1	< 0.1	< 0.1	<0.079	<0.079	< 0.1	< 0.1	< 0.073	<0.072	<0.079
Muscular tissue	< 0.006	0.01±0.004	< 0.004	<0.075	<0.068	< 0.1	0.012±0.002	< 0.070	<0.073	<0.069
Bone tissue	< 1.0	< 1.0	< 1.0	<1.3	<1.3	< 1.0	< 1.1	< 0.94	<1.3	<1.1
Testes	< 0.1	< 0.1	< 0.1	<0.080	<0.079	< 0.1	< 0.1	< 0.072	<0.077	<0.070
Spleen	< 0.2	< 0.1	< 0.1	<0.066	<0.080	< 0.1	< 0.1	< 0.070	<0.082	<0.10
Wool	1.3±0.3	< 0.37	< 0.40	< 0.36	< 0.31	0.93±0.24	< 0.37	< 0.41	<0.56	< 0.41

The table below (Table 24) presents data on the specific activity of sheep organs at different periods of ^{241}Am intake with food and water.

Table 24.

Specific activity of ^{241}Am in organs and tissues of sheep at different periods of its intake with food and water

Organs and tissues	Average daily intake with food, Bq/kg					Average daily intake with water, Bq/kg				
	1.2×10^1	1.4×10^1	2.0×10^1	2.8×10^1	2.4×10^1	4.8	8.3	4.4	2.4	2.6
	Number of husbandry days , days					Number of husbandry days , days				
	7	14	28	56	112	7	14	28	56	112
	Activity of organs, Bq/kg									
Heart	< 0.3	<0.2	< 0.3	<0.6	<0.4	< 0.4	< 0.4	< 0.4	<0.4	<0.4
Kidney	< 0.3	<0.2	0.7±0.03	<0.5	<0.6	< 0.3	< 0.4	< 0.4	<0.3	<0.4
Lung	< 0.3	<0.2	< 0.3	<0.3	<0.4	< 0.3	< 0.3	< 0.3	<0.3	<0.3
Liver	< 0.2	<0.2	< 0.3	0.7±0.03	<0.4	< 0.3	0.7±0.02	< 0.3	<0.2	<0.3
Muscular tissue	< 0.2	<0.02	< 0.03	<0.03	<0.05	< 0.2	< 0.2	< 0.03	<0.02	<0.03
Bone tissue	< 0.7	<0.6	< 0.9	<1.9	<2.4	< 0.4	< 0.4	1.2±0.4	<0.7	<1.3
Testes	< 0.4	0.6±0.02	< 0.3	<0.8	<0.3	< 0.4	0.6±0.02	< 0.3	<0.3	<0.3
Spleen	0.7±0.03	< 0.5	< 0.5	<0.7	<0.6	< 0.4	< 0.7	< 0.5	<0.8	<0.5
Wool	0.15±0.06	< 0.07	< 0.19	<0.183	0.22±0.09	0.25±0.1	< 0.08	0. 9±0.34	< 0.16	< 0.074
Skin	0.23±0.09	0.18±0.08	< 0.14	<0.09	<0.09	< 0.10	< 0.11	< 0.11	< 0.11	0.37±0.09

In the majority of cases, specific activity of $^{239+240}$Pu and ^{241}Am in tissues and organs was below the detection limit, but quantitative data allow a preliminary assessment of $^{239+240}$Pu and ^{241}Am transfer into the tissues and organs of sheep. Thus, the average K_n of $^{239+240}$Pu in the muscular tissue is $<1.3\times10^{-4}$, and K_n of ^{241}Am in the muscular tissue is $<1.5\times10^{-3}$.

The highest concentrations of ^{241}Am were registered only in the bone tissue of one animal; some concentrations were found in kidneys, liver, spleen and testes of several animals. Specific activity of ^{241}Am in the muscular tissue is below the detection limit, which amounts to 0.02 Bq/kg. A similar distribution pattern is observed in studies on exchange kinetics of ^{241}Am in the body of dogs [65]. It was determined that at prolonged intake with food or water in small concentrations, there is no process of depositing $^{239+240}$Pu and ^{241}Am in the tissues and organs of sheep.

CONCLUSION

In general, in our research we confirmed the existing data on the nature of radionuclides metabolism in animals. However, obtained transfer coefficients of ^{90}Sr and ^{137}Cs into muscular tissue of sheep at equilibrium are for an order of magnitude smaller than the average value of K_n in the IAEA database. Maybe their data rely on the different forms of ingestion. Thus, K_n of radionuclides, according to our data, at different intakes, i.e. with water or food, can vary within one order of magnitude. Perhaps the value of K_n is affected by the natural and climatic features (differences of micro-, macro element of the environment components, especially the concentrations of stable Sr and Cs and their chemical analogues in animal food), and radionuclides species in environmental media. All this points out to the fact that one can not

use K_n as a constant without knowing the conditions under which it was received. Despite the fact that these studies failed to adequately explore the characteristics of radionuclides $^{239+240}$Pu and ^{241}Am, on the basis of the results obtained we can say that these radionuclides are not dangerous in terms of dose formation for the population consuming animal products from "Degelen" site.

The results allow us to regulate the daily rate of radionuclides, excluding the excess of hygienic requirements for food products that are essential for the transfer of STS lands to commercial utilization.

Established dependence of the distribution of radionuclides ^{90}Sr, ^{137}Cs and T between the organs and tissues allows determining the content of radionuclides in the whole organism by an organ, or even, without butchering the animal, to give a preliminary assessment of radionuclide concentrations in organs and tissues by its wool. It should be noted that no similar studies carried out previously were found.

Territory of the STS is a unique "natural laboratory" where it is possible to study and solve a number of open questions of radioecology in natural conditions. This study is only the first step in this direction. In the future we are to create the site for radioecological agricultural research within the STS.

We are grateful to the staff of Environmental Engineering Laboratory of the Institute of Nuclear Physics (Almaty) for spectrometric and radiochemical analyses, to all employees of the department of integrated ecosystem studies of IRSE for their assistance in organizing and conducting radioecological works, and also express special appreciation to R.Yu. Magasheva for scientific advice and support.

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ССП ЖАҒДАЙЫНДА ЖАСАНДЫ РАДИОНУКЛИДТЕРДІҢ ҚОЙДЫҢ ОРГАНДАРЫ МЕН ҰЛПАЛАРЫНА АУЫСУ ЕРЕКШЕЛІКТЕРІН ТӘЖІРИБЕЛІ ЗЕРТТЕУ

Ж.А. Байғазинов, А.В. Паницкий, С.Н. Лукашенко, С.А. Келлер, С.С. Қаратаев, О.Р. Иванова

Бұл жұмыста жасанды радионуклидтердің әр түрлі түсу жағдайы мен мерзімі кезіндегі қойдың органдары мен ұлпаларына ауысуын зерттеу бойынша табиғи зерттеу нәтижелері ұсынылған. Т, ^{137}Cs, ^{90}Sr, $^{239+240}$Pu және ^{241}Am радионуклидтерінің қоршаған орта құраушыларымен: "Дегелең" таулы массивінің ауасы, суы және экожүйесі өсімдіктерімен түсуінің сандық бағасы берілген. Штольня суларымен түскен ^{137}Cs және ^{90}Sr биологиялық қолжетімділігі қойдың ор-

гандары мен ұлпалары үшін жемшөп өсімдіктеріне қарағанда жоғары. Жемшөппен түскен Т биологиялық қолжетімділігі сумен түскен Т қарағанда көп. Түсу мерзімі мен түріне қарамастан, ^{137}Cs , ^{90}Sr және Т органдар мен ұлпаларда таралуының нақты тәуелділігі бар, бұл олардың біреуі бойынша түрлі органдардағы радионуклидтердің құрамын бағалауға мүмкіндік береді.

Түйін сөздер: Семей сынақ полигоны (ССП), өту коэффициенті (Өк), қойлар, мүшелер мен ұлпалар, радионуклидтің биологиялық қол жетімділігі, тритий (Т), цезий–137 (^{137}Cs), стронций–90 (^{90}Sr), плутоний–239+240 ($^{239+240}$Pu), америций–241 (^{241}Am).

ЭКСПЕРИМЕНТАЛЬНОЕ ИССЛЕДОВАНИЕ ОСОБЕННОСТЕЙ ПЕРЕХОДА ИСКУССТВЕННЫХ РАДИОНУКЛИДОВ В ОРГАНЫ И ТКАНИ ОВЕЦ В УСЛОВИЯХ СИП

Ж.А. Байгазинов, А.В. Паницкий, С.Н. Лукашенко, С.А. Келлер, С.С. Каратаев, А.Р. Иванова

В работе представлены результаты натурных исследований по изучению особенностей перехода искусственных радионуклидов в органы и ткани овец при различных условиях и сроках их поступления. Дана количественная оценка поступления радионуклидов Т, ^{137}Cs, ^{90}Sr, $^{239+240}$Pu и ^{241}Am с компонентами окружающей среды: воздухом, водой и растительностью, при их содержании на опытно–экспериментальной площадке "Дегелен". Установлено, что биологическая доступность ^{137}Cs и ^{90}Sr для органов и тканей овец, поступивших со штольневой водой, выше, чем из кормовых растений. Биологическая доступность Т, поступившего с кормом, больше, чем Т, поступившего с водой. Определено, что независимо от времени и вида поступления, распределение ^{137}Cs , ^{90}Sr и Т в органах и тканях, имеет определенную зависимость, что дает возможность оценки содержания этих радионуклидов в различных органах по одному из них.

Ключевые слова: Семипалатинский испытательный полигон (СИП), коэффициент перехода ($К_п$), овцы, органы и ткани, биологическая доступность радионуклида, тритий (Т), цезий–137 (^{137}Cs), стронций–90 (^{90}Sr), плутоний–239+240 ($^{239+240}$Pu), америций–241 (^{241}Am).

УДК 577.391:504.74:591.146:546.11.02.3

STUDY OF TRITIUM CONCENTRATION DYNAMICS IN LACTOSERUM OF CATTLE FOR VARIOUS FORMS OF ITS INTAKE UNDER NATURAL CONDITIONS

Z.A. Baigazinov, A.V. Panitskiy, S.N. Lukashenko, S.A. Keller, S.S. Karataev, O.N. Lyakhova, L.F. Subbotina

Studies were carried out at "Degelen" experimental site located within the former STS. The paper presents findings on the dynamics of tritium transfer (hereinafter - T) into lactoserum under various conditions and timing of its intake in the body of cattle with components of environment: air, water and vegetation. We determined that the equilibrium state of T concentration in lactoserum is reached after 5-6 days of its prolonged intake with food. There has also been studied impoverishment of T concentration after the cessation of its intake. It was revealed that at prolonged intake of T with vegetable food it takes ~14 to impoverish its content in lactoserum for 2 times, at intake with water - 2 days.

Key words: tritium (T); cattle; lactoserum; organically bound T (OBT); free water, transfer (Kn).

INTRODUCTION

In contemporary literature on risk assessments of radiation exposure, we increasingly see the term "tritium problem" the study of which is devoted a significant amount of work. T, as an isotope of hydrogen, can replace it in all the molecules inside the cell. Its radioactive β - decay leads to violation of the molecular structures and intermolecular bonds under the action of β - radiation, as well as a result of conversion of T into an isotope of helium.

T is among the major dose-forming radionuclides existing in the territory of the former Semipalatinsk test site (STS). Of particular concern is intake of T into the human body through animal products, obtained under conditions of grazing on sites contaminated with tritium of Degelen massif and Shagan River, which necessitates a careful study of T transfer in various animal products.

So far, only few works have been carried out to study transfer of T into different components of cow milk (hereinafter - milk) [1]. When cows were watered with tritiated water single maximal content of T was found in the aqueous fraction of milk and a small amount in dry matter. Content of T in the organic fraction of milk after prolonged intake (25 days) becomes significant and can reach > 4% [2]. Transfer coefficient of T into milk during prolonged oral intake with tritiated water into cow bodies is 8 times higher than after single intake [3]. After the cessation of chronic administration of tritiated water periods it takes 6-6.5 and 7.5-8 days to impoverish its concentration two-fold in whole milk and dry matter, respectively [4].

We studied incorporation of T into organic components of milk - protein, fat and lactose, and fat tissue - caul fat and adipose capsule when feeding cows 28 and 160-170 days with dried grass grown in vegetative pot under irrigation with tritiated water [5]. It was found that at daily consumption of 1 kBq of organically bound T (hereinafter - OBT) ~ 50% of it in milk was incorporated in organic fractions, while the other half was in the aqueous fraction. Under the experiment [6], where two cows were injected hay contaminated with tritium through a fistula to rumen during 4 weeks, transfer coefficient of T into some fractions of milk was as follows: 0.84 for fat, 0.49 - casein, 0.05 – lactose, 0.10 - water.

No information have been found on this issue obtained from conditions as close as possible to natural ones, i.e. at real contamination of biological components - air, water and vegetation. There are no comprehensive full-scale studies that include a variety of conditions and timing of T intake into body of cattle. It should be noted that we have not found works carried out to study inhalation of T into animal organism and its transfer into milk.

This paper investigates the peculiarities of T transfer into lactoserum under various conditions and timing by field experiments under natural conditions of radioactive contamination.

1. BASIC RESEARCH METHODOLOGY

To study the change dynamics of T concentrations in lactoserum of cattle under natural conditions, we selected the ecosystem of watercourse at tunnel # 177 of "Degelen" experimental site. Scheme of watercourse ecosystem area, where an experiment was conducted is shown in the figure (Figure 1). Watercourse was formed with groundwater which flow through the cavity of the explosion, taking the radionuclides to the surface [7], thus forming the basic ecosystem contamination with tritium and other radionuclides over a large territory. In tunnel #177 on March 30, 1983 test of a nuclear charge with capacity of 0.001-20 kt with the purpose of nuclear weapons development was carried out [8].

Width of the watercourse at tunnel #177 mostly is less than 1 m. Annual average water output is $4.4*10^8$ litres (more than 500 l/min) [9]. Studies performed in 2007-2008 say that the concentration of T during this period in tunnel water ranged within 300-350 kBq/l, in atmospheric air - up to 1,400 Bq/m^3, the highest concentration of T in the vegetation reaches $2.4*10^5$ Bq/kg [10].

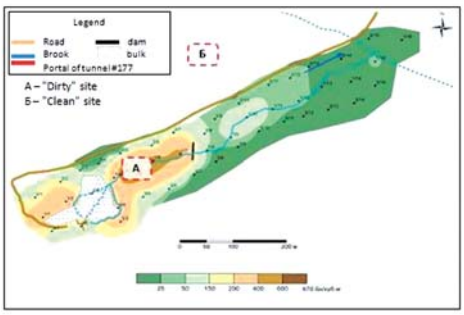


Figure 1. Scheme of watercourse ecosystem at tunnel #177

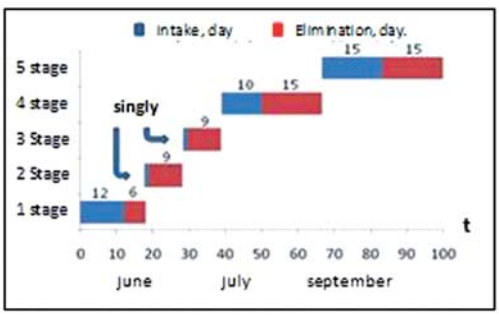


Figure 2. Experiment stages

Venue of the experiment is conventionally divided into two sites. "Clean" - a site with background concentrations of T in the air and plants (0,6±0,06 Bq/m^3, 60±7 Bq/kg, respectively), "dirty" - the area where the previous studies registered the highest concentration of T in the air, plants and water (streambed of watercourse at tunnel #177).At these sites we have set paddocks for cattle. The research site has been split into areas of different types of T intake into animal body:

- prolonged intake with atmospheric air (stage 1);
- one-fold intake with water (stage 2);
- one-fold intake with meadow grass (Stage 3);

- prolonged intake with water (Stage 4);
- prolonged intake with meadow grass (Stage 5).

Procedure of the experiment and duration of the stages are represented in the figure (Figure 2).

As a subject of investigation we chose 8-9 years of age local manorial cow with live weight of 250-260 kg, with daily average milk yield of 6-8 kg. Throughout the study we used tethered husbandry. Milking and feeding with concentrated feed (bran) was carried out in the morning and evening. During the experiment, the overall condition of the animal was assessed as normal, the behaviour was active, had good reaction to sound and pain stimulation, stance is natural, but after 10-15 day of the experiment we marked decrease in the average daily milk yield by 5-6 kg, which persisted until the end of the study. Apparently, this is due to changes in the conditions of husbandry. Main sources of T intake into animal body were air, water and tunnel meadow vegetation of radioactively contaminated ecosystems of the watercourse at tunnel #177.

2. RESEARCH METHODOLOGY IN STAGES

2.1. At prolonged intake with atmospheric air

An animal was kept for 12 days in the "dirty" area with high content of T in the air, and then was relocated to a "clean" area to continue observation. Milking was twice daily, morning and evening. We took 1.5 l of milk each day after the morning milking for radionuclide analysis. To determine the intake of T into body of cattle with atmospheric air we studied the dynamics of changes in the concentration of T in the air at the cattle husbandry venue.

Determination of T concentration in the air was carried out by the method of cryogenic freezing. The method is based on the ability of water vapour in air to condense on the surface with a temperature below the dew point. So, the radiator with refrigerant is placed in a Dewar flask filled with liquid nitrogen for 30-50 minutes. Contact of water vapour in the air with the surface of cooling fin leads to crystallization of vapour. Fins were at an altitude of about 60-70 cm from the surface. After the freezing the sample is placed in a container for thawing and then filtered. Sampling was conducted around the clock every 2 hours on the third and eighth day of the experiment.

2.2. At one-fold intake with water

Experimental animal was fed with ~ 20 ± 0.5 tunnel water in a single-shot. Control sample of milk was pre-taken. First 3 days after intake of tunnel water milking and milk sampling were conducted every 4 hours. Further, within 6 days milking was twice a day, morning and evening. Milk for analysis was taken after the morning milking.

2.3. At one-fold intake with meadow grass

For an experimental animal we harvested ~ 10 kg of meadow vegetation from "dirty" area. Immediately after the morning milking the animal was fed on new-mown grass. An intense eating of grass was for 3 hours, after which we collected and weighed the remains

of food that amounted to about 2.0 ± 0.2 kg. Already in the evening of the same day, we fed and watered the animal on "clean" hay and water. Over the next nine days we were observing the change dynamics of T concentration in the lactoserum. The first 4 days milking and milk sampling were conducted every 6 hours after the start of feeding on "dirty" grass. In the next five days milking was twice a day, morning and evening; for analysis we took milk after morning milking.

2.4. At prolonged intake with water

Daily for 10 days, experimental animals was fed with "dirty" water. And then animal had "clean" water and within 30 days were observing elimination of T with milk. The animal was fed with "clean" grass throughout. Milking was twice a day, morning and evening. For analysis we took milk after morning milking. When watering we recorded daily quantity of water drunk by experimental animal.

2.5. At prolonged intake with meadow grass

Daily for 15 days twice a day the experimental animal was fed with mown vegetation growing in the "dirty" area. Later the animal had "clean" grass and within 20 days we were monitoring the concentration of T in lactoserum. Milking was carried out 2 times a day, morning and evening. For analysis, we took milk after the morning milking. When feeding we recorded daily amount of grass eaten by the experimental animal. Also we assessed the content of T in the vegetation that is eaten by animal.

2.6. Sample preparation and the method for T measurements

At each milking throughout the experiment the cow was milked fully. Preparation of samples for radiological analysis was carried out by curdling the milk, keeping it for ~2 days at 20-22 ºC (without acidulating). Milk curds went through a sieve of 1 mm. From the resulting lactoserum we prepared an analytical probe to determine T, which was obtained by distillation at a temperature of 100 ºC. First obtained condensate of 10 ml was eliminated, and for analysis, we took the following 5-6 ml. Activity of tritium in tunnel water, air and water fraction of lactoserum and plant free water were measured on a beta spectrometer TRI CARB 2900 TR, by liquid scintillation method of research method according to ISO 9698 [11]. Analytical measurement error is no more than 30 %.

2.7. Calculation of T transfer coefficient in lactoserum

As the parameter of T intake level from ambient medium into lactoserum, we used radionuclide transfer coefficient K_n, calculated as the ratio of specific activity of lactoserum (Bq/l) to the total amount of radionuclides intaken during the day into body of the animal with air or diet (Bq/day), expressed as a percentage:

$$K_n = \frac{A_l}{A_{day}} * 100 \text{ , where}$$

A_l – specific activity of T in lactoserum, (Bq/l);
A_{day} – total activity of T intaken by animal body within 24 hours, Bq/day.

3. RESULTS AND DISCUSSION

3.1. Studies T concentration dynamics for lactoserum at prolonged intake with atmospheric air

3.1.1. Assessment of T intake into animal body

Based on average values obtained from the results of two measurements, we plotted the dynamics in concentration of T in atmospheric air. As one can see (Figure 3), the concentration of T in atmospheric air during the day varies greatly. Therefore, when calculating we divided the average daily intake into 2 segments, since the volume of inhaled air is changed during rest, wakefulness and feeding. Thus, average concentrations of T in the air during the daytime from 06,00 am to 18.00 pm and night-time from 18.00 pm to 06.00 am were ~133 Bq/m^3 and ~55 Bq/m^3, respectively. According to the literature [12], the volume of air inhaled daytime during 12 hours is ~ 72 m^3, at night-time ~ 36 m^3. On the average, intake of T for 24 hours with atmospheric air into the cow's body was ~ 11.5 kBq/day.

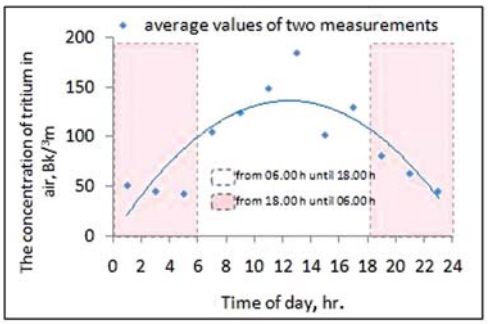


Figure 3. T concentration dynamics in atmospheric air at cattle husbandry venue round the clock

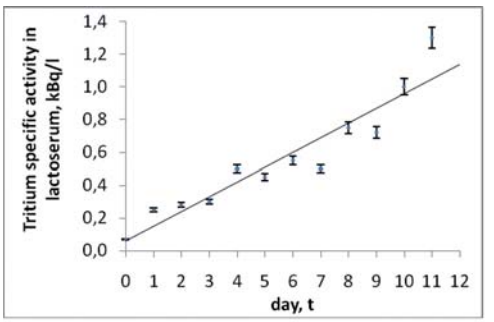


Figure 4. Activity of T in lactoserum at intake with atmospheric air

3.1.2. Concentration of T in lactoserum

Findings of first stage are shown in the figure (Figure 4), which shows that when T is intaken with atmospheric air into the body of cattle within 12 days there is almost a linear increase in T concentrations in lactoserum and milk and no balanced state is reached. Upon that at 11th day after commence of the experiment the concentration of T in the lactoserum was 1.3 ± 0.1 kBq/l (K_n = ~ 11.3%).

Our results show that aerial intake of T into the animal can make a significant contribution to the contamination of animal products.

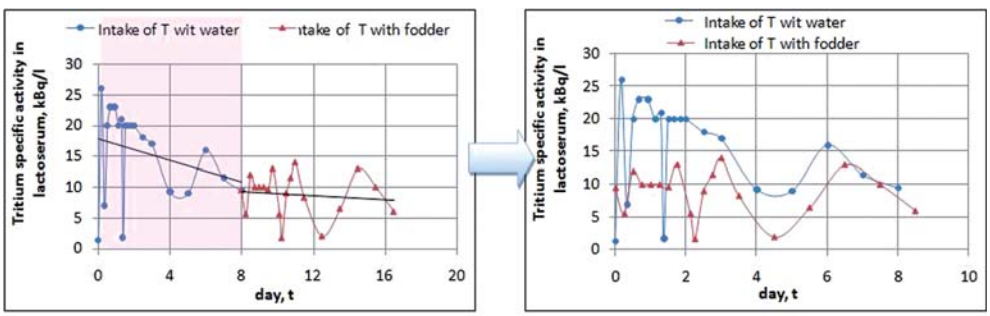
3.2. Study of T concentration dynamics in lactoserum at one-fold intake with tunnel water

3.2.1. Assessment of T intake into animal body

Based on data on concentration of T in tunnel water, which is average 330 kBq/l, and amount of water given we calculated T intake into experimental animal, which amounted to ~ 6,500 kBq/day.

3.2.2. Assessment of T content in lactoserum

Dynamics of changes in concentration of T in lactoserum at one-fold intake with water is shown in the figure (Figure 5, A). As a consequence of T intake with water, the maximum concentration of T in the lactoserum was 26±2.6 kBq/l (K_n = ~ 0.4) and recorded in 4 hours after watering. Further, we observed decrease in concentration of T in the lactoserum and sharp drop after 8 hours and 33 hours after animal had "clean" diet. On the third day the character of decrease in concentration becomes abrupt.



a) b)

Figure 5. Dynamics of T concentration in lactoserum at one-fold intake with water and meadow grass

3.3. Study of T concentration dynamics in lactoserum at one-fold intake with meadow grass

3.3.1. Assessment of T in the animal

To assess T intake into animal organism there has been calculated T intake with the free and organically bound water of plants. Determination of plant free water was carried out by the standard method for determining the initial and hygroscopic moisture [13]. It was determined that the amount of plant free water on average is 70 ± 10% (min - 56%, max - 83%). Based on our measurements of T concentration in plant free water, taken from the total mass of forage before feeding, it was found that the activity of T on the average is 270±30 kBq/l. We used computational method to determine organically bound water. We were based on the assessment that the organically bound water in the dry matter of plants is ~60%. Activity of T in the organically bound water of plant was taken equal to the activity of T in plant free water. Thus, the total amount of T at single intake (within 3 hours) was ~ 1,900 kBq.

3.3.2. Assessment of T content in lactoserum

Changes dynamics in concentration of T in lactoserum at one-fold intake with grass is shown in the figure (Figure 5, a). The curve has a complex character that is paranormal at this stage and requires further research. However, if you compare the two curves (Figure 5, b) one can see monotonous oscillations, starting from the third day after feeding and watering, which might suggest the physiological characteristics of the animal during the formation of milk. Maximum concentrations of T in lactoserum - 14 kBq/l (K_n = ~ 0.6) were registered only after 72 hours after feeding.

3.4. Study of T concentration dynamics in lactoserum at prolonged intake with drinking water

3.4.1. Assessment of T intake into animal body

To assess T intake with water during 10 days we were registering the amount of "dirty" water consumed. In ten days the animal drank ~220 litres of water. To assess the intake of T with water into experimental animal, we studied the dynamics of change in concentration of T in water of the watercourse at tunnel #177. It was found that concentration of T in water during the experiment did not change and was fairly constant (320-340 kBq/l). Based on these data, we calculated the daily intakes of T with water which was ~7,300 kBq/day.

3.4.2. Assessment of T content in lactoserum

Dynamics of T concentration in the lactoserum at prolonged intake with water is shown in the figure (Figure 6).

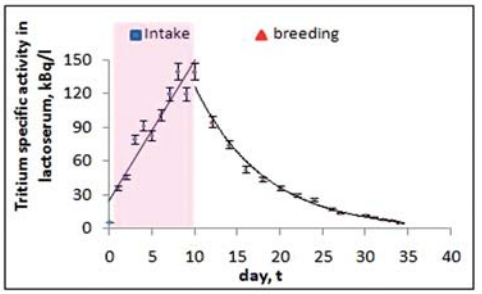


Figure 6. Dynamics of T concentration in lactoserum at prolonged intake with water

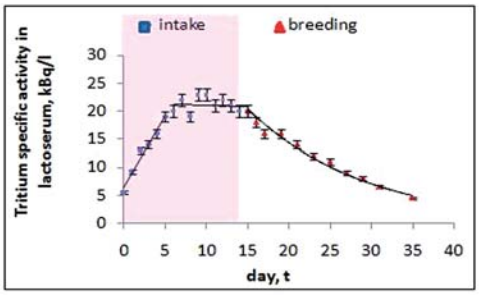


Figure 7. Dynamics of T concentration in lactoserum at prolonged intake with meadow grass

As it can be seen from the figures above, concentration of T in the lactoserum grew continuously during the entire 10 days of intake with water and did not reach its equilibrium state. However, in final days T concentrations in lactoserum ranged within 120-140 kBq/l. Perhaps in this period the organism is saturated with radionuclides and there comes balanced state of T concentration in lactoserum. Maximum concentration of T in lactoserum at 8-10th day of intake was 140 kBq/l (K_n = ~2%). Once tunnel water in no more consumed, the T concentration in lactoserum decreases exponentially that can be approximated by the following equation (1):

$$A=410e^{-0,12t} \quad (1)$$

Thus, period of impoverishing the concentration of T in lactoserum two times is ~2.5 days.

3.5. Study of T concentration dynamics in lactoserum at prolonged intake with meadow grass

3.5.1. Assessment of T intake into animal

To monitor T intake into animal we performed daily records of eaten meadow grasses. Determination of T in plant free water was carried out on the basis of analysis of plant samples

taken every three days. Average concentration of T in plant free water was 80±9 kBq/litre. Calculation of amount of organically bound water in the plant and T activity in it was carried out similarly to the procedure described above. Given the amount of food consumed, average amount of free and organically bound water in it, we calculated the total daily intake of T into body (Table 1). To calculate transfer of T into lactoserum, we used the average daily activity of a radionuclide in the food that is 1,100 kBq/day.

Table 1.

Daily intake of T into the body of experimental animal with meadow grass

Number of days	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Amount of food consumed, kg	18	18	15	15	16	16	14	13	15	15	16	15	15	16	16
Daily intake of T with PFW*, kBq	1033	1033	861	861	918	918	804	746	861	861	918	861	861	918	918
Daily intake of T with OBPW**, kBq	266	266	221	221	236	236	207	192	221	221	236	221	221	236	236
Total intake, Bq/day	1,299	1,299	1,082	1,082	1,155	1,155	1,010	938	1,082	1,082	1,155	1,082	1,082	1,155	1,155
Note: *PFW – plant free water; **OBPW – organically bound plant water															

3.5.2. Assessment of T content in lactoserum

Dynamics curve (Figure 7) of T concentrations in lactoserum at chronic injection with meadow grass at the stage of "intake" can be divided into two phases: from 1st to 5th day - phase of an intensive increasing of T concentration in lactoserum, from 6th to 15th day – phase of balanced state of T concentration in lactoserum. Maximum concentration of T in lactoserum was registered in 9-10th day (23 kBq/l, K_n = ~1.8%). Once we stopped feeding an animal with meadow grass contaminated with tritium, the dynamics of reducing the concentration of T in lactoserum can be described as exponential curve approximated by equation (2):

$$y=58e^{-0,07t} \quad (2)$$

Thus, it takes ~14 days to impoverish T concentration two-fold in lactoserum at prolonged intake with meadow grass, which is 6-7 times longer than at prolonged intake with water.

CONCLUSIONS

When T comes with atmospheric air into the body of cattle within 12 days, there is almost a linear increase in T concentration in lactoserum, upon that no balanced state is reached. These results show that aerial intake of T into animal can make a significant contribution to the contamination of animal products.

Dynamics of T concentration in lactoserum at single intake with meadow grass and water has a complicated nature, possibly related with physiological properties of the animal during the formation of milk, which requires additional research.

Conducted research determined that at Degelen Mountain, under chronic T intake with meadow grass equilibrium state of its concentration in lactoserum occurs at 5-6th day. At chronic T intake with atmospheric air and water equilibrium state of its concentration at selected time intervals has not been achieved.

In its turn, impoverishment of T concentration in lactoserum after chronic T intake with meadow grass is much slower than after chronic T intake with water. Thus, reduction of T concentration by 50% after long-term intake with water occurred after ~2 days, after long-term intake with meadow grass - ~14 days.

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ТРИТИЙДІҢ ТАБИҒИ ЖАҒДАЙЛАРДА ТҮСУІНІҢ ӘР ТҮРЛІ ФОРМАЛАРЫ КЕЗІНДЕГІ ОЛАРДЫҢ ІҚМ СҮТІ САРЫСУЫНДАҒЫ ШОҒЫРЛАНУЫНЫҢ ӨЗГЕРУ ДИНАМИКАСЫН ЗЕРТТЕУ

Ж.А. Байғазинов, А.В. Паницкий, С.Н. Лукашенко, С.А. Келлер, С.С. Қаратаев, О.Н. Ляхова, Л.Ф. Субботина

Зерттеу бұрынғы ССП аумағында орналасқан "Дегелең" тәжірибелік-эксперименттік алаңда жүргізілді. Бұл жұмыста тритийдің (ары қарай Т) қоршаған орта құраушылары: ауа, су және өсімдіктермен ІҚМ ағзасына әр түрлі түсу жағдайлары мен мерзімі кезінде сүт сарысуына ауысуының динамикасын зерттеу бойынша нәтижелер берілген. Т сүт сарысуында шоғырлануының тепе-теңдік жағдайы оның жемшөппен ұзақ уақыт түсуі кезінде 5–6 тәулікте анықталатыны белгіленді. Т сүт сарысуында шоғырлануының оның түсуі тоқталған соң төмендеуі зерттелді. Т өсімдік жемшөбімен ұзақ уақыт түсуі кезінде оның сүт сарысуындағы құрамының төмендеу уақыты екі реттен ~ 14 күн, ал ұзақ уақыт сумен түскенде – 2 күн құрайтыны анықталды.

Түйін сөздер: тритий (Т); ірі қара мал (ІҚМ); сүттің сарысуы; органикалық байланысқан Т (ОБТ); еркін су; өту коэффициенті (Өк).

ИССЛЕДОВАНИЕ ДИНАМИКИ ИЗМЕНЕНИЯ КОНЦЕНТРАЦИИ ТРИТИЯ В СЫВОРОТКЕ МОЛОКА КРС ПРИ РАЗЛИЧНЫХ ФОРМАХ ЕГО ПОСТУПЛЕНИЯ В ЕСТЕСТВЕННЫХ УСЛОВИЯХ

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Исследования проведены на опытно-экспериментальной площадке "Дегелен", расположенной на территории бывшего СИП. В работе представлены результаты по изучению динамики перехода трития (далее – Т) в молочную сыворотку при различных условиях и сроках поступления его в организм КРС с компонентами окружающей среды: воздухом, водой и растительностью. Установлено, что равновесное состояние концентрации Т в молочной сыворотке при его длительном поступлении с кормом устанавливается на 5-6 сутки. Изучено снижение концентрации Т в молочной сыворотке после прекращения его поступления. Выявлено, что при длительном поступлении Т с растительным кормом время снижения его содержания в сыворотке молока в 2 раза составляет ~14 дней, а при поступлении с водой – 2 дня.

Ключевые слова: тритий (Т); крупнорогатый скот (КРС); сыворотка молока; органически связанный Т (ОСТ); свободная вода; коэффициент перехода ($К_п$).

CHAPTER: "NON-RADIATION" SAFETY CONCERNS AT STS

УДК 577.4: 622.278: 621.039.9

ASSESSMENT OF POSSIBILITIES FOR CATASTROFIC EVENTS AT "BALAPAN" SITE

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The article presents the results of studies on the effects of underground nuclear explosions (UNE) related to subsidence phenomena and processes of underground gasification of rocks. Gathering information about the "warfare" boreholes on the test site "Balapan" according to literary sources and data funds allowed compiling a complete catalog and making their location layout. As a result of reconnaissance works there was found subsidence phenomenon in a number of boreholes, according to the topographic monitoring of epicentral zones information was obtained about their dynamics for the first time. The paper also presents the outcomes of the studying the composition and concentration of gases coming from blocks of rocks containing the central zone of UNE, to the surface. The estimation was done for the significance of hazards associated with long-term effects of residual manifestations of geothermal activity in the epicentral areas of UNE as local processes of the catastrophic nature. To increase the information content of work done, recommendations were developed for the use of additional research methods.

Key words: Semipalatinsk test site, carbon monoxide, methane, hydrogen sulfide, underground gasification, thermal digestion, gas emission.

INTRODUCTION

Besides the threat of radiation impact in the area of Semipalatinsk Test Site (STS) there is a range of other hazards which have to do with residual effects of long-term geothermal activity in the epicenter areas of underground nuclear explosions (UNE) and existence of uncollapsed cavities.

Military and civil specialists who worked in Kurchatov which UNE took place often witnessed gas effervescence. There were several cases of gases self-inflammation and there sustainable burning for many days. The most serious catastrophic demonstration of rock gasification was in April of 1992 at Balaban site at the wellhead of "Glubokaya" borehole (UNE was made in 1977). In April of 1992 there was a sudden funnel-shaped surface draw at the wellhead of this borehole. Draw formations was accompanied by an explosion and fire. Currently this funnel has a diameter of 200 meters with depths of 20 meters. This event can be classified as a local catastrophic event. This happened 15 years after nuclear explosion in this borehole. In the history of UNE at STS there were no similar events during explosions or after them.

For a long time the event at "Glubokaya" borehole was covered by rumors, but systematic and detailed study of this event started in 2007 within the frame of special program 011 "STS Safety provision". There was a logical question if this event can repetitiously occur and what is situation at other warfare boreholes.

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1. PRECONDITIONS FOR OCCURANCE OF CATASTROFIC EVENTS AT "BALAPAN" SITE

1.1. Geological structure and hydrogeological conditions at "Balapan" site

STS territory is part of eastern sector of big Paleozoic Ural-Mongol belt. Fold rock located at above mentioned territory belong to two major tectonic structures – Caledon Chingiz-Tarbagatay meganticlinorium and gertzinsky Zaisan fold system. Border between these strictures goes along steep deep Kalba-Chingiz fault. These fold structures differ not only by different development histories, but also by age and structure of fold complex base. Below is their brief characteristic.

"Balapan" site is located within STS at its south-eastern part, 100 km from Kurchatov city. Geological structure of "Balapan" site (Figure 1) is determined by its location at south-western slope of Zharminskaya synclinorium which has formed during gertzinsky cycle of geotectogenesis.

Cambrian system. At the south, behind Kalba-Chingiz fault caledonides of Chingiz-Tarbagataiskiy meganticlinorium with medium-upper coal-bed intrusions crop out. Sediments of this age are combined into maidan assise subdivided into two stratums: lower volcanogenic-terragenic and upper terragenic – siliceous. This Starata thickness is up to 1120 meter.

Carbonic rock system. Carbonic rock system occupies most of "Balapan" site. At the south-west of the site there are mainly Kokon assises located, within Chinrau geoblock enclosed between Kalba-Chingiz and Chirau faults. Rock is poorly disposed and split with faults into tectonic blocks. Under impact of intrusions and tectonic processes, sediment rock of assise are orogenic, schistose, silicified. Assise composition is carbon siliceous sandy silt with rare layers of lava and andesite and andesite-basalt tuff porphyrite. Internal structure of assise is complicated by facial instability of sections, wide development of contact and dislocation metamorphosis. Total thickness of sediments is 2,300 m.

Arkalyk assise sedimants witin "Balapan" site have abruptions onto original ground in a form of north-west band in Zharminskaya zone at the north-east side of the site behind Baiguzin-Bulak fault. From north-east Arkalyk assise are unevenly covered (with basalt conglomerates at the base) by sediments of Sirektas assise and from south-west – by conglomerates of upper-high carbon. Sediments are represented by conglomerates, gravelites, sandstone and siltstone with layers of basalt porphyrites and their tuffs, limestone-siliceous seltstones. Sediments of Arkalyk assise form the most thick layers, total thickness is up to 4,000 m.

Sirektau assise sediments have wide areal distribution at the north-east and occupy tectonic block limited by Baiguzin-Bulak and North-Zhanan faults. Contacts with lower layers are tectonic. In assise cross-section main and medium composition tuffs occupy most of space. Andesite prophyrite tuffs of light green and sometimes light grey colour of psammo-pseuphitic structure with irregular texture. Assise sediments are bulged into folds of north-west direction and broken into separate blocks by multiple fracturing breaches running into different direction. Total assise thickness is 1,500–3,700 m.

Bukon assise sediments form Zhanan goeblock located between Baiguzin-Bulak and Chinrau faults. Sediments are formed by continental and sub continental sediments subdivided into two assises according to their lithologic composition. Cross section is represented by narrow alternating sandstone, conglomerates, siltstone, and carbon schist with coil layers lenses. Total thickness of Bukon assise is 2,500–3,000 m.

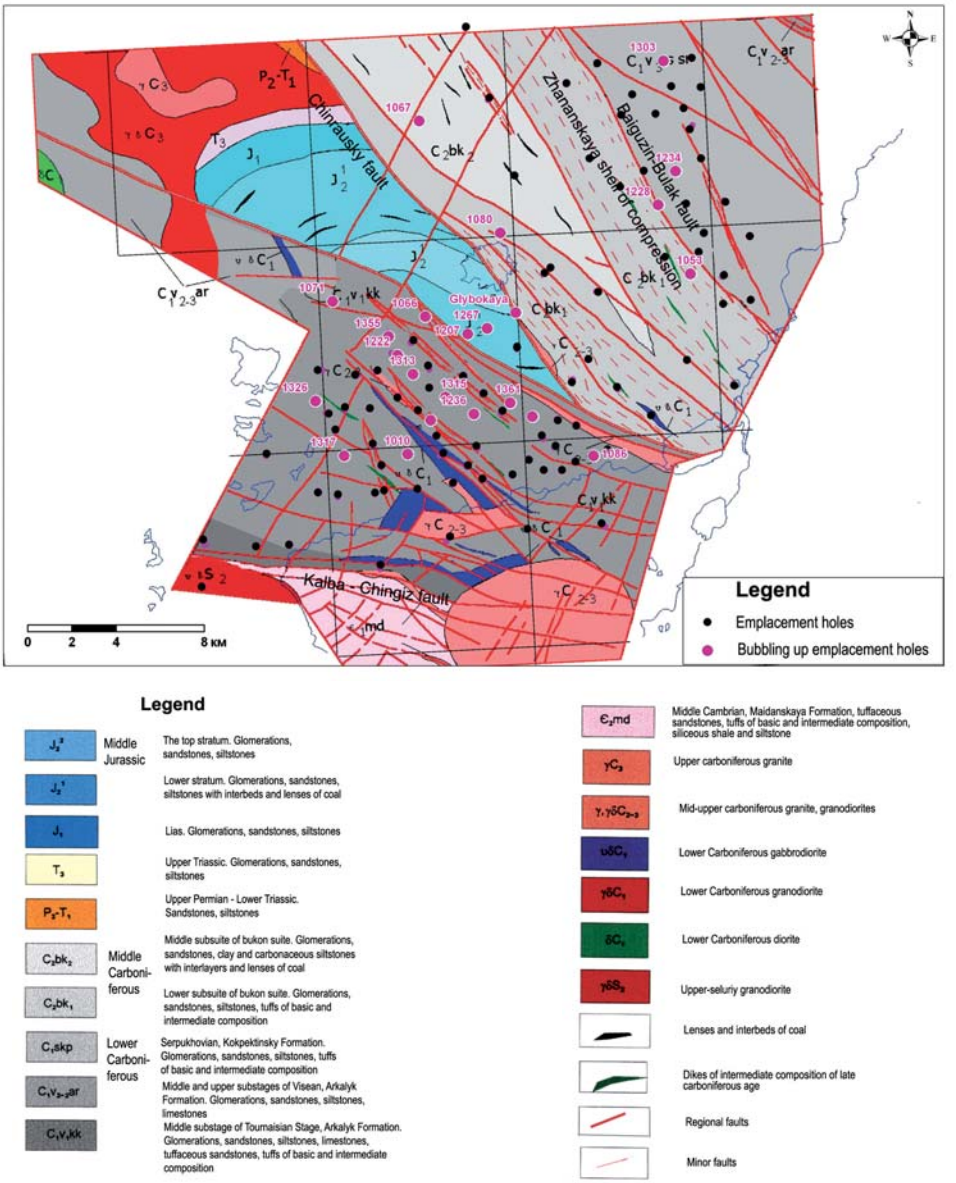


Figure 1. Geological map of "Balapan", 1:50 000 scale

Triassic system. Triassic system sediments were found at the northern side of the site where they form southern part of Akbota fault through of 1.5–2 km, observed at south-western side of Chinrau fault of 30 km length. At the bases of profile basalt conglomerates are present. They contain fine-grained sandstone and alerovites with lens and coal layers. Here

there are small horizons of basalts and andesite. Upper part of profile consists of bigger grain sandstone and gravelites. Sediments thickness is up to 350 m.

Jurassic system. Lias sediments located either at upper triass or directly on granite foundation and subdivided into two complexes.

Upper coal bearing complex is mainly represented by alevrolites and sandstone with coal layers. Gravelites, conglomerates and shale oil are rarer. Dogger rock thinckness is up to 100 m. Above carboniferous complex is characterized by alternating layers of speckled conglomerates, gravelites and sandstone. Alevrolites and mudstones are rarer. Complex thickness is 180 m.

Neogenic system. Practically all territory of "Balapan" site is covered by a layer of neogenic clay, excluding north-western and south side. Clay cover layer thickness alternates from 10 to 70–80 m.

Quaternary system. Formations of this age are represented by gravel pebble rock, different fraction size sand, clay. Sediment thickness at different sites varies from 5 to 70 m.

Hydrogeological conditions. There are two hydrogeological complexes at the described territory, sometimes connected with each other. First complex includes water located in local hydrogeological basins. Water bearing rock is represented by loose deposits from neogen to modern age. Second complex is included into regional hydrogeological system. This includes fracture water of Paleozoic foundation, Mesozoic residual soil water and paleogenic deposits water. Typically they form one aquifer of 100–150 m thickness.

At periphery of "Balapan" site where drainage relief forms are located, fracture water level is close to original ground. At the sites, covered by waterproof neogen clays, fracture water gain artesian property. Water head is directly proportional to clay thinkness which alternates from several meters to 80 meters.

Above described nature of geological structure and hydrological conditions suggests that rock located within "Balapan" site limits are subject to occurrence and underground burning due to destructing UNE factor. Deposits of central area of the site are more prone to this process – these are Jura deposits and also deposits of carbonic system (coal layers, coaly siltstone and schist). Warfare boreholes layout (Figure 1) shows that some of boreholes are located close to coal structure of secondary mesozoic Jubilee trough and some of boreholes is located right in deposits of Bukon set of rocks. Rock in these areas are characterized by higher content of coaly material which is one of the factors for possible occurrence of ground gasification process and pyrolysis after UNE [1, 2].

1.2. Nuclear explosions at "Balapan" site and their consiquences

1.2.1. Mechanical consequences of UNE (technogenic changes in surface)

Nuclear explosion energy release happens within one micro second exceeding charge temperature up to tens of millions of degrees. Charge substance heated to such high temperatures releases energy in a form of electromagnetic radiation of very wide spectrum – from roentgen to long wave [3]. Estimates made for Rainier explosion [4], had shown that within several microseconds the temperature in the explosion chamber was 10^6 K and pressure 10^7

aph. In such conditions a charge substance and some of adjoining rock turns into partially ionized plasma.

Joint impact of strong blast and intensive thermal radiation evapourates adjoining rock and forms steam sphere called evapouration cavity [5]. Cavity size is mainly determined by explosion power and rock properties. On the average 70 tonns of silicate rock evaporate per kiloton of explosion power. During distribution of spherical last it melts about 350 tons of rock per kiloton of explosion power and heats roughly up to 800 ºK around 600 t/kt. Temperature in the rock after the blast is not distributed evenly: it decreases from the rock evapouration temperature at inner wall of stabilization cavity to the the surrounding formations temperature within tens and hundreds of meters from the cavity [6]. Cavity radius may be presented as an empiric correlation connecting explosion power W, charge location depth and upper rock density ρ:

$$R_c = C\frac{W^{1/3}}{(ph)^{1/4}}$$,where

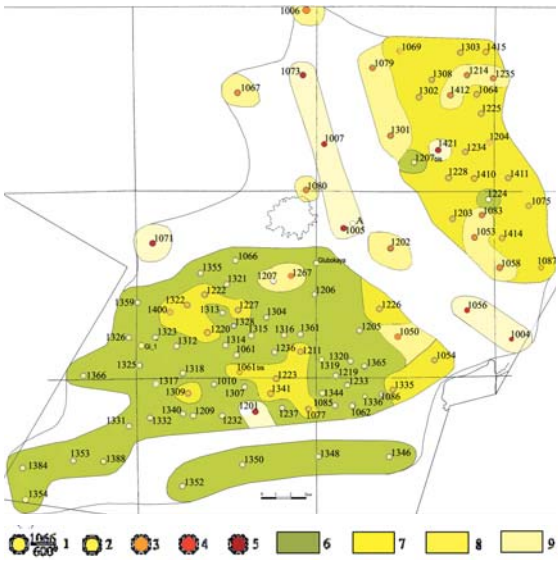
W – explosion power;
C – empirically determined coefficient;
p – upper rock density;
h – charge location depth.

Calculation results of UNE cavity radiuses at "Balapan" site are presented in a table (Table 3). Data presented in the table shows that quantitative values of cavity radiuses alternate from 10 to 40 meters.

In the collapse process steam-gas components contact with relatively cool surfaces of falling rick debris. Starting from this moment, processes of mass transfer regulate formation of radioactive contamination of the rock in the collapse column, rock debris fall into melted mass, gases and steam which is pushed through the collapse column contact debris and cool down rapidly. Formation of a column continues until quasi stable cove is formed which in a range of experiments leads to formation of a cavity underneath this cove [7]. During explosions in weak rock (sediment rock) growth of collapse column to an original ground is possible. In such cases so called "sediment funnel" is formed. Accroding to performed studies, a collapse column has a cylinder shape with radius slightly different from radius of initial cavity. From the top it is limited by a cope of slightly destroyed rock. Lower part of the column is a preserved or slightly transformed lower cavity semi sphere.

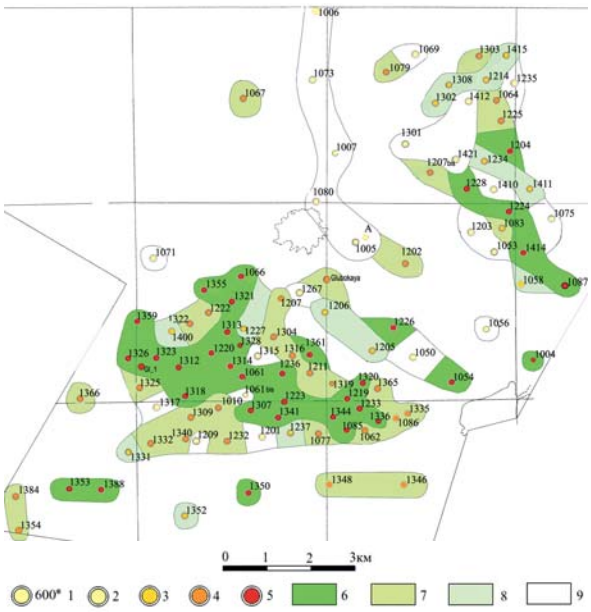
1.2.2. General information on nuclear tests at "Balapan" site

All together at "Balapan" site there are 106 warfare boreholes, where UNE was made [8]. Geological map (Figure 1) shows borehole location layout. With the purpose of typing of conditions in rock volume covered with UNE, data on warfare boreholes were systematized according to depth, power and magnitude of explosions. Received schemes of technogenic load on interior depending of the depth of nuclear charge are presented in Figure 2a, and nuclear charges power distribution is presented in Figure 2 b, density of warfare boreholes - Figure 2c [9].



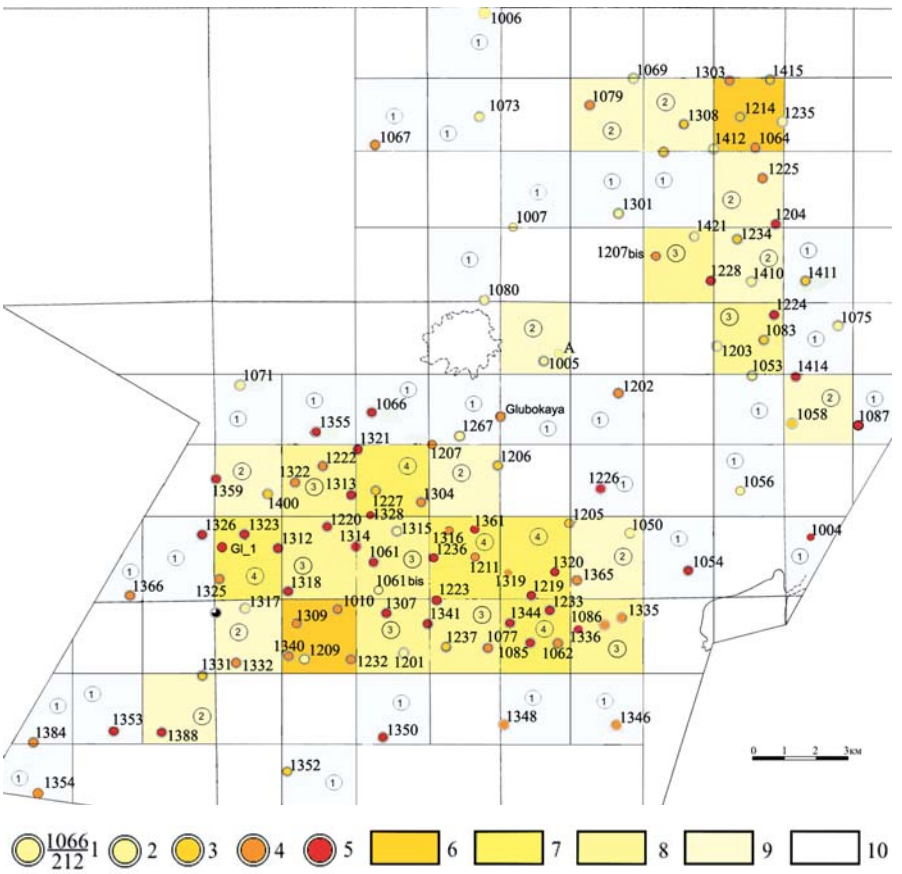
1 – warfare borehole (number/nuclear charge depth). Nuclear charge depth: 2 – more than 500 m; 3 – 400–500 m; 4 – 300–400 m; 5 – less than 300 m. Area with nuclear explosions at the depths of: 6 – more than 500 m; 7 – more than 500 m; 8 – 400–500 m; 8 – 400–300 m; 9 – less than 300 m.

a



1 – borehole. Nuclear charge power: 2 – up to 20 kt;
3 – 21–50 kt; 4 – 51–100 kt; 5 – more than 100 kt. Area with nuclear explosions with power of:
6 – more than 100 kt; 7 – 51–100 kt; 8 – 21–50 kt; 9 –. до 20 kt

b



warfare borehole (number/nuclear charge power). Nuclear explosion power (kilotons): 2 – up to 20; 3 – 21–50; 4 – 51–100; 5 – more than 100. Area with great number of nuclear explosions (explosion): 6 – 5–6; 7 – 4; 8 – 3; 9 – 2; 10 – 1

c

Figure 2. "Balapan" site. Technogenic load scheme: a – from nuclear charge location depth; b – nuclear charge power; c – explosion density

Data analysis has shown that after a nuclear device test, technogenic effect is manly demonstrated in the form of soil blistering (boreholes 1062, 1075, 1079, 1087, 1220, 1225, 1227, 1235, 1400, 1410), and sometimes – in the form of insignificant depressions (1233, 1236, 1314, 1316).

At some boreholes absolute marker values stayed unchanged (1061bis, 1080, 1202, 1237, 1317). Maximum changes of a original ground were at wellheads 1062 and 1220, where soil blistering was 2.8 and 3.9 m. Usually technogenic changes are registered within 500 meters radius from wellhead, but at some boreholes depressions and blistering occurred within 1500 meters radius (borehole 1062). At five sites (1061bis, 1080, 1202, 1237, 1317) there was no technogenic changes of original ground instrumentally registered after nuclear device test.

On picture (Figure 3) there are examples shown on original ground changes (in absolute markers); on one of four profiles in wellheads of boreholes 1220 and 1314 – before and after UNE.

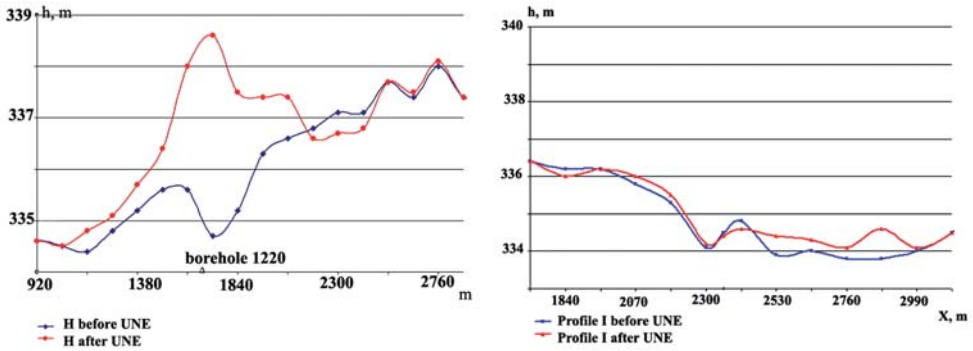


Figure 3. "Balapan" site. Boreholes 1220 and 1314. Original ground changes after UNE

As we can see from Figure 3, after UNE original ground demonstrate soil blistering and also depression with fluctuations of absolute markers from (–0.4) m to (+3.9) m.

Figure 4 below demonstrates changes of original ground under impact of UNE as a difference of results of topogeodesic measurements before and after explosion in the borehole 1206 [9].

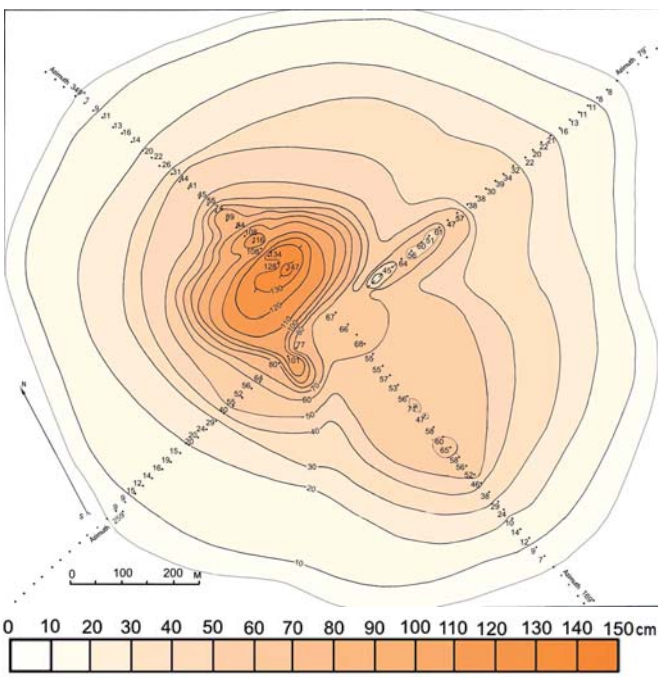


Figure 4. "Balapan" site. Borehole 1206. Difference in original surface height before and after UNE

1.2.3. Thermal changes as a result of UNE

Study results of thermal regime of underground nuclear explosions at original ground are presented in work [10, 11]. For the first time there were circle IR-spectra luminescent lines covering slab areas of underground nuclear explosions. It was determined that chamber cavities were not absolutely sealed after UNE. Technogenic impact: maximum cracking areas, collapse column and rock destruction area – make it prone for heat and gases release to the surface and in case of high temperatures in cavities and their slow cooling, this process becomes rather long. Thermal measurements on the surface had shown that by all studied UNE at the epicenter surface area there were circle and arch type thermal structures registered which cover slabbing explosion areas. In other cases, usually corresponding to low energy explosions or made in boreholes, thermal anomalies do not have strictly defined geometry and constitute separate thermal spots. Maximum radius of thermal anomaly changes from 80 to 250 meters and a thermal circle width – from 20 to 60 meters.

Review of materials on longterm thermal regime in rock blocks with center areas of UNE shows that for many years they have high internal temperature slow decreasing in time [12, 13]. Detailed studies of temperature fields performed for 1.7 kiloton Rainer explosion (USA) show that maximum temperature of a cavity within half a year was 94 °C, and during Rulison explosion – cavity temperature decrease only to 290 °C within 6 years [14]. Thus UNE cavity are a longterm storage and sources of thermal power.

1.3. Processes of underground gasification and thermal destruction of carbon containing rock

All coil mainly consists of carbon (60–90 %), hydrogen (<4–8 %) and oxygen (2–30 %); there are also small quantities of nitrogen (0.5–2 %) and sulfur (0.5–3 %). At higher temperatures coil and also coil containing rock are subject to thermal destruction, pyrolysis. Gaseous products composition in the process in pyrolysis or thermal destruction of coil in case of absence of any reagents including oxygen, significantly depends on coil particles heating and temperature.

Thermal transformation of coil starts at temperature around 200 °C. Although at reaching 120 °C fixed moisture and absorbed gases (carbon dioxide, methane etc.) begin releasing. With that there is no obvious digestion of coil mass, although some changes in internal structure are possible. At temperatures more than 200 °C some water is released which occurs under thermal digestion of organic coil mass and also carbon oxide and dioxide. This is a result of rather complicated chemical processes. Within 250–325 °C range coil digestion is more intensive. There is in intensive release of water steam, carbon oxides, some hydrogen sulfur and organic sulfur compounds. Deep digestion of coil organic mass, release of usually liquid substances (tare) is completed at 550 °C. At high temperature transformation (up to 850 °C) gas composition changes: methane and high molecular hydrocarbons decrease with significant increase of hydrogen, HCN occurs in gas. This way, in pyrolitic digestion of carbon containing materials we should expect release of [15].

Other process, which may lead to formation of gases is a process of gasification of carbon containing materials, transformation of fossil fuel (coil, peat, shale) into gases at high temperature in presence of oxidizers (gasifying agents), which can also be water. At the same time there can be many other reactions, which may be presented in the following way [16]:

$$(C, C_nH_{2n}) + H_2O \rightarrow CO, CO_2, CH_4, H_2$$

It is necessary to consider possibility of secondary reactions, like:

$$(CH_4, CO) + (H_2, H_2O) \rightarrow CO_2, CH_4, H_2O, H_2$$

With that some of thes reactions are exothermal, which may lead to increase of temperature in reaction amount.

This way, combination of the following conditions: presence of carbon containing materials, high temperatures and presence of water are factors contributing to occurrence non-oxygen destruction of rock. In other words in such conditions we witness a process of partial or not full gasification of carbon containing rock without access to oxygen in combination with pyrolitical processes which may lead to generation of the following gases: CO, CO_2, H_2, CH_4 etc., some of which are flammable (CO, H_2, CH_4).

2. DETECTION OF "CRITICAL" BOREHOLES ON THE BASIS OF ARCHIVE DATA AND SURVEY DATA

Collection of data on boreholes at "Balapan" site in literature and archive sources allowed creating their full catalog and construct their layout. Altogether the site has 105 warfare and 13 unused boreholes. Most of underground nuclear test were performed from 1965 to 1989 in boreholes of bigger diameter and 400–600 meters. Besides warfare boreholes there were more than 200 shuttle boreholes drilled (structural, instrumental, hydrogeologic, testing etc.).

Along with study on warfare boreholes, there were data collected and processed on primary geophysics surface and borehole data performed nearby and directly in warfare and structural boreholes at "Balapan" site. Using this data there was a layout of site constructed using surface geodesic methods with 1:50,000. Prior to explosion state of geological media and post explosion deformations in the area of warfare boreholes were studied by field team № 27 of Second Hydrological Department of the Ministry of Geology of USSR. During period from 1964 till 1974 there were geophysical work performed in the scale 1:50,000 which allowed building STS map with scale 1:200,000. After 1974 there was a study performed on central area of UNE on warfare boreholes with 0.3–5 km radius using complex geophysics work. Usually measurements were made before and after explosions which allowed registering deformation adjoining UNE epicenter and also with structural boundaries (fractures, lithological contacts, etc.). Partially they were traced at original surface (soil blistering, depressions, cracks of different intensity and length).

Warfare boreholes inventory. In order to perform survey to clarify conditions of warfare borehole wellheads at "Balapan" site there was warfare boreholes inventory performed. Catalog and layout of 106 warfare boreholes developed upon results of data collection and processing were used as baseline data (section 1). Survey included physical search of warfare boreholes, taking wellhead coordinates (using GARMIN GPS+), taking pictures of wellheads and visually noticeable relief peculiarities near a borehole. Upon results of survey of 106 wellheads of boreholes there was a catalog of coordinates developed and 1:50,000 scale map.

2.1. Instrumentation and survey methods

2.1.1. Instrumentation and methods of original surface changes survey

At the initial stage there was a preliminary survey performed at all warfare borehole at "Balapan" site. Survey included boreholes search, pictures of wellheads and peculiarities of relief, description and evaluation of registered craters. Work was performed at 106 boreholes

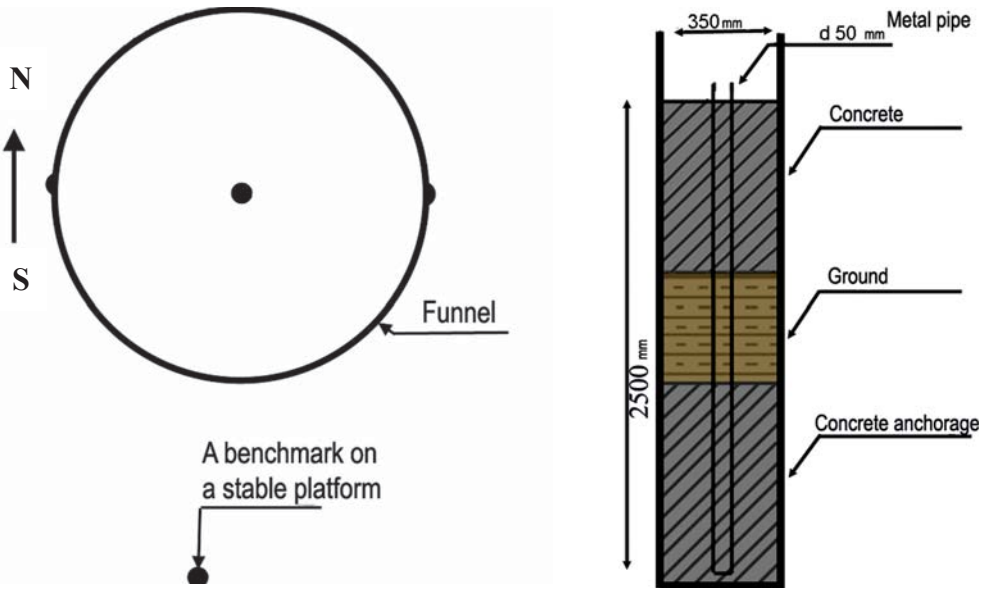
of "Balapan" site. Pictures of original ground near boreholes were made as the same angle from southern side of sites. Descriptions and measurements determined list of boreholes with depression which is presented in Table 1.

Table 1.

"Balapan" site. List of boreholes with craters on and close to 9 wellhead

#	Borehole #	Power, kiloton	Crater diameter, m		Crater depth, m	
			2005	2008	2005	2008
1	1004	140	>200	500		
2	1066	212	~200	250		17
3	1069	15	~10	8		4.5
4	1203	20	>50	50		Up to 30
5	1204	140	~200	300	25-30	15-17
6	1207	140	~200	250	25-30	15
7	1226	180	~150	200	~15	20
8	1301	3	~10	10		6
9	1414	150	~10	10		3
10	"Glubokaya"	70	>200	250		15
11	1355	130	-	20	-	8

To monitor craters dynamics at three boreholes – 1207, 1066 and 1414 – there were long term reference points installed. Scheme for reference points location and their design is shown at (Figure 5, a, b).



a b

Figure 5. "Balapan" site. Borehole 1414. Topographic monitoring of deppresions:
a – reference points layout; b – design

In all cases two reference points were placed at the edges of craters and one – at the bottom. At boreholes 1207 and 1066 distance between reference points is about 250 meter and depth of a central reference point is 16 and 17 meters and at borehole 1414 – 6 and 3 meters. Year later topographic monitoring began at these boreholes.

At boreholes 1053, 1066, 1207, 1236, 1222, 1414 at the sites of 1,000x1,000 size there was a tacheometric survey performed with scale 1:2,000. At all sites there was layout geodesics survey network created as an independent geodesic basis. Network is created by a polar method; length of polar sector did not exceed 1000 meters. Geodesic survey points are marked by temporary marks – metallic pins of 40 cm height. Measurements performed by electronic tachometer SOKKIA SET 510 #33746. Angle measurement preciseness is 5″ (ISO 12857-2:1997). Distance measurement preciseness ±(2mm+2ppm x D, where D – distance measured in kilometers). Coordinates and height points of survey justification were received in 1985 coordinates system. Height marks system is Baltic. Average error of objects and contours on engeneering topographic plan do not exceed 0.5 mm in plan scale. Topographic plans were built using AutoCAD 2005 and software complex GeoniCS Topoplan v. 5.1.

2.1.2. Instrumentation and methods of work on gas composition detection

Determination of methane, carbon dioxide content in soil and atmosphere was made with an interferential gas analyzer IGA. Instrument operation is based on measurement of a shift of interferential picture due to changes in tested air sample which is on the way of one of the two beams capable of interfere. Different gases concentration limits are the following: CH_4 – from 0 to 6 %; CO_2 – from 0 to 6 %; O_2 – from 5 to 20.9 % (by volume). Device gives accurate readings on ambient temperature from +10 to +30 °C and atmospheric pressure from 720 to 800 torr.

Hydrogen sulphide (H_2S), carbon oxide (CO) detection was made by gas analyzer ANKAT-7664M, used for continuous automatic measurement of volume share of oxygen, carbon oxide II, Hydrogen sulphide, concentrated total of paraffin hydrocarbon (Ex). Sampling method is diffuse or forced, forced sampling is provided by means of fitted circulator of by mean of rubber bellows.

For detection of carbon dioxide, sulfur anhydride, indicator tubes method was used [17]. Essence of a method is formation of colour reaction of detected substances in air sample during its movement through an indicator tube with reagent applied on a hard surface. Length of coloured column of indicator powder in the tube is proportional to the concentration of analyzed gas in the air and is measured according to the scale divided in mg/m^3. Measuring system consists of an indicator tube and universal gas analyzer UG-2 performing a function of a pump. According to the passport data, tube design provides colouring length not less than 15 mm at maximum concentration of detected substances in a measuring range and had preciseness not less than ± 25 %. Operation principle of an universal analyzer UG-2 is based on pumping air with admixture through an indicator tube with air sampling device.

Detection of hydrogen content in gas sample was performed using mobile gas analyzer VTI-2 [18], it's action is based on a fraction burnup of an oxygen over copper oxide with temperature limits 260–270 °C at a speed of gas in the furnace tube of 20 ml/min. Metrological parameters of instruments and methods used for detection of gas content are given in Table 2.

Table 2.

Metrological parameters of instruments and methods used fir detection of gas composition

Measuring channel	Physical value unit	Measurement range	Segment of a measurement range where absolute error is determined	Allowable main, absolute error Δ_o
O_2	Volume share, %	0–30	All range	± 0,9
E_x	%, LCLD	0–50	All range	± 5
CO	мг/м^3	0–200	0–20	± 5
			20–200	± (5+0.25*($C_{вх}$–20))
H_2S	mg/m^3	0–40	0–10	± 2.5
			10–40	± (2.5+0.25*(C_{in}–10))
CH_4	Volume share, %	0–6		
Remark: LCLD – lower concentration limit of distribution (flame)				

Sampling of soil air. Sampling of soil air was performed by means of storage capacitors (polyethylene 10 liter canisters with hose for air sampling). Tightness is provided by embedding the tank to 5-7 cm and compression of soil adjoining to its walls. Duration of gas collection was not less than one day. After one day gas was sampled from the tank.

For transportation, storage of soil air, glass aspirator was used. Operational principle of this instrument is based on pushing gas out by means of water in a compensating tube [19]. Gas sampling into a glass aspirator from borehole is presented on the Figure 6).

Figure 6. "Balapan" site. Gas sampling from a borehole.

2.2. RESULTS

2.2.1. Results of initial survey of "Balapan" site boreholes

In 2008 at some boreholes with coil containing rock in the area of UNE hypocenter and where potentially destructive processes were registered, there were long term reference points installed and tacheometric survey was performed. Below are the results of topographic survey results are presented for some boreholes.

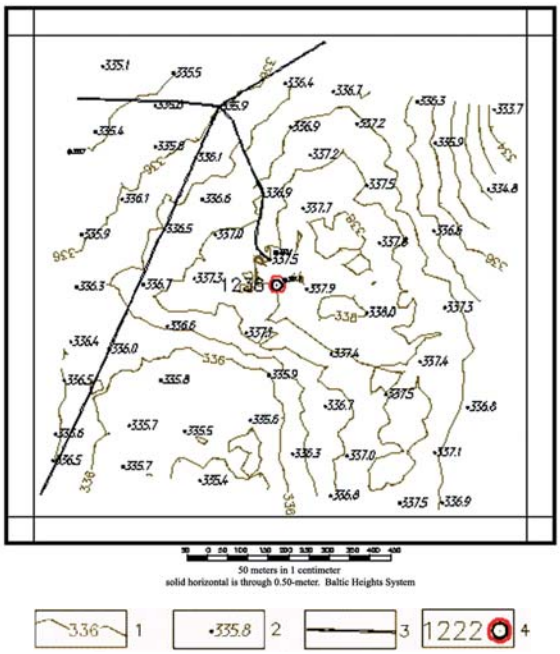
2.2.1.1. Topographic survey results

Borehole 1236. Borehole is located in the central part of the site and in the area with highest test density. Current view of wellhead area is presented at the picture (Figure 7).

a b c

Figure 7. "Balapan" site. Warfare borehole 1236: a – wellhead; b – south view; c – surface depression from wellhead side

Earlier, during initial survey in 2005 and 2008 there were post explosion surface changes registered, mainly to the west of wellhead, in the form of blistering of depressions and cracks with fluctuations of height marks up to 1.5 meters (Figure 7 c). Results of tachometric survey were presented in Figure 8.



1 – isoline of height marks, m; 2 – reference point in connection with height mark; 3 – gravel road; 4 – borehole and its number

Figure 8. "Balapan" site. Warfare borehole 1236. Tachometry results at near the wellhead site (out of scale copy of a map with 1:2,000 scale)

Central part of the site with wellhead of a borehole 1236 is a small height with weak slope to the north-east. Absolute marks are measured with 336.5–333.7 m limits. Maximal values of absolute marks 338.0 m (archive data say – 336 m [17]). Tachometry data shows that absolute mark of a wellhead is 337.7 m (data of the year 1982 – 335 m). According to the existing materials of 1982 and 2009. (received by use of instruments of different preciseness type and with reference to different trigonometry marks) there were meridian profiles (Figure 9a) and gradient schedule of absolute values of height marks constructed (Figure 9, b).

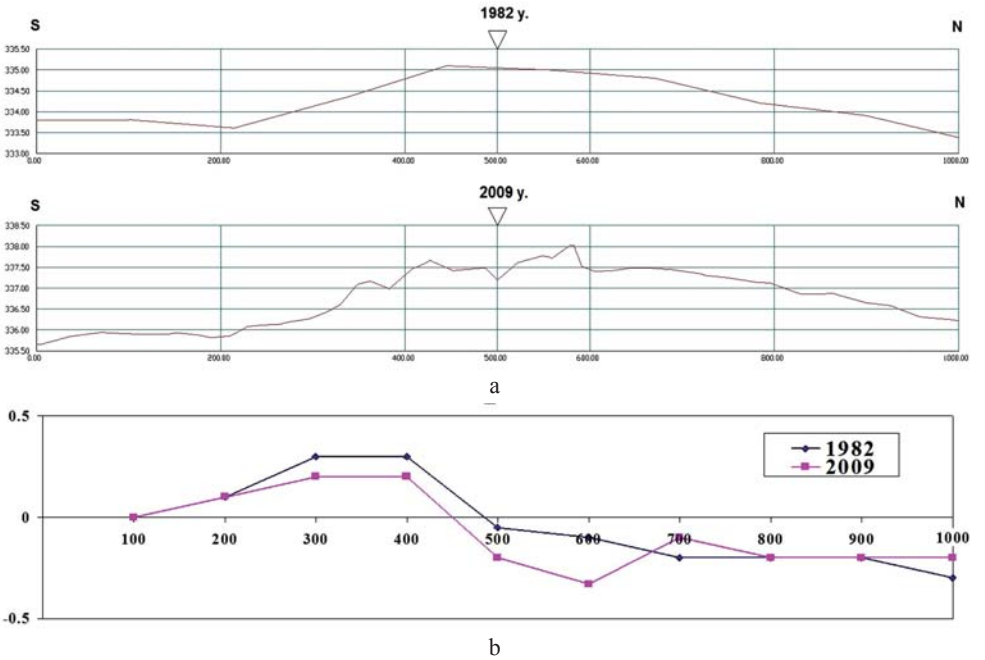


Figure 9. "Balapan" site. Borehole 1236. Relief of wellhead area of the site: a – absolute marks, m; b – gradient of height marks, m

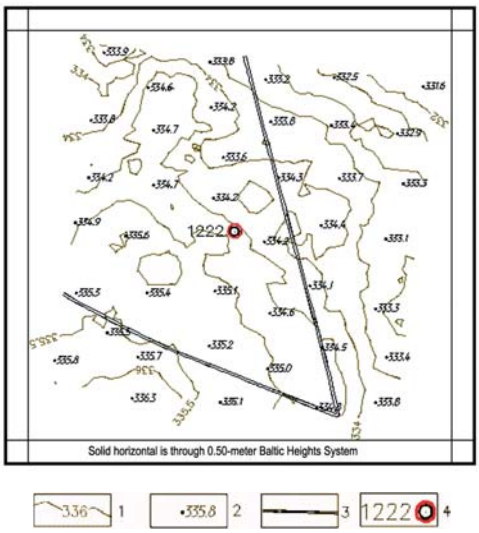
It may be seen that during the period beginning from 1982 there have been a depression of original ground right around wellhead of a warfare borehole in the radius of 200 - 250 m.

Borehole 1222. Warfare borehole 1222 is located practically at the center of "Balapan" site in the area with the highest test density. Initial surveys in borehole1222 were performed in 2005, 2008 and 2009. Original surface is slightly wavy, differences in absolute marks up to 0,5 m. Modern condition of wellhead area is presented on picture 10 – casing has been removed (ditch has 4 meter depth and 8 meter diameter).

a b

Picture 10. "Balapan" site. Warfare borehole 1222 (2009): a – wellhead: b – general view

Picture 11 shows results of tachometry data which demonstrate that wellhead has a wavy surface and a weak slope to the north-east. Height marks are measured in the range 336.3–331.6 meters. There are some lower areas in relief.



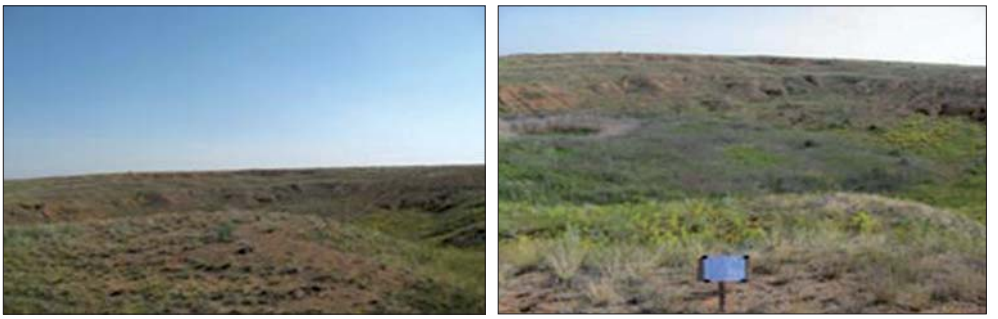
1 – relief isoline, meters; 2 – control point and its height mark, m;
3 – gravel road; 4 – borehole and its number

Figure 11. "Balapan" site. Warfare borehole 1222. Results of tachometry at wellhead site (no scale copy of a map with 1:2,000 scale)

Borehole 1207. Borehole 1207 is located in the center of "Balapan" site. Initial surveys at a borehole 1207 were performed in 2005, 2008 and 2009. Modern state of wellhead is shown at picture 25. Original ground significantly changed after explosion. A crater was formed with 15 meters depth and 250 meter diameter.

Results of tachometry show that borehole area is a flat plain with absolute heights from 329.9 to 325.6 m, with some slope from south-west to north-east (Figure 12). At the north relief is complicated by technogenic forms in shape of caponiers and pits which were

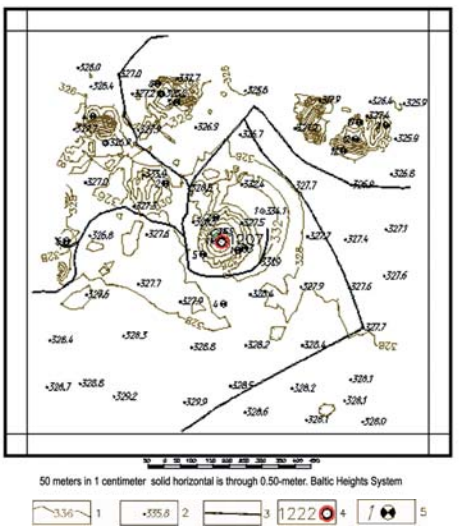
formed as a result of liquidation of military infrastructure of a military complex at STS. At the center of the site relief is significantly changed. There is a crater from a nuclear explosion. Tachometry data provides the following dimensions of the crater – (150–200) × (150–250) m (within crater limits isolines are rare), crater depth is 12 m (marks 332–320 m). Total deformations area – 300×450 m. At the north-west from the crater there is a significant blistered are, maximum height of which 334.1 m.



a b

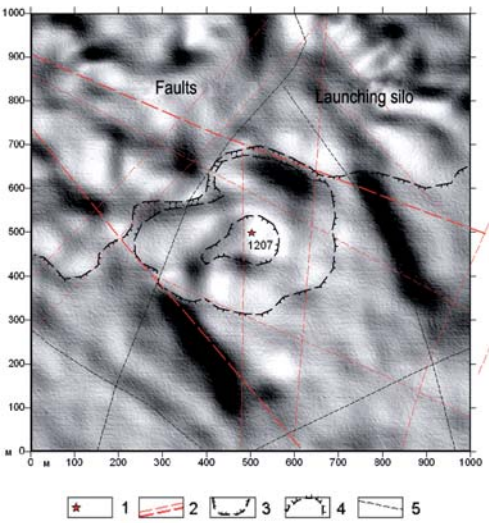
Figure 12. "Balapan" site. Warfare borehole 1207 (2009): a – view of site from the south; b – view the borehole from the south

Detailed half tone panchromatic shot (Figure 13, b) demonstrates oval border of ground crater of 350×450 m. Size of a central crater on the plan is - 100×120 m, depth - up to 10 m.



1 – isolines of equal value, m; 2 – reference points and height marks, m; 3 – gravel roads; 4 – borehole and its number; 5 – constant reference point GR type

a



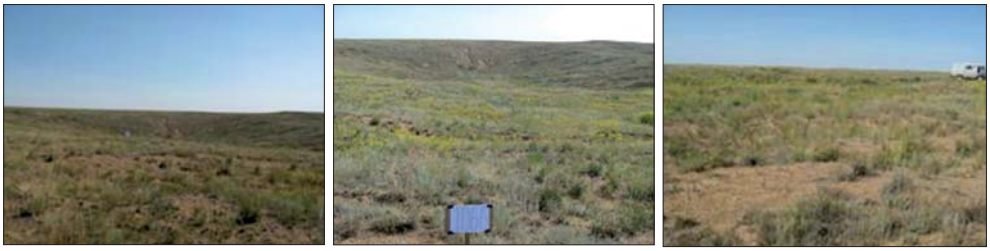
1 – warfare borehole; 2 – tectonic cracks and faults; 3 – crater outline; 4 – relief elevation contour and main rock ejection; 5 – old dirt roads

b

Figure 13. "Balapan" site. Warfare borehole 1207:
a – tachometry results (out of scale copy of a map in 1:2000 scale);
b – pseudo relief view of borehole site with elements of geo morphological interpretation

In 2008 in the area of borehole 1207 log term reference points were installed, 2-GR type, for geo morphological monitoring (Figure 13). In November 2009 there was a local leveling network was created in a form of an enclosed test site and II class leveling has been performed.

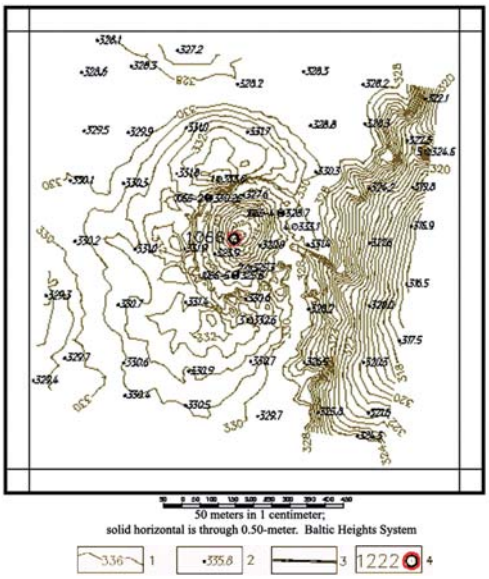
Borehole 1066. Warfare borehole 1066 is located at the center of "Balapan" site. The most powerful explosion at "Balapan" site was made in this borehole. Initial survey at borehole 1066 was performed in 2005, 2008 and 2009. Modern state of wellhead area is shown on the picture (Figure 14).



a b c

Figure 14. "Balapan" site. Warfare borehole 1066 (2009): a – general view from the south; b – wellhead view; c – depression and blistered soil 100-150 m to the south from the crater

Crater is filled with water (200–250 m diameter, depth – up to 17 m). Surface deformation is registered up to 500 meters radius from a warfare borehole. Relief from the south side is significantly changed. Continuous depression and soil blisters are registered 100-150 meters south to the wellhead. Results of tachometry are presented on the Figure (Figure 15).



1 – isolines of equal values, m; 2 – reference points and height points values, m; 3 – gravel roads; 4 – borehole and its number; 5 – full time reference points, GR type

Figure 15. "Balapan" site. Warfare borehole 1066: a – tachometry results at wellhead area (out of scale copy of the map with 1:2,000 scale)

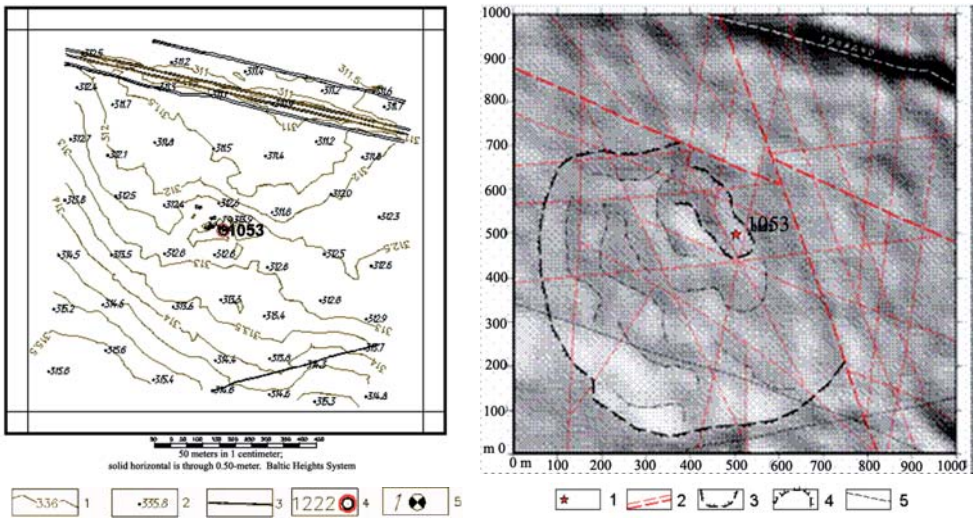
Survey results show a plain with absolute points from 329.7 to 327.2 m, with small slope from the south to the north (Figure 15). At the east, relief is complicated by a sudden slope (329.2–316.5 m) At the center of the site the relief is significantly changed. Here there is a depressed crater 200×250 m size (isolines are rare within crater limits). Crater depth is 21 m. Total area of deformations – (550–600)×(700–800) m. At the north, east and south of this big area of deformations there is a significant blistered area. To the south-west of a crater there smaller relief forms in a form of depressions and ground blisters. In 2008 there were long term reference points, 2-GR type installed for systematic geo morphological control.

Borehole 1053. Warfare borehole 1053 is located at the east of "Balapan" site. From presented data, it is clear that an explosion was made at an average depth and was not accompanied by steam-gas products release into the atmosphere. Initial surveys at borehole 1053 were performed in 2005, 2008 and 2009. Modern view of wellhead area is presented at the Figure (Figure 16).

a b

Figure 16. "Balapan" site. Borehole 1053: a – general view from the south; b – wellhead, 2009 г.

In 2005 it was noticed that the surface did not have any technogenic changes. In 2008 there was a depression of original ground registered at around 3 meter depth in relation to the rest of relief.



a b

Figure 17. "Balapan" site. Warfare borehole 1053: a – tachometry results at wellhead area (out of scale cope of as map, 1:2,000 scale);
b – pseudo relief picture of a borehole site with elements of geo-morphological interpretation

Tachometry results confirmed that relief has a slightly wavy surface with smooth slope from south-west to north-east. At the north it is complicated by an earth road which has dirt roads running along on both sides. At the central part there is a smooth depression which occurrence has to do with UNE made in a borehole 1053. Here differences of absolute points are 2.5 m (from 314,5 to 312 m).

According to interpreted data from satellite pictures (Figure 17 b) borehole site is located in the intensively fractured zone. That is possible why contour relief elevation has a significantly stretched size (650×650 m). From north-east, contour is limited by two tectonic cracks of north-west direction. Inside this contour there is wavy localized blistering of the surface. Right close to the borehole there is a well seen contour of relief depression of a stretched form from north-west of 50×200 m.

Borehole 1414. Borehole 1414 is at the east of "Balapan" site, at the center of boreholes cluster (1087, 1058, 1053, 1083 и 1075). Modern view of wellhead area is shown on the picture (Figure 18).

a b

Figure 18. "Balapan" site. Warfare borehole 1414 (2009): a – wellhead; b – general view from the south

Initial surveys of borehole 1414 were performed in 2005, 2008 and 2009. Post explosion changes of original surface were registered in a form of blistered depressions and cracks in the radius of 200–300 m from wellhead of the borehole. % meters from the wellhead there is a depressed crater filled with water, 3 meters deep [20].

This way, results of topographic work in the epicenters of warfare boreholes show theta as a result of UNE around many of boreholes circled post explosion structures were formed and also other deformations of the ground surface. At that technogenic relief elements (banks, terraces, etc.) reach kilometers in length and several meters on height. In wellhead part of warfare boreholes there depressed crates up to 30 meters diameter and several meters depth. Above mentioned facts show that processed which have to do with deformation of surface in epicenters of warfare boreholes are not complete yet.

2.2.1.2. Gas survey of warfare boreholes

Initial survey of epicenter zones of warfare boreholes was performed to reveal in soil air the presence of gases typical for underground gasification processes in coil contacting rock. Study of wellhead areas was made within 50 m diameter. Sampling was made at several points, close to and at some distance from wellheads of boreholes. In 2008 105 warfare boreholes were surveyed in regard to gasification at Bakapan site where UNE were made. Table (Table 3) has general data on air content at wellhead area of boreholes.

Data presented in the table on rock containing central zones of UNE, it can be mentioned that for boreholes which cavities are in sediment rock, gasification process occurrence has no doubt, but for boreholes in tuffs of andesite porphyrite (1361, 1234, 1303, 1319, 1071). One of explanation for rock gasification at mentioned boreholes may be that in tuffs widespread at "Balapan" site usually have inclusions (xenolith) represented by coaly and siltstone.

This way, upon results of initial survey at the "Balapan" site, there have been several borehole found with higher background methane concentrations (boreholes 1234, 1207),

carbon oxide (boreholes 1317, 1303, 1228, 1066, 1267, 1236, 1086, 1326, 1319, 1207, 1222, "Glubokaya"), carbon dioxide (boreholes 1067, 1303, 1228), hydrogen sulfur (boreholes 1326, 1080), sulfurous anhydride (boreholes 1071, 1326, 1317), hydrocarbons (boreholes 1317, 1010). Maximum concentrations of all measured gases were found in borehole 1010 [20].

It should be mentioned that in all presented data on UNE there are no any specific factors impacting occurrence of ground depression and gasification. Thus in most of UNE higher than 100 kilotons performed in sediment rock, there was no ground depression or soil gasification registered. From the other side in the range of determined gas releasing boreholes (1067, 1071, 1080, 1234, 1267,1317,1319, "Glubokaya"), there were UNE performed less than 100 kilotons. With that practically in all boreholes there is presence of ground water which in most cases is pressure water and located at 5,8 to 34 m. Presence of loose sediments mantle does not have any significant impact.

May be one of main factor impacting presence of obvious and hidden signs of ground gasification is peculiarities of structural tectonic structure of the rock containing central areas of UNE at "Balapan" site. It seems that territory of the site is a complex tectonic magmatic center for intrusions development and long term faults. It is connecting a range of regional north-west faults and big meridian and north-east faults. Introduction of granite type inter fault intrusions promoted loosening of hercyian rock with formation of weak areas. As a result, complex of factors combining peculiarities of geological structure of the territory and results of underground nuclear explosions contributed to the formation of complex geological physical conditions for occurrence of ground depression and rock gasification. This statement allows to suggest occurrence of gasification process in all boreholes drilled in sediment rock and to less extent, in tuffs of andesite porphyries. In this context there is probability of explaining possibility of ground gasification processes in a catastrophic manifestation. The fact is that presence of tectonic defects within limits of UNE areas contributes to formation of gas transferring zones. In this case, conditions are created for slow pressure release of central areas from generated gases. In more monolithic rock, it is likely that episodic ("volley") release of gases occurs due to pressure growth. One of obvious facts of "volley" release of gases is an above described cases at "Glubokaya" borehole.

Table 3.

Characteristics of "Balapan" site boreholes according to archive data and results of initial survey

№ borehole	C_nH_{2n}, mg/m^3	CO mg/m^3	CO_2 mg/m^3	H_2 mg/m^3	SO_2, mg/m^3	H_2S, mg/m^3	CH_4 mg/m^3	Explosion date	Explosion power, kilotons	Charge depth , m	Cavity radius, m	Ground water level, m	Rock	Loose rock thickness, m	Ground depression
1005	none	none	none		none	none	none	16.10.1974	43	535	24.5	21.4	Alternation of post and slate layers	55	none
1006	none	none	none		none	none	none	01.02.1979	18	309	18.3	12	Alternation of conglomerates and post	20	none
1007	none	none	none		none	none	none	10.02.1972	16	295	17.6	9.5	Post	20	Depression, 15 m from the wellhead
1010	850	47	574.5	8.3	92.07	303.6	7.14	11.06.1978	60	556	27.4	13.2	Alternation of post and siltstone	58	none
1050	none	none	none		none	none	none	29.07.1974	15	535	17.3	30	Porphyrines at 405 m	88,8	none
1053	none	none	1.94	0.31	57.1	none	none	19.06.1968	22	316	19.6	none	Post		none
1054	none	none	none		none	none	none	30.11.1969	160	472	38	11.5	Conglomerate at 391 m	84,6	none
1056	none	none	none		none	none	none	30.06.1971	8	217	14	none	Conglomerate	Up to 50	none
1058	none	none	none		none	none	none	27.12.1974	51	535	26	12	Conglomerate at 393 m	86.5	Small depression at wellhead

№ borehole	C_nH_{2n}, mg/m³	CO mg/m³	CO_2, mg/m³	H_2, mg/m³	SO_2, mg/m³	H_2S, mg/m³	CH_4, mg/m³	Explosion date	Explosion power, kilotons	Charge depth , m	Cavity radius, m	Ground water level, m	Rock	Loose rock thickness, m	Ground depression
1061	none	7	3.02	none	none	none	0.92	02.11.1972	165	521	38.4	11.1	Black mudstone	35	Bumpy relief, difference of absolute points up to 1 m
1062	none	none	none		none	none	none	04.07.1976	90	535	31.4	9	Post	39	Bumpy-wavy relief, difference of absolute points up to 1 m
1064	none	none	none		orc.	orc.	orc.	14.12.1973	150	478	37.2	15.8	Tuff of diabase porphyrite	33	none
1066	none	5	2.05		none	none	3.57	23.07.1973	60	535	27.4	26.7	Granite at 422 m, conglomerate at 574 m	27	Depressed crater. 17 m depth, 250 meters - diameter.
1067	none	none	7.95		none	none	none	25.12.1975	90	535	31.4	19.9	Alternation of conglomerate and post	33	none
1069	none	none	none		none	none	none	04.11.1973	15	535	17.3	16.6	Cristal litho classic tuffs of diabase porphyrite at 490 meters	26	Crater. Depth - up to 4.5 meters and 8 meter diameter.
1071	none	none	2		3.5	none	none	25.04.1980	19	330	18.7	8.4	Tuff of quartz porphyrites at 331 m	2	none
1073	none	none	none		none	none	none	12.11.1977	15	535	17.3	10.1	Post at 306 m	37.5	none
1075	none	none	none		none	none	none	09.06.1976	25	298	20.5	8.9	Tuff of andesite porphyrite	60	none

№ borehole	C_nH_{2n}, mg/m³	CO mg/m³	CO_2, mg/m³	H_2, mg/m³	SO_2, mg/m³	H_2S, mg/m³	CH_4, mg/m³	Explosion date	Explosion power, kilotons	Charge depth , m	Cavity radius, m	Ground water level, m	Rock	Loose rock thickness, m	Ground depression
1077	none	none	none		none	none	none	05.07.1978	60	445	27.4	15.4	Siltstone	29	Bumpy relief, Рельеф бугристый, difference of absolute points up to 0.5 m
1079	none	none	none		none	none	none	05.09.1977	80	535	30.2	16.5	Siltstone at 498 m	50	none
1080	none	none	none		none	none	none	29.06.1977	9	340	14.6	5.2	Conglomerate	none	none
1083	none	none	none		none	none	none	12.06.1980	37	354	23.3	13	Tuff post	65	Wavy relief, bank height up to 0.5 m
1085	none	none	none		none	none	none	04.08.1979	150	590	37.2	16	Carbonized post	40	Changed relief
1086	none	28	2.01	0.17	none	none	none	14.12.1980	100	530	32.5	14	Slate	70	Wavy relief, bank height up to 0.5 m
1087								12.10.1980	102	436	32.7	н/д	none	none	none
1201	none	none	none		none	none	none	21.04.1976	20	535	19	13.5	andesite porphyrite	30	none
1202	none	none	none		none	none	none	28.08.1976	58	535	27.1	12.5	Post at 455 m	90	Wavy relief
1203	none	none	none		none	none	none	27.05.1981	20	535	19	11.6	Siltstone at 485 m	62	Crater. Depth – up to 30 m, diameter – up to 50 m.
1204	none	none	none		none	none	none	10.12.1972	140	478	36.3	12.2	Tuff of andesite porphyrite	30	Crater. Depth – up to 300 m, diameter – up to 20 m.

№ borehole	C_nH_{2n}, mg/m³	CO mg/m³	CO_2 mg/m³	H_2 mg/m³	SO_2, mg/m³	H_2S, mg/m³	CH_4 mg/m³	Explosion date	Explosion power, kilotons	Charge depth , m	Cavity radius, m	Ground water level, m	Rock	Loose rock thickness, m	Ground depression
1205	none	none	none		none	none	none	27.04.1975	60	535	27.4	13.6	Siltstones	29	Depression to the SE from borehole wellhead, 2009
1206	none	3	none		none	none	none	29.10.1975	90	535	31.4	8.5	porphyrite	70	none
1207	none	6	none		none	none	none	31.05.1974	140	535	36.3	n/d	Conglomerate	Up to 90 m	Crater. Depth – up to 15 m, diameter – up to 250 m.
1209	none	none	none		none	none	none	07.12.1976	15	535	17.3	11.6	Microdiorite	53	none
1211	none	none	none		none	none	none	15.09.1978	80	535	30.2	15	Siltstone at 520 m	none	Bumpy relief
1214	none	none	none		none	none	none	29.10.1977	50	535	25.8	18.7	Tuff of andesite porphyrite 446 m	none	Slightly wavy
1219	none	none	none		none	none	none	25.04.1982	145	548	36.8	15.5	Slate	none	Bumpy relief
1220	none	none	none		none	none	none	14.09.1980	190	483	40.2	10.8	Plagio granite	25	Bumpy-wavy relief
1222	none	3	none		none	none	none	29.11.1978	100	535	32.5	9.3	Siltstone	none	none
1223	none	none	none		none	none	none	23.06.1979	150	490	37.2	18	Sandstone	16	none
1224	none	none	none		none	none	none	28.10.1979	140	529	36.3	12	Tuff sandstone	none	Changes in a form of banks and bumps, up to 2,5 m
1225	none	none	none		none	none	none	07.07.1979	100	469	32.5	22	Tuff of andesite porphyrite	none	Bumpy relief, differences in absolute heights is up to 1.0
1226	none	none	none		none	none	none	18.08.1979	180	535	39.5	19.8	Conglomerate	none	Crater, up to 20 m depth, diameter – up to 200 m

№ borehole	C_nH_{2n}, mg/m^3	CO mg/m^3	CO_2 mg/m^3	H_2 mg/m^3	SO_2, mg/m^3	H_2S, mg/m^3	CH_4 mg/m^3	Explosion date	Explosion power, kilotons	Charge depth , m	Cavity radius, m	Ground water level, m	Rock	Loose rock thickness, m	Ground depression
1227	none	none	none		none	none	none	29.06.1980	44	535	24.7	5.8	Siltstone	none	none
1228	none	25	8.3		none	none	none	29.08.1978	120	535	34.5	14.2	tuff	none	Significant tuff 20 m from borehole wellhead from the southern side
1232	none	none	none		none	none	none	22.04.1981	90	535	31.4	none	Lava breccia	97	Bumpy relief, differences in absolute heights is up to 1.0
1233	none	none	none		none	none	none	13.09.1981	163	531	38.2	16	Siltstone	none	Wavy-bumpy relief
1234	none	none	2.43		none	none	3.57	29.03.1981	30	433	21.8	24.5	Crystal lithoclase tuff	44	none
1235	none	none	none		none	none	none	20.11.1983	20	305	19	23.7	Tuff of andesite porphyrite	none	none
1236	none	35	2.9	0.53	1.22	none	none	18.10.1981	110	525	33.5	n/d	Siltstone	15	none
1237	none	none	none		none	none	none	29.11.1981	30	538	21.8	19.5	Sandstone	50	none
1267	none	8	1.81	0.97	59.4	none	0.74	20.09.1973	15	535	17.3	16.7	Mudstone at 372 m	58	none
1301	none	none	none		none	none	none	16.04.1974	3	535	10.1	20.5	Conglomerate at 500	40	Crater. 10 m diameter, 6 м depth.
1302	none	none	none		none	none	none	04.11.1978	45	535	24.9	15	Tuff at 500 m	45	None
1303	none	120	38.9		none	none	none	27.12.1980	100	440	32.5	19.1	Tuff of andesite porphyrite	26	none

№ borehole	C_nH_{2n}, mg/m³	CO mg/m³	CO_2 mg/m³	H_2 mg/m³	SO_2, mg/m³	H_2S, mg/m³	CH_4 mg/m³	Explosion date	Explosion power, kilotons	Charge depth , m	Cavity radius, m	Ground water level, m	Rock	Loose rock thickness, m	Ground depression
1304	none	none	none		none	none	none	07.12.1976	55	520	26.6	11	Conglomerate	none	none
1307	none	none	none		none	none	none	26.10.1983	115	528	34	18.03	Sandstone	32	Wavy relief, banks height up to 1.0 m
1308	none	none	none		none	none	none	07.03.1984	42	465	24.3	21.3	Tuff of andesite porphyrite	none	none
1309	none	none	none		none	none	none	02.12.1979	93	535	31.7	10.8	Carbon mudstone	32	none
1312	none	none	none		none	none	none	27.12.1981	156	529	37.7	13.35	Metamorphous sandstone	20	none
1313	none	none	2.85	1.6	29.64	none	3.7	16.12.1984	137	543	36.1	10.85	porphyrite	none	Wavy relief – blisters, ground depression, cracks
1314	none	none	none		none	none	none	05.12.1982	120	529	34.5	15	Orogenic Carbon siltstone	30	none
1315	none	none	0.78	0.09	none	none	none	12.03.1987	58	529	27.1	none	Carbon siltstone	20	none
1316	none	none	none		none	none	none	25.04.1984	76	540	29.7	15.3	Sandstone	19	none
1317	50	5	4.14	0.09	1	none	none	31.08.1982	8	532	14	15	Carbon siltstone	none	none
1318	none	none	none		none	none	none	03.04.1987	140	535	36.3	n/d	Sandstone	50	Wavy relief, banks height up to 1.0 m
1319	none	4	2		none	none	none	25.04.1985	74	531	29.4	29	Tuff lava	22	none
1320	none	none	none		none	none	none	12.06.1983	138	530	36,2	17,4	Siltstone	16	Wavy relief, banks height 0.5 to 1.0 m

№ borehole	C_nH_{2n}, mg/m^3	CO mg/m^3	CO_2 mg/m^3	H_2 mg/m^3	SO_2, mg/m^3	H_2S, mg/m^3	CH_4 mg/m^3	Explosion date	Explosion power, kilotons	Charge depth , m	Cavity radius, m	Ground water level, m	Rock	Loose rock thickness, m	Ground depression
1321	none	none	none		none	none	none	04.07.1982	136	526	36	17,1	Aleuropelite	none	Wavy relief, banks height 0.5 to 1.0 m
1322	none	none	none		none	none	none	20.07.1985	74	466	29.4	none	Granite	22	none
1323	none	none	none		none	none	none	27.10.1984	165	524	38.4	34	Siltstone	none	Slightly wavy relief
1325	none	none	none		none	none	none	06.10.1983	82	508	30.4	none	Sandstone	none	none
1326	none	6	5		4	3	none	20.06.1987	126	530	35.1	17,32	Siltstone	none	There is general elevation at the epicenter
1328	none	none	none		none	none	none	22.01.1989	118	535	34.3	none	none	none	none
1331	none	none	none		none	none	none	19.02.1984	50	532	25.8	15,4	Lava of andesite porphyrite	32	none
1332	none	none	none		none	none	none	15.11.1987	100	531	32.5	17,1	Granite	none	Bumpy relief, absolute height differences around 0.5 m
1335	none	none	none		none	none	none	29.03.1984	83	475	30.5	17	Andesite lava	58	Bumpy relief, absolute height differences around 0.5 m
1336	none	none	none		none	none	none	03.04.1988	135	530	35.9	none	none	none	none
1340	none	none	none		none	none	none	10.02.1985	60	529	27.4	none	none	none	Slightly wavy relief
1341	none	none	none		none	none	none	15.06.1985	115	535	34	19	Tuff sandstone	39	Bumpy relief, absolute height differences around 1 m

№ borehole	C_nH_{2n}, mg/m³	CO mg/m³	CO_2 mg/m³	H_2 mg/m³	SO_2, mg/m³	H_2S, mg/m³	CH_4 mg/m³	Explosion date	Explosion power, kilotons	Charge depth , m	Cavity radius, m	Ground water level, m	Rock	Loose rock thickness, m	Ground depression
1344	none	none	none		none	none	none	14.07.1984	135	528	35.9	none	Carbonaceous shale	none	Bumpy relief, absolute height differences around 1 m
1346								17.12.1988	68	535	28.6	10.65	Siltstone	17	none
1348	none	none	none		none	none	none	02.08.1987	100	526	32.5	16.36	Peach stone	46	Bumpy relief, absolute height differences around 1 m
1350								14.09.1988	108	642	33.3	9	Granosite	31	Wavy relief
1352	none	none	none		none	none	none	08.07.1989	22	535	19.6	17.7	Granosite	5	none
1353	none	none	none		none	none	none	28.12.1984	105	531	33	17.78	Tuff sandstone	46	Bumpy relief, absolute height differences around 0.5 m
1354	none	none	none		none	none	none	30.06.1985	86	525	30.9	none	none	none	Lowering of relief around wellhead
1355	none	none	4.09	0.71	55.6	none	3.56	13.12.1987	130	530	35.5	none	Siliceous schist	none	Crater dimensions: diameter – around 20 m, depth – 12 m
1359	none	none	none		none	none	none	04.05.1988	132	519	35.6	none	Siltstone	none	Slightly bumpy relief
1361	none	none	0.4	0.18	59.6	none	none	13.02.1988	125	527	35	19.2	Tuff sandstone	15	none
1365	none	none	none		none	none	none	19.10.1989	70	628	28.8	20	Siliceous sand at 585 m	22	none

№ borehole	C_nH_{2n}, mg/m³	CO mg/m³	CO_2 mg/m³	H_2 mg/m³	SO_2, mg/m³	H_2S, mg/m³	CH_4 mg/m³	Explosion date	Explosion power, kilotons	Charge depth , m	Cavity radius, m	Ground water level, m	Rock	Loose rock thickness, m	Ground depression
1366	none	none	none		none	none	none	12.02.1989	135	535	35.9	none	Tuff breccia	none	none
1384	none	none	none		none	none	none	17.04.1987	126	535	35.1	none	none	none	none
1388	none	none	none		none	none	none	27.12.1987	117	530	34.2	17.76	Breccia	67	Bumpy relief, absolute height differences around 1.5 m. Lowering of relief around wellhead
1400	none	none	none		none	none	none	29.05.1977	44	393	24.7	7.15	Siliceous hornstone	28	Bumpy relief, absolute height differences around 0.5 m
1410	none	none	none		none	none	none	02.09.1989	6	478	12.7	none	Tuff of andesite porphyrite	none	none
1411	none	none	none		none	none	none	02.12.1984	80	450	30.2	17.8	Tuff of crystal andesite porphyrite	n/d	Bumpy relief, absolute height differences around 2 m
1412	none	none	none		none	none	none	12.11.1988	15	395	17.3	20.6	Tuff of andesite porphyrite	24	Bumpy relief, absolute height differences around 1 m
1414	none	none	none		none	none	none	26.05.1984	150	489	37.2	12.1	Siltstone	87	Crater dimensions: diameter – around 10 m, depth – 3 m
1415	none	none	none		none	none	none	26.12.1982	42	441	24.3	25	Tuff of andesite porphyrite	20	none

№ borehole	C_nH_{2n}, mg/m^3	CO mg/m^3	CO_2 mg/m^3	H_2, mg/m^3	SO_2, mg/m^3	H_2S, mg/m^3	CH_4 mg/m^3	Explosion date	Explosion power, kilotons	Charge depth , m	Cavity radius, m	Ground water level, m	Rock	Loose rock thickness, m	Ground depression
1421	none	none	none		none	none	none	14.06.1988	4	271	11.1	none	Tuff of andesite porphyrite	35	none
1061-bis	none	none	none		none	none	none	15.06.1985	115	463	34	none	none	none	none
1207-bis	none	none	none		none	none	none	23.11.1976	74	535	29.4	19.05	Sandstone at 514 m	45	Slightly wavy relief
Glubokaya	none	none	0.78	0.81	28.8	none	none	30.11.1977	70	535	28.8	74	Conglomerate	59.7	Crater dimensions: diameter – around 15 m, depth – 250 m
Glubokaya-1	none	none	none		none	none	none	23.12.1979	137	535	36.1	none	none	none	Elevation over an epicenter, up to 1 m
A	none	none	none		none	none	none	30.06.1975	4	535	11.1	none	Dark grey siltstone	56	none
Remark: none – no data															

2.2.2. Evaluation of gas composition stability in "critical" boreholes

With the purpose of studying of rock gasification processes dynamics there was a gas monitoring performed in 2009 at "critical" boreholes (1010, 1355, 1086, 1236, 1061, 1267,1053, 1313) [20]. For air sampling there were monitoring wells drilled and equipped. Monitoring was performed from August to November 2009. Air samples for determining of gas compositions and concentration were taken at three points by warfare boreholes well-heads: right in a monitoring well, from storage capacitor and from atmospheric air in the place of where storage capacitor is stationed.

Results of gas monitoring at the borehole 1010 are presented at the picture (Figure 19).

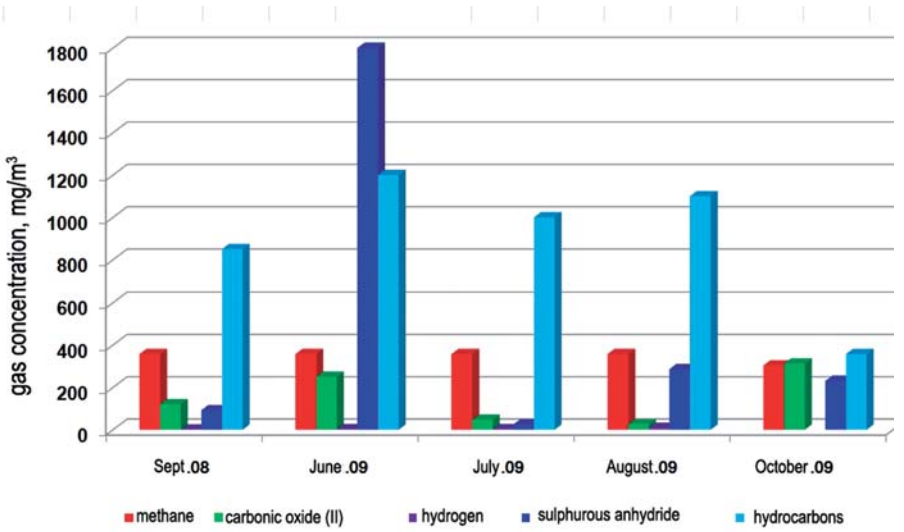
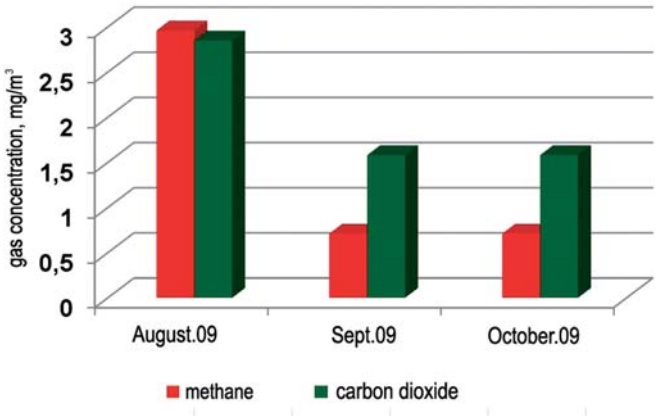


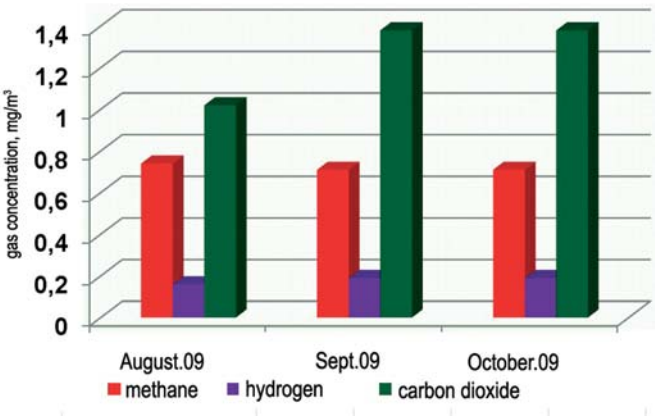
Figure 19. Result of gas monitoring at the borehole 1010

During gas monitoring the following features were registered in the dynamics of gas concentration. From June to October there has been relative stability of hydrocarbons in ground air registered, in October concentration significantly dropped down from 1,100 to 357 mg/m^3. In July there have been abrupt decrease of sulfurous anhydride concentration from 1,800 to 25 mg/m^3, further in August there have been significant increase up to 286 mg/m^3 and October – again insignificant concentration decrease to 230 mg/m^3. In other gases dynamics (methane, carbon oxide, carbon dioxide and hydrogen) there have not been any significant changes. There was some increase and some decrease of concentration in ground air.

Monitoring results show that in most of boreholes gas environment is stable. In the picture (Figure 20) there are gas monitoring results presented for boreholes 1313 and 1267 as an example.



a



b

Figure 20. Gas monitoring results for boreholes: a) 1313, b) 1267

There are some insignificant decreases and increases of gas concentration registered. There haven't been any patterns found in dynamics of gas mixture composition, which means that were no typical established signs of increase or decrease of rock gasification established.

2.2.3. Study of rock gas egress to an original ground

In order to clarified features for rock gas egress to an original ground there have been gas survey performed at wellheads of boreholes 1010 and 1355 [20]. There were methane and carbon dioxide concentrations measured at survey points. These gases were chosen because they are more typical for coal containing rock burning and these gases concentrations significantly higher that background values around borehole 1010.

Borehole 1010. Survey results show that methane concentrations do not increase background values in surveyed area. Analysis results on carbon dioxide concentration at monitoring points are shown at the pictures (Figure 21).

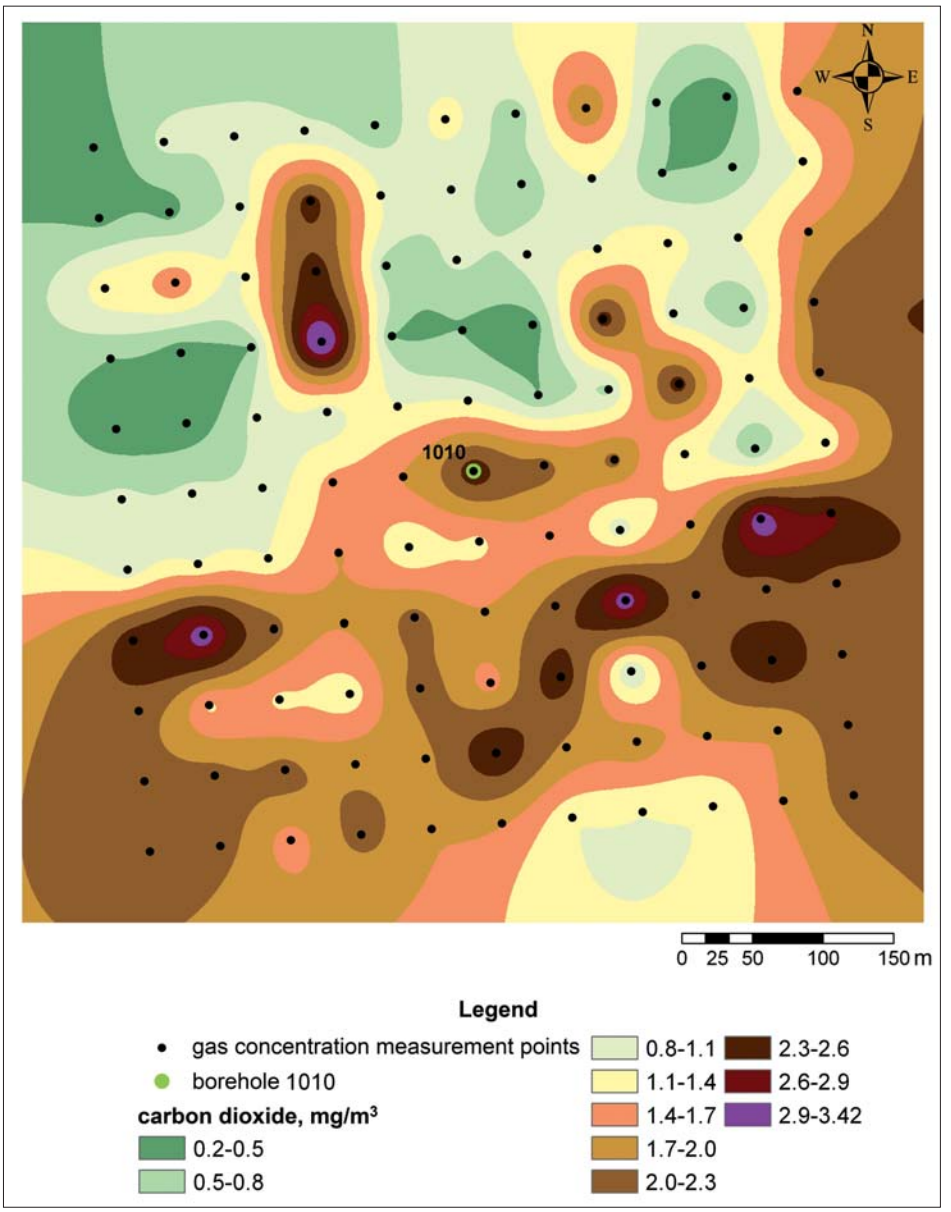


Figure 21. Carbon dioxide concentration distribution around borehole 1010 area

Data presented in the scheme demonstrates that higher CO_2 concentration area is located closer to UNE epicenter from its southern side. Within this area, from south-east and south-west areas of the site, two zones with higher CO_2 concentrations are clearly seen. This area may be characterized as local permeable structures with higher gas transfer. This estimate is supported by results of geophysical study (Figure 22)

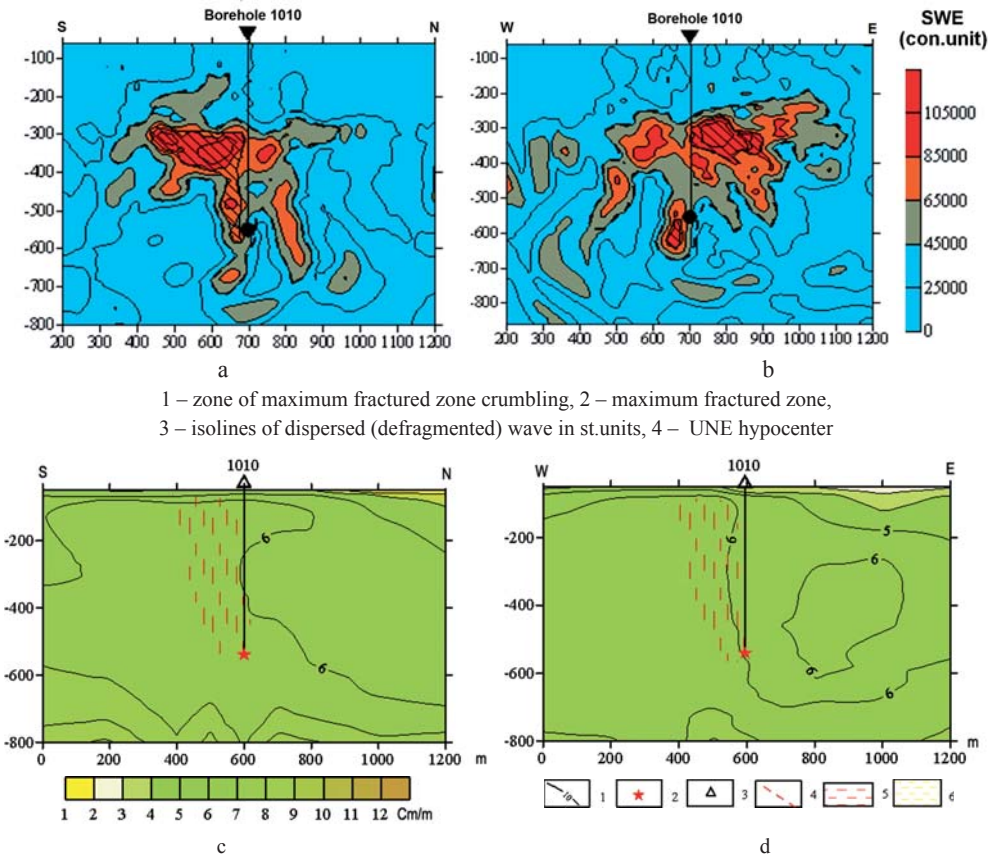


Figure 22. Results of geophysics studies on borehole 1010. Vertical cross section of dispersed (defragmented) waves according to MDSW data: a – meridian; b – latitudinal. Geoelectric cross section of seemingly lengthwise electric conductivity according to NFS: data – meridian; d – latitudial

Borehole 1010 profiles is mady up from alternating sediment rosk of Koyandin measure of low coil age – sandstone and coily-clay schis. Seismic data shows that most crumbling and fractured rock area is mostly located at south-east area and to less extent – at south-west area of the site which, in general, agrees with configuration of higher CO_2 values. Drilling data shows that in this interval (300 – 450 m), the highest tectonic activity was registered which means that UNE was made in uneven rock layer with destructive impact on weakened layers located higher than its epicenter. Reliability of this interpretation is significantly supported by electric survey data. Geoelectric situation by borehole № 1010 also demonstrate presence of a structure, characterized by higher lengthwise conductivity. It is actually a weakened area which occurred at southern and eastern directions. Above described conditions provide basis for assumption that created weakened areas are main permeable structures for gas and can be local area for rock gasification.

Borehole 1355. Nature of carbon dioxide and methane distribution at the borehole 1355 (Figure 23) [21].

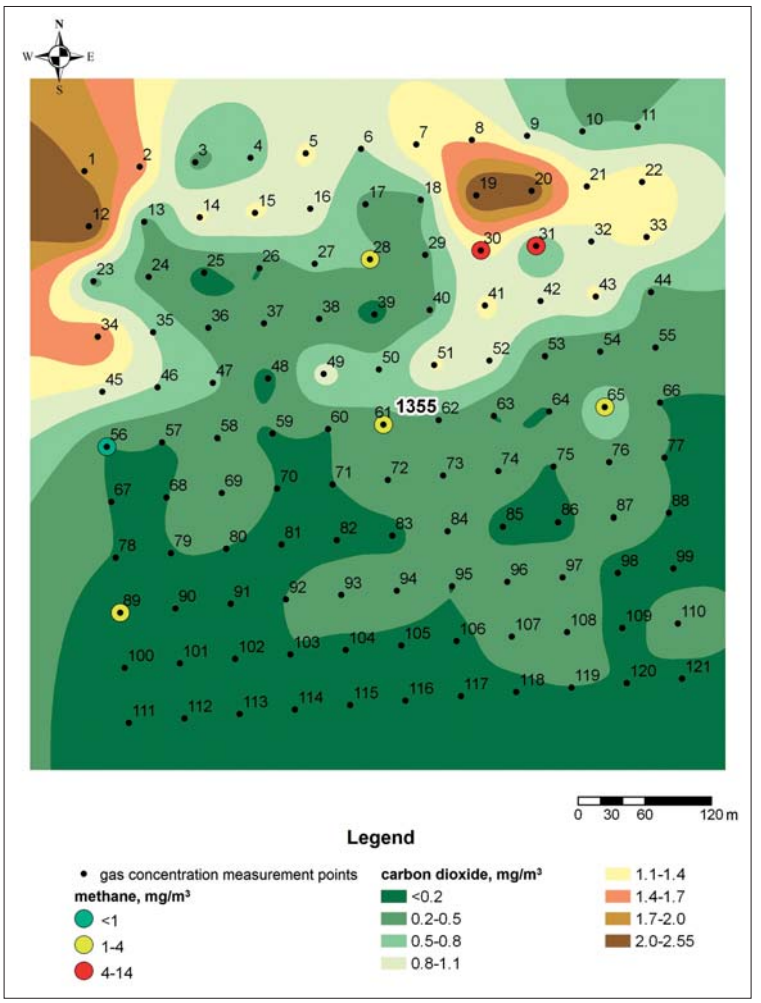


Figure 23. Borehole 1355. Areal distribution of carbon dioxide and methane

Data presented at the scheme demonstrates higher concentrations of carbon dioxide at the area close to UNE epicenter from the north side. Within this area there are two zones with higher concentration of this gas. Distribution of methane in this area has a local nature (Figure 23). Only at 7 points there have been quantitative values receive. With that, maximum concentration, up to 14 mg/m^3 were estimated within north-eastern side with higher carbon dioxide values. This way, these areas can be characterized as locally permeable structures with higher gas transfer.

Tritium. Besides above mentioned gases, there was tritium presence determined in ground and atmospheric air at some critical boreholes. [21]. Sampling methodology uses ca-

pability of water steam present I air to condense on radiator plates cooled by liquid nitrogen to condensation point. This way, if radiator plate temperature is significantly lower than 0°C, than water steam contact with plate surface leads to crystallization. After completion of freezing process of sufficient water steam amount, radiator with plates was removed from Dewar vessel and placed into the Petri dish. After melting, sample of not less than 5 ml was placed into a flask, sealed and transported for tritium detection. Detection of tritium concentration in water steam samples was made at liquid scintillation counter "TriCarb-2900 TR". Lab analyses are presented in table 4.

Table 4.

Tritium content in soil ground and atmospheric air

Sampling point	Sampling date	^3H, Bq/m^3		$^3H_{ground}$ / $^3H_{atm}$
		soil	atmospheric	
1010/1	15.09.2009	0.4 ± 0.04	0.2 ± 0.02	2
1313/1	15.09.2009	1.8 ± 0.1	1.4 ± 0.1	1.29
1267/1	16.09.2009	4.6 ± 0.4	0.6 ± 0.06	7.67
1236/1	16.09.2009	0.1 ± 0.01	0.35 ± 0.03	0.29
1086/1	16.09.2009	0.9 ± 0.09	0.3 ± 0.03	3
1061/1	16.09.2009	0.8 ± 0.08	0.2 ± 0.02	4
1355/1	17.09.2009	2.6 ± 0.2	2.1 ± 0.2	1.24

Based on presented data, it may be mentioned that at all boreholes except for borehole 1236, tritium concentration in ground air is higher than in atmospheric air. It is likely that tritium concentration in air depends on UNE specifics and degree of rock massif deformation since there is no mentioned dependence on loose deposits thickness. It is necessary to continue studying the tritium egress from central UNE areas up to day surface since this factor may be used for evaluation of gas permeability of rock mass during gas monitoring of "critical" boreholes. It should be considered that above described method for tritium concentration detection in soil and atmospheric air does not consider tritium contained in hydrogen, methane and other hydrocarbons.

3. DISCUSSION OF THE RESULTS

One of the main consequences of rock gasification is formation of cavities in UNE area and also expansion of uncollapsed cavities. This threat is confirmed by catastrophic collapse of rock and explosion in "Glubokaya" borehole. It should be understood that collapse of cavities may happen at any moment and may be accompanied by:

- Ground depression;
- Gas blow-up, explosion and burning;
- Escape of highly radioactive products from UNE.

Main threat from such event is posed toward entities involved in deposits exploration at "Balapan" site (coal deposit "Karazhara" and copper-molybdenum deposit "Ar-

man"). Nearest warfare boreholes are about 2 km from deposits boundaries. Collapse of cavities may be provoked by seismic vibrations from mining equipment and mining explosions. With that any gas blow-up from boreholes may be accompanied by escape of radioactive products which in case of unfavorable environmental conditions may lead contamination of adjoining territories and consequently – to accident irradiation of local population. Original ground depression is an evidence of formation and expansion of cavities. For example, at borehole 1355 in 2005 there was slightly bumpy surface registered and in 2008, 25 meter from the wellhead there have been a crater found with 20 meter diameter анв 12 meters depth (Figure 24).

a b c

Figure 24. "Balapan" site: a) general view of borehole 1355 from the south, 2005 ; b) depressed crater, 25 m from the wellhead, 2008; c) same in 2009.

Received data are not sufficient yet in order to find an answer to the question on dynamics of rock gasification located in central zone of UNE. For some boreholes there is still an open issue in nature and mechanism on gound depression. Ground depressions may happen as a result of expansion of UNE central area due to ground rock gasification. Or filling of a cavity and collapsed column with rock. Anyway since this process continues for more than 40 years, it is right to say that it will continue in the nearest future. To foresee processes in UNE cavities, and evaluate threat danger and prevent possible accidents, it is necessary to implement the whole range of measures.

Studies have shown that rock gasification processes at "Balapan" site continue and have rather stable nature. There have been local post explosion events of catastrophic nature registered. Due to the fact that there is active economic activity at "Balapan" site, it is necessary to perform study on evaluation of risk in connection with ground gasification processes with possible fires, collapse of UNE cavities and escape of conserved radioactive substances. In order to study process dynamics it is necessary to organized all year round gas monitoring at all critical boreholes with survey being performed not less than once a month. In order to increase awareness about performed work, below are recommendations listed on use of additional study methods.

Monitoring wells. Main issue of gas presence of the UNE area at "Balapan" site is existence of loose sediments of 90 m thickness. Loose sediments, like clay and silt, hinder gas escape to original ground. Most effective method of gas monitoring is monitoring wells drilled to the top of the base rock. With that use of drilled wells will be most effective if they are drilled in the areas of highest gas presence. To do that, it is necessary to detect anomalies of increased gas generation in epicenters of UNE. Study results have shown that gas release structures may be more than 100 meters from wellhead of warfare boreholes. Since increased

gas release and heat transfer happens in the same permeable structures, it would be reasonable to perform mercury metric, thermal and gas survey.

Mathematical modeling. It is interesting to create a mathematical model in order to quantify impact of underground nuclear explosions on the processes of heat transfer in ground. Proposed model created between sciences (geology, deformation and destruction mechanics, physics and chemistry), may be used for prognoses which has to do with evaluation of occurrence and special-time character of rock gasification in connection to specifics of structural-tectonic rock structure.

CONCLUSIONS

Rock gasification processes in the areas of indentified "critical" boreholes at "Balapan" sites continue and have stable character. Special attention should be paid to borehole 1010.

To the fact that processes of rock gasification in the areas of UNE at STS may cause catastrophic events, is necessary to continue studies on organization of effective monitoring and resolution of prognosis tasks with have to do with special-time evaluation of their development in dependence with peculiarities of structural-tectonic rock formation and composition.

Based on the fact that there is significant difference in composition and concentration of gases in identified "critical" boreholes, it will be right to state that specifics of geological structure and hydrogeological conditions play significant role in occurring gasification process. Issue of further study on this issue is complicated by absence of materials on geological structure of STS test area in Kazakhstan. Using this opportunity, authors of this article are asking Russian side to provide necessary documents and participate in study of "critical" boreholes condition at STS.

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"БАЛАПАН" АЛАҢЫНДАҒЫ АПАТТЫҚ СИПАТТАҒЫ ҮРДІСТЕРДІҢ АҒЫСЫНЫҢ МҮМКІНДІГІН БАҒАЛАУ

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Бұл мақалада тау жыныстарының жерасты газдануы үрдістерімен және отырықшы көріністермен байланысты жерасты ядролық жарылыстардың (ЖЯЖ) зардабын зерттеу нәтижелері берілген. "Балапан" сынақ алаңындағы "әскери" ұңғымалар жайлы әдеби және қордағы көздердегі мәліметтер жинағы, олар жайлы толық жинақ құруға және орналастыру сұлбасын құруға мүмкіндік берді. Алдын ала тексеру жұмыстарын өткізу барысында алынған зерттеулердің нәтижелері бойынша бірқатар ұңғымалардағы ойық көріністер анықталды, эпицентралды аймақтарға жасалған топографикалық мониторинг деректері бойынша олардың қарқыны жайлы алғашқы деректер алынды. Бұл мақалада сонымен қатар, ЖЯЖ орталық аймақтары орналасқан, тау жыныстарының қатпарларынан жердің бетіне шығатын газдардың шоғырлануын және құрамын зерттеу бойынша нәтижелер ұсынылды. ЖЯЖ эпицентралды аймақтарындағы ұзақ мерзімді геотермалды белсенділіктің қалдықты көріністеріне байланысты қауіптілік мәніне, апаттық сипаттағы жергілікті үрдістер сипатындағы баға берілді. Өткізіліп жатқан жұмыстардың ақпараттылығын арттыру үшін, зерттеудің қосымша әдістерін қолдану бойынша кеңестер берілді.

*Түйін сөздер***:** Семей сынақ полигоны, көміртегі оксиді, метан, күкіртті сутек, жерасты газдану, термиялық деструкция, газ бөліну.

ОЦЕНКА ВОЗМОЖНОСТЕЙ ПРОТЕКАНИЯ ПРОЦЕССОВ КАТАСТРОФИЧЕСКОГО ХАРАКТЕРА НА ПЛОЩАДКЕ "БАЛАПАН"

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В статье представлены результаты исследования последствий подземных ядерных взрывов (ПЯВ), связанных просадочными явлениями и процессами подземной газификации горных пород. Сбор сведений о "боевых" скважинах на участке Балапан по литературным и фондовым источникам позволил составить полный их каталог и построить схему размещения. По результатам рекогносцировочных работ установлены провальные явления на ряде скважин, по данным топографического мониторинга эпицентральных зон впервые получены сведения об их динамике. В статье также представлены результаты по изучению состава и концентрации газов поступающих из блоков горных пород, вмещающих центральные зоны ПЯВ, на дневную поверхность. Дана оценка значимости опасностей, связанных с эффектами остаточных проявлений долговременной геотермальной активности в эпицентральных зонах ПЯВ, как локальных процессов катострафического характера. Для повышения информативности проводимых работ разработаны рекомендации по использованию дополнительных методов исследований.

Ключевые слова: Семипалатинский испытательный полигон, оксид углерода, метан, сероводород, подземная газификация, термическая деструкция, газовыделение

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GEOCHEMICAL BACKGROUND OF STS. MICROELEMENTS IN SOILS OF SITES ADJACENT TO DEGELEN MOUNTAIN MASSIF

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Total content of more than 30 chemical elements in soils at the areas adjacent to Degelen mountain massif was determined. Abnormally high contents of several elements in relation to their clarke content have been revealed. A criterion was selected for evaluation of anomalies over average values of trace elements concentrations on their values and the heterogeneity of areal distribution. The greatest excess of content in relation to their clarke values was revealed for molybdenum, arsenic and cadmium. Areal distribution of concentrations showed its differentiated character, expressed in local patches of irregular shape at all sites.

Key words: clarke, element, concentration, anomaly, content, distribution, heavy metals, exposure.

Besides the radioactive contamination at the Semipalatinsk Nuclear Test Site there are areas with abnormally high content of non-radioactive toxicants (heavy metals and toxic elements). Heavy metals occupy a special place [1] by their effect on the biological objects due to their high toxicity and carcinogenic effect. Moreover, such effect may result in cell-level modifications in living organisms and gene mutation the consequences of which are similar to those of the ionizing radiation effect.

The data obtained in different years of research [2] were a basis for conducting the research works to assess the geochemical background of Semipalatinsk NTS and to identify areas with abnormally high contents of some chemical elements such as beryllium, manganese and molybdenum in water (tunnels with water inflow at Degelen site), soils (soil samples from tunnel portals) and bottom sediments.

Table 1 presents the data on the maximal contents of elements in Degelen waters compared to the mean value for the arid climate [3] and MPC for drinking water [4].

Table 1.

Comparison of the trace elements contents in natural waters of Degelen massif with the respective mean value for arid climate waters and MPC for potable water

Element	Concentration min-max	Average concentrations for arid climate	MPC for potable water
	mkg/l		
Be	60÷445	0.3	0.2
Li	25÷120	40	3
Mn	20÷87,000	135	100
Mo	20÷990	4	240
Zn	50÷8,000	85	5,000

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Data in Table 1 allow us to make a conclusion that concentrations of trace elements in natural waters of Degelen exceed the mean value for the arid climate and, what is more important, the maximum permissible concentrations for potable water.

Additionally, the content of a sum of rare-earth elements (REE) in the soil sample leachates of tunnel No 176 portal has been compared with their mean (Clarke) content in the soil where almost threefold excess over the Clarke value is recorded. The leachates are 1MHCl solution containing 100 grams of soil sample. The data are summarized in Table 2.

Table 2.

Content of rare-earth elements in the soil sample leachates from tunnel No 176

Element	Arithmetical mean value for concentrations in samples	Average contents in soil (clarke)	Element	Arithmetical mean value for concentrations in samples	Average contents in soil (clarke)
Dy	$6 \cdot 10^{-4}$	$5.2 \cdot 10^{-4}$	Nd	$0.3 \cdot 10^{-4}$	$0.3 \cdot 10^{-4}$
Er	$3.5 \cdot 10^{-4}$	$2.8 \cdot 10^{-4}$	Pr	$8 \cdot 10^{-4}$	$6.2 \cdot 10^{-4}$
Eu	$1 \cdot 10^{-4}$	$1.2 \cdot 10^{-4}$	Sm	$4.4 \cdot 10^{-4}$	$7.5 \cdot 10^{-4}$
Gd	$5 \cdot 10^{-4}$	$7.5 \cdot 10^{-4}$	Tb	$7.2 \cdot 10^{-3}$	$1.2 \cdot 10^{-4}$
Ho	$0.8 \cdot 10^{-4}$	$1.5 \cdot 10^{-4}$	Tm	$3 \cdot 10^{-3}$	$0.2 \cdot 10^{-4}$
La	$4.8 \cdot 10^{-3}$	$3 \cdot 10^{-3}$	Yb	$1 \cdot 10^{-4}$	$0.8 \cdot 10^{-4}$
Lu	$0.4 \cdot 10^{-4}$	$0.8 \cdot 10^{-4}$	$\sum$ REE	$1.8 \cdot 10^{-2}$	$6.5 \cdot 10^{-3}$

Data in Table 2 allow us to assume that the content of REE in soil is higher since the leaching method extracts just their small portion to the water phase.

1. MINERAL DEPOSITS ADJACENT TO DEGELEN MASSIF

According to geological data, the most typical elements for Degelen massif are tungsten, beryllium and molybdenum. Zirconium, antimony and tin have also been found. A group of greisens with tungsten mineralization forming an ore body is situated at the west slope of Degelen Mountains. The greatest amount of greisens hosting the rare-earth mineralization is concentrated at the South and North sites. The ore contains tungstate, hematite, pyrite, sphalerite, galena, scheelite, molybdenite, beryl and other minerals. The content of tungsten oxide is 0.5–4.5 %, molybdenum – 0.15 % and bismuth – 01 %. Degelen East, Degelen West and Zhambasbulak ore manifestations and Satbai zirconium manifestation are located at the deposit flanges.

The weakly studied tungsten-molybdenum ore manifestation, Mailykara is located among the Late Carboniferous - Early Permian granites 30 km north of Degelen Mountains. The content of tungsten oxide in soil samples is up to 3.96% and molybdenum – 0.73%.

The Karadzhal deposit is located in the northern part of Degelen massif. 9 ore skarns are known. The biggest body has the size of 500×100 m. The following skarns are distinguished as to the mineral composition: magnetite, garnet, garnet-vesuvian, pyroxene-garnet and garnet-fluorite. Iron content in magnetite skarnsranges from 6.36 to 37 %. The by-

metals are copper (0.13–3 %), lead (up to 1.2 %) and zinc (0.19–1.14 %). The beryl minerals are chryzoberyl and beryl-containing vesuvian; other beryl minerals exist in small amounts. The beryllium oxide content is 0.2 %, tungsten oxide – 0.4 %, molybdenum – 0.02 % and tin – 0.02 %.

2. EXPERIMENTAL PART

Topographically, the study area was determined based on the geological data on the explored deposits adjacent to Degelen massif. The sites to be studied were determined considering a possible effect of deposits on the geochemical background. On detailed investigation of the deposit pattern, three sites in the immediate vicinity of Degelen massif were chosen. The sites to be studied and locations of explored mineral deposits are shown in Figure 1.

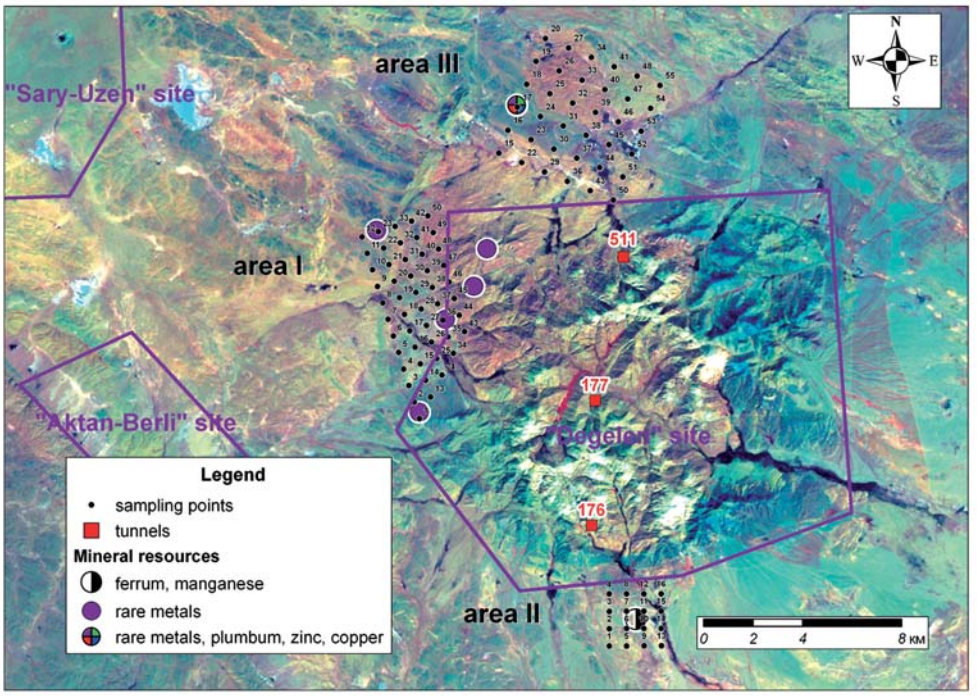

Figure 1. Sampling grid and location of explored mineral deposits

To recognize anomalies at these sites there was used litho-chemical mapping on dispersion flows over the scale 1:100,000 with 1000×1000 m grid at sampling. The scale was specified considering the task to recognize geochemical anomalies. Soil samples were collected from three sites adjacent to Degelen massif and on the streams from tunnels No 176, No 177, No 511 of Degelen site.

At site No 1 on the northwest side of the massif there were 50 sampling points. Site No 2 on the south side included 16 sampling points and site No 3 – 36 points.

Soil sampling along tunnel streams was conducted 0.5m of the waterway approximately every 100 m, starting from the stream exit.

Soil sampling was done by the puncture method using a soil sampling probe. The depth of soil sampling was 0–5 cm and the area – 100 cm^2.

Several analytical methods were used to obtain quantitative data on the concentration of elements in soil samples, such as X-ray fluorescence analysis (XFA), neutron-activation analysis (NAA) and mass-spectroscopy with the ion excitation source by inductively coupled plasma (ICP-MS).

Soil samples from sites No 1 and No 2 were analyzed by XFA method. Data on the content of the following elements were obtained: *As, Ba, Bi, Br, Ce, Co, Cs, Cr, Cu, Fe, Ga , La, Mn, Mo, Nb, Nd, Ni, Pb, Pr, Zn, Rb, Sb, Sn, Sr, Th, Ti, U, Y and Zr* with 1–700 μg/g detection limit and 5–20 % uncertainty.

For the preparation of soil sample for measurements it was dried at 40 °C. The resulting sample was ground to 50 mesh size and packed in a special container for measurements. Measurements lasted 15 min. The obtained spectrometry data were processed by the spectrometer software to obtain a quantitative result.

The ICP-MS method was used to determine the total content of elements at site No 3. The following elements were detected by this method: *Al, As, Ba, Be, Cd, Ce, Co, Cr, Cs, Cu, Dy, Er, Eu, Fe, Gd, Ho, La, Lu, Mo, Nb, Ni, Pb, Pr, Sb, Sc, Sm, Sn, Tb, Ti, Th, Tm, U, W, Y, Yb and Zn* with 0.05-5 μg/g detection limits and 5–15 % uncertainty. The detection was performed using "ICP MASS Spectrometer ELAN 9000" apparatus.

The method of autoclave digestion of soil samples was used. The method is based on soil mineralization in a sealed autoclave under the influence of elevated temperature and pressure. To achieve full mineralization, the autoclaving of soil samples was performed in two stages.

The 1st stage of autoclaving: 0.2 g sample was placed in a fluoroplastic insertion piece (reaction tank), and 5 cm^3 of nitric and hydrofluoric acid was successively added. For the check determination (blank control sample) a mixture of reactants without the tested sample addition was placed into the reaction tank. The autoclave was sealed, placed in a drying cabinet heated to 160 ^{0}C and left there for 5 h. The resulting solution with the precipitate was transferred to a centrifuge tube and centrifuged at a rate of 4500 revolutions per minute for 10 min. The centrifugate was quantitatively transferred into a fluoroplastic beaker (of 50 cm^3 volume), placed on an electric stove and evaporated to wet salts. 2 cm^3 of concentrated HNO_3 was added, and the solution was heated until dissolution of precipitate, cooled to room temperature and quantitatively transferred into a graduated flask (V=25–30 cm^3). The beaker was washed with 10–15 cm^3 bi-distilled water and brought to 20 cm^3 volume. The same procedure was performed with the blank sample. For the ICP-MC measurements the obtained solution was diluted tenfold.

The 2nd stage of autoclaving included the following: the residue (after the 1st stage) was transferred from the centrifuge tube into the reaction tank and 3 cm^3 of concentrated nitric acid and 1 cm^3 of hydrochloric acid were added. The autoclave was sealed and placed in a drying cabinet heated to 160 ^{0}C temperature and kept there for 5 h. The same procedure was performed with the blank sample. On completion of mineralization, the autoclave was removed from the drying cabinet and cooled to room temperature. For the ICP-MC measurements the obtained solution was diluted tenfold.

The reactants used were of especially pure grade. The solutions only after the 1st stage of digestion were used for measuring REE concentrations.

Soil samples from tunnels No 176, No 177 and No 511 of Degelen site were analyzed by neutron-activation method to detect the following elements: *Fe, Ce, Co, La, Rb* and *Lu* with 1–50 μg/g detection limits and 5–15 % uncertainty. The NAA method is based on the analysis of radioactive isotopes forming in the sample under the influence of irradiation by thermal neutrons. A soil sample was ground in a ball mill (to 50 mesh size). 0.3 g sample was weighed on an analytical balance, placed in special plastic capsules and then packed in metal foil and forwarded for irradiation. The irradiation was performed at NPI NNC RK in WWR-K reactor in channel No 11 in a passive zone; the thermal neutron flux was $6.7 \cdot 10^{12}$ $n \cdot cm^{-2}$ s^{-1} and the irradiation time - 5 hours. The samples were irradiated with IAEA-Soil-1 standard sample. The samples were exposed for 7 days to decay short-lived isotopes. Spectrometric measurements of samples lasted 30 min. The measurements were performed on HPGeOrtec GMX20 + MCA Ortec DSPEC jr. The concentration was estimated by comparison (i.e. the relative method of measurement) with the concentration of elements in the standard.

3. RESULTS AND DISCUSSION

Total contents of 26 trace elements in the studies soil samples from three sites of the lands adjacent to the Delegen massif are summarized in Table 3 below.

Table 3.

Table of results for soil samples from sites No 1, No 2 and No 3

Element	Site #1		Site #2		Site #3		Clarke as by Vinogradov [5]
	(min–max)/average	$\frac{C_{max}}{C_{min}}$	(min–max)/average	$\frac{C_{max}}{C_{min}}$	(min–max)/average	$\frac{C_{max}}{C_{min}}$	
Ca, %	(0.3÷3.0)/1.0	10	(0.5÷23.0)/3.0	46			3.3
Fe, %	(2÷7)/4	4	(1÷6)/4	6	(0.60÷3.80)/2	6	4.6
K, %	(1÷2)/2	2.5	(1.0÷2.0)/1.5	2			2.5
Mn, %	(0.04÷0.20)/0.10	4	(0.10÷0.14)/0.10	1.5			0.1
Ti, %	(0.1÷1.0)/0.4	0.1	(0.1÷0.8)/0.4	8	(0.2÷0.7)/0.4	3.7	0.45
Al, ppm					(220÷29,000)/18,000	135	80,000
As, ppm					(2.2÷24)/6	11	1.7
Ba, ppm	(200÷1,500)/680	7.5	(220÷835)/620	4	(6÷300)/120	55	650
Be, ppm					(1.0÷20)/4	18	3.8
Cd, ppm					(0.3÷7.5)/0.7	30	0.13
Ce, ppm	(40÷130)/85	3.5	(16÷100)/60	6.5	(0.4÷20)/9	50	70
Co, ppm	(6÷33)/20	5.5	(4÷40)/20	9.5	(3÷14)/8	4.5	18

Element	Site #1		Site #2		Site #3		Clarke as by Vinogradov [5]
	(min–max)/average	$\frac{C_{max}}{C_{min}}$	(min–max)/average	$\frac{C_{max}}{C_{min}}$	(min–max)/average	$\frac{C_{max}}{C_{min}}$	
Cr, ppm	(85÷140)/120	2	(40÷130)/100	3.5	(10÷60)/30	5.7	83
Cu, ppm	(20÷0)/30	3.5	(20÷50)/30	3	(8÷60)/20	7.9	47
La, ppm	(20÷90)/50	5	(20÷40)/30	2	<0.2		49
Lu, ppm	(0.3÷1.0)/1	5	(0.1÷0.8)/0.4	7	<0.1		0.8
Mo, ppm	(4÷50)/10	11.5	(4÷10)/6	2.5	(2÷20)/7	8	1
Nb, ppm	(8÷120)/20	15	(9÷24)/16	3			20
Ni, ppm	(20÷65)/35	3	(25÷50)/34	2	(6÷34)/17	6	58
Pb, ppm	(20÷50)/30	2.5	(20÷30)/25	1.5	(12÷34)/20	3	16
Rb, ppm	(70÷170)/100	2.6	(20÷100)/70	5			150
Sr, ppm	(50÷700)/300	13	(220÷820)/340	4			340
W, ppm					(1.5÷22)/4	15	1.3
Y, ppm	(20÷50)/30	2.2	(5÷40)/30	7	<0.2		29
Zn, ppm	(50÷200)/80	4	(44÷90)/70	2	(50÷120)/90	2.3	83
Zr, ppm	(160÷530)/240	3.3	(30÷270)/210	8.5			170

Concentrations of rare-earth elements are below the detection level of the ICP-MC method and have not been presented for consideration. The detection limits for Sm, Dy, Yb, Gd, Pr, Er< 0.2 μg/g, and for Eu, Ho, Tb, Tm < 0.1 μg/g.

Such elements as germanium, selenium, bromine, tin, antimony and cesium have not been detected at neither sites surveyed. The concentrations of these elements are below the detection level of XFA method: the concentration of Se, Br < 10 μg/g, Sn < 700 μg/g, Sb < 500 μg/g and Cs < 100 μg/g. Other analytical methods with a lower detection limit should be used for the detection of these elements.

The following have been chosen as a criterion for assessment of the results: 1 – excess of mean concentration of elements over their Clarke value; and 2 – existence of non-uniformity of distribution of elements at the sites surveyed.

For the surveyed sites, non-uniformity of the content of elements in the soil of the following sites has been detected: at site No.1 the maximum concentrations exceed the minimum concentrations for 2–15 times; at site No.2 the concentration excess is respectively 1.5–46 times; and at site No 3 the excess for beryllium, cerium and aluminum is found to be respectively 18, 50 and 135 times.

According to the data in Table 3, molybdenum shows a considerable excess (for 4–50 times) over the Clarke value in the soil. Concentrations of other elements are at the level of their Clarke contents in soil or are below the detection limit of the methods used.

At site No.1, north-west of Degelen massif, there has been detected an elevated content of molybdenum (4 ppm to 46 ppm) in the soil in relation to their Clarke (1.1 ppm) concentrations.

At site No.2, south of Degelen massif, elevated concentrations of molybdenum (4–10 ppm) have also been detected for 10 times higher than the Clarke values. No excess over the Clarke content has been noted for the remaining elements at Degelen massif site, however, maximum concentrations of strontium exceeding the Clarke and 2-2.5 times the mean value have been found. The spread of mean concentrations of elements at two sites No.1 and No.2 is 5–20 %.

At site No.3, north of Degelen massif located in the intermediate vicinity of Karadzhal fluorite deposit, elevated contents of molybdenum, cadmium, and arsenic have been detected. Thus the content of molybdenum exceeds the Clarke value for 7 times. The mean content of cadmium, 0.67 μg/g, exceeds the Clarke 0.13 μg/g for ~5 times. Accordingly, content of arsenic exceeds the Clarke 1.7 μg/g for 5 times. The other detected elements are at the level of their Clarke contents in the soil.

At the three surveyed sites, a significant excess over the Clarke content in the soil for such elements as arsenic, cadmium and molybdenum is respectively 3.5, 5 and 7 times.

Table 4 reports on total contents of 6 elements from the streams of tunnels No.176, No.177 and No.511 at Degelen site.

Table 4.

Table of results of analysis of soil samples from the streams of Degelen site

Element	Tunnel No.511		Tunnel No.176		Tunnel No.177		Clarke as by Vinogradov
	(min–max)/ average	C max / C min	(min–max)/ average	C max / C min	(min–max)/average	C max / C min	
Fe, %	(1.4÷2.1)/1.8	1.5	(1.5÷3.0)/2.2	2	(1.4÷2.4)/2.0	2	46000
Co, ppm	(4÷16)/10	4	(3÷15)/9	5	(3÷10)/6	3.5	18
Rb, ppm	(30÷60)/50	2	(70÷100)/80	1.5	(12÷80)/50	7	150
Ce, ppm	(70÷170)/120	2.5	(70÷120)/90	2.0	(93÷125)/110	1.5	70
La, ppm	(35÷70)/50	2	(40÷70)/60	2	(40÷60)/50	1.5	49
Lu, ppm	(0.6÷2.4)/1.5	4	(0.5÷1.5)/1	3	(0.6÷2.0)/1.3	3.5	0.80

No abnormal concentrations of detected elements have been found in soil from the streams of tunnels No.176, No.177 and No.511.

Based on the adopted criteria, the following elements have been distinguished for the sites No.1, No.2 and No.3: molybdenum, arsenic and cadmium. Areal distribution maps for the content of these elements in soil have been plotted (see Figures 2-4.)

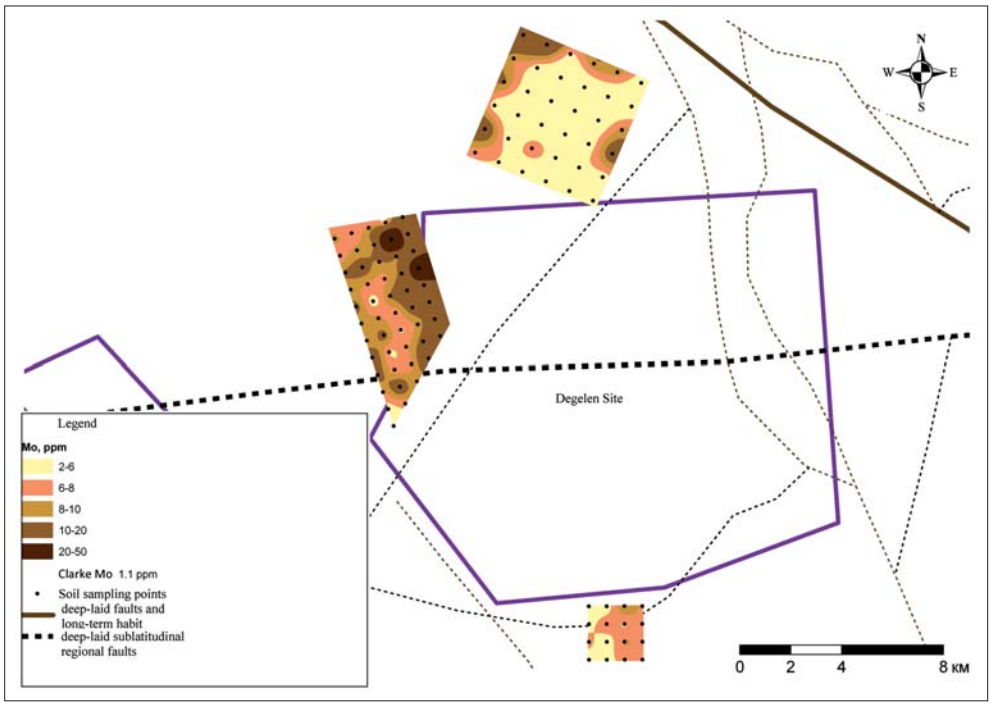

Figure 2. Schematic map of molybdenum content distribution in the soil of surveyed sites

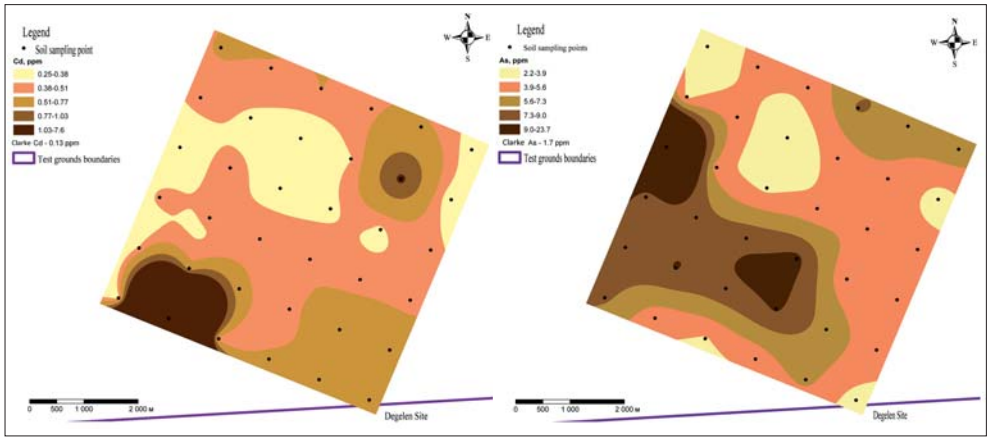

Figure 3. Schematic map of cadmium content distribution in the soil of site No.3

Figure 4. Schematic map of arsenic content distribution in the soil of site No.3

The schematic maps presented in Figures 2, 3 and 4 indicate an excess of Mo content at all three sites surveyed. The excess of Cd and As concentrations over their respective Clarke values has been recorded for site No.3, and the excess is respectively 4 to 32 times for Mo, 2 to 33 times for Cd and 2 to 10 times for As.

4. CONCLUSION

In this paper the preliminary data on the total content of 30 chemical elements have been summarized. A considerable excess over the Clarke content for such elements as arsenic, cadmium and molybdenum in soil has been detected at all three sites surveyed. The areal distribution of the concentration of elements has a differentiated character manifested in the local patches of irregular shape at all sites.

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ССП ГЕОХИМИЯЛЫҚ АЯСЫ. ДЕГЕЛЕҢ ТАУЛЫ МАССИВІНЕ ЖАҚЫН ЖАТҚАН ТЕЛІМДЕРДІҢ ТОПЫРАҒЫНДАҒЫ МИКРОЭЛЕМЕНТТЕР

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Дегелең таулы массивіне жақын жатқан телімдердің топырағы 30-дан аса жалпы химиялық элементтің құрамына зерттелді. Бірқатар элементтердің олардың кларктарына байланысты құрамы ауытқушылығы жоғарғы деңгейде белгіленді. Алаңдық таралудың әртектілігі және олардың кларкты мәні бойынша, элементтердің орта мәнінің шоғырлану мәнінің жоғарылауы бойынша ауытқуды бағалау критерийлері іріктеп алынды. Олардың кларктарына қатысты құрамының молибден, мышьяк және кадмийге қатысты жоғарылаудың орын алғаны анықталды. Шоғырлану бойынша элементтердің алаңдық таралуы барлық телімдерде анық байқалатын жергілікті дұрыс емес пішіндегі дақтары бар, диффренциалды сипатта орын алған.

Түйінді сөздер: кларк, элемент, шоғырлану, ауытқу, құрамы, таралу, ауыр металлдар, әсер ету.

ГЕОХИМИЧЕСКИЙ ФОН СИП. МИКРОЭЛЕМЕНТЫ В ПОЧВАХ УЧАСТКОВ, ПРИЛЕГАЮЩИХ К ГОРНОМУ МАССИВУ ДЕГЕЛЕН

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В почвах на участках, прилегающих к горному массиву Дегелен, исследовано валовое содержание более 30 химических элементов. Было установлено аномально высокое содержание ряда элементов по отношению к их кларкам. Выбраны критерии оценки аномалий по превышению средних значений концентраций элементов над их кларковыми значениями и по неоднородности площадного распределения. Установлено, что наибольшее превышение содержания по отношению к их кларкам имеет место для молибдена, мышьяка и кадмия. Площадное распределение элементов по концентрациям имеет дифференцированный характер, выраженный локальными пятнами неправильной формы на всех участках.

Ключевые слова: кларк, элемент, концентрация, аномалия, содержание, распределение, тяжелые металлы, воздействие.

CHAPTER: BROAD ISSUES OF ENSURING RADIATION SAFETY

УДК 504.75.05:577.4:615.849:612.461:546.799.4

CALCULATION OF DOSES FROM PLUTONIUM ISOTOPES BASED ON THE STUDY OF ITS CONCENTRATION IN URINE OF THE POPULATION IN THE STS IMPACT ZONE

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The article presents a model for calculation of internal doses from plutonium isotopes on the population, as measured in the urine. IAEA and ICRP biokinetic models were used for the calculations. Various scenarios of intake of plutonium isotopes into the body have been considered and assessments of internal doses based on these scenarios were performed. Influence of different ratios of plutonium isotopes on the dose rates has been considered. It was shown that when the content of $^{239+240}$Pu in urine is about 1 μBq/l, the dose of internal radiation from plutonium isotopes comprises $n*10^{-1} - n*10^{-2}$ mSv. The maximum contribution to internal dose is provided by $^{239+240}$Pu: 96–97% of the total dose.

Key words: plutonium radionuclides, internal exposure, absorption, excretion, dose of internal exposure, biokinetic model.

INTRODUCTION

Problems of radiation safety of the population living in the territories of the former Semipalatinsk test site (STS) attract much attention. In vicinity of nuclear testing site the population has been exposed to chronic irradiation due to inhalation of radionuclides and intake of radionuclides with food. At the present time the urgent point is to obtain reliable data about the dose loads on local people who lived there over a long period of nuclear testing and after in areas adjacent to the STS.

One of the most biologically hazardous radionuclides is plutonium. Toxicity of plutonium is associated primarily with the irradiation of tissue with α-particles, where it is accumulated after getting into the body. Period of biological elimination of half of the accumulated plutonium is comparable to the duration of human life. Direct determination of plutonium in the body, such as, for example, for gamma-emitting radionuclides ^{137}Cs or ^{241}Am, is impossible. According to some researchers, the urine formed in the kidney is reflecting the concentration of plutonium in the circulatory system, therefore the measurement of plutonium content in urine is the main source of data on total deposition of plutonium in the body [1].

The literature contains very little data on the calculation of internal exposure from plutonium isotopes, determined by their content in urine. In general, such calculations have been made for employees of the nuclear industry, who deal with plutonium at their work. Thus, the documents of the International Commission on Radiological Protection (ICRP) and the International Atomic Energy Agency (IAEA) suggest biokinetic models for dose assessment of so-called "standard" employee [2]: adult male with normal nasal breathing, rate of breathing is 1.2 m^3/h when doing light work, activity median aerodynamic diameter (AMAD) of respirable particles is 5 microns. Assessment of internal exposure is considered for the period of monitoring of workplaces - from several days to several months depending on the type of radionuclides getting into body.

No such model for population is described in literature. The IAEA publication [3] considers the assessment of internal radiation doses to the public during a long time after

getting the radionuclides into body - up to 20 000 days (approximately 55 years), used activity median aerodynamic diameter of respirable particles is 1 micron. These methods for assessing doses are recommended for use in the study of dose loads on the population after a long period of time that has elapsed since exposure. The recommendations contained in this publication can be used to assess dose loads on people exposed to surface nuclear tests at the test site during the years of 1949-1963.

1. THEORETICAL PART

1.1. Content of plutonium in a body

According to documents of the IAEA and the ICRP [2, 3, 4, 5, 6], the calculation of radionuclide intake into body and internal exposure is performed using biokinetic models - mathematical models describing the intake, absorption and retention of radionuclides in various organs or tissues and subsequent elimination from the body in different ways. Biokinetic models describing the behaviour of different radionuclides in the body have been published by ICRP and are based on conditional man and on observations of radionuclide behaviour in humans and animals.

Radionuclides get into the upper airway and then thoracic lung when inhaled. Deposition level of respirable particles is calculated for every division of respiratory tract. In inhalation model, the rate of dissolution and absorption of various radionuclides in blood must be taken into account. Compounds of plutonium in terms of absorption are moderately and slowly soluble. Intake and deposition of particles in the respiratory tract depend only on the particle size. Feature of plutonium isotopes is their ability to accumulate in bone tissue. When plutonium gets into the lungs through inhalation it partially settles on the surface of the lungs, partially passes into the bloodstream. Body retains about 5-25% of inhaled particles. Depending on the particle size and chemical form, plutonium getting through the respiratory is localized in the lungs or the lymph system or is absorbed by blood and transported mainly in the liver and bone. Approximately 60% get into the bone, 30% - in the liver and about 10% is eliminated in a natural way [1]. Elimination rate of incorporated plutonium from the body is very low. By some estimates, half-life of plutonium deposited in bone is about 50-100 years, and in the liver - approximately 20-40 years [1, 7].

When radionuclides are intaken through peroral way they get into stomach, small intestine, upper and lower divisions of large intestine. Average time of presence in the sections of the gastrointestinal tract (GIT) is 1, 4, 13 and 24 hours respectively. Absorption by blood comes from the small intestine and defined by coefficient of fractional absorption (f_1). There is evidence that only about 0.05% of soluble plutonium compounds and only 0.001% of insoluble compounds arrived with food get into adults' blood. The rest of it passes through the digestive tract and is eliminated from the body [1].

According to some researchers, ^{239}Pu is most intensively excreted in the first years after intake. When intake of ^{239}Pu into the body is one-fold only about 9% of the radionuclide is excreted with urine after 20 years. Upon that about half of that amount is excreted after about 3 years, and further the process of excrete slows down [8]. Urine that is formed in the kidney reflects the concentration of plutonium in the circulatory system; therefore the measurement of plutonium in urine is the main source of data on total deposition of plutonium in the body [1].

According to research conducted earlier in the Institute of Radiation Safety and Ecology, we can assume that the contents of $^{239+240}$Pu in urine was, on average, at the level of n*10^{-6} Bq. The Institute calculated the ratio of isotopes of plutonium-238, 239, 240 and 241 in different territories of STS in accordance with their half-lives for the initial time, today. According to our calculations, ratio of ^{238}Pu to the sum of^{239}Pu and ^{240}Pu in the northern territories is 0.02, ratio of ^{241}Pu to $^{239+240}$Pu is 0.75. At Degelen massif the ratio of ^{238}Pu to the sum of^{239}Pu and ^{240}Pu is 0.01, the ratio of ^{241}Pu to $^{239+240}$Pu is 0.9. In the territory of "Balapan" test site the ratio of ^{238}Pu to the sum of^{239}Pu and ^{240}Pu is 0.4, the ratio of ^{241}Pu to $^{239+240}$Pu is 0.7.

Based on these data, it is necessary to consider the content of plutonium isotopes in the urine of people living in different territories of STS. Thus, for example, one can assume that the content of ^{238}Pu in urine of the inhabitants of northern territories would be approximately n*10^{-8} Bq, and ^{241}Pu - n*10^{-7} Bq.

1.2. Assessment of internal exposure

To assess the peroral or inhalation intake of radionuclides by using analysis of excreta (urine or excrement) one uses data on the radionuclide content in the excreta and the time elapsed after the intake into body [2, 3, 4].

In accordance with a certain intake the expected effective dose for the period of time since intake is calculated as follows:

$$E = \frac{M}{m(t)} e(g)_j \text{ , where:}$$

$e(g)_j$ – dose coefficient for the radionuclide *j*;

M – measured absolute concentration of radionuclide in the daily excreta, Bq;

m(t) – fraction of the radionuclide, which remains in the body (for direct measurement) or removable from the body (for indirect measurement) at the time t after intake. The value of m(t) depends on the biokinetics of the radionuclide, i.e., its chemical form, ways of intake and time elapsed after intake, usually measured in days.

Since M - the value obtained after the analysis of urine and displays the content of a radionuclide in urine, then the focus should be the correct selection of the values of dose coefficient $e(g)_j$ and the value of m(t), which are contained in the publications of the ICRP, but depend on many factors. These values are determined by biokinetic models of radionuclides behavior in the body and eliminate from it.

Thus, in essence, correct choice of biokinetic model parameters, and then based on this selection of values $e(g)_j$, and m (t) will be the base for all of the calculations.

1.3. Rationale and choice of biokinetic models for assessment of internal exposure

To select biokinetic model one can use the following procedure:

1) Choice of dose coefficient:

a) Identify the way and intake time of the radionuclide (peroral, inhalation).

At peroral intake the absorption coefficient through intestine f_1 is used.

At inhalation intake the value of particle size, i.e. activity median aerodynamic diameter is used.

b) In both cases it is necessary to take into account the chemical form (type) of the incoming compounds, which determines the behaviour of the radionuclide at intake and its subsequent biokinetics in the body. There are three types of compounds: F - compounds with fast half-period, M - compounds with a moderate half-period and S - compounds with long half-period (Table 1).

Table1.

Types of chemical compounds

Half-period		Examples
Type F	100% absorption with half-period of about 10 minutes. Rapid absorption of almost all material deposited in the lungs. Half of the deposited material is taken to the gastrointestinal tract by transporting of particles and the other half is absorbed.	All compounds of cesium, iodine
Type M	10% absorption with half-period of 10 minutes and 90% - with a half-period of 140 days. There is rapid absorption of approximately 10% of materials deposited in the bronchial divisions of the lungs and bronchial tubes, and 5% of the material deposited in the lower respiratory tract. Approximately 70% of radionuclides deposited in the alveolar division of the lungs, is body fluids.	All compounds of radium and americium
Type S	0,1% absorption with half-period of 10 minutes, and 99,9% - with a half-period of 7,000 days. Small percentage of absorption is from the respiratory tract, bronchial and alveolar divisions of the lungs, approximately 10% of deposited radionuclides reach body fluids.	Insoluble compounds of uranium and plutonium

c) The dose coefficient e $(g)_j$ is chosen depending on AMAD, f_1 and type of absorption (Table 2).

Table2.

Dose coefficients for radionuclides of plutonium, depending on AMAD and the type of compound at inhalation

Radionuclide	Type/form	Inhalation		Peroral intake	
		$e(g)_{inh}$ (Sv/Bq)		f_1	$e(g)_j$ (Sv/Bq)
		AMAD=1 micron	AMAD=5micron		
Pu-238	M	$4.3*10^{-5}$	$3.0*10^{-5}$	$5*10^{-4}$	$2.3*10^{-7}$
	S	$1.5*10^{-5}$	$1.1*10^{-5}$	$1*10^{-5}$	$8.8*10^{-9}$
Pu-239	M	$4.7*10^{-5}$	$3.2*10^{-5}$	$5*10^{-4}$	$2.5*10^{-7}$
	S	$1.5*10^{-5}$	$8.3*10^{-6}$	$1*10^{-5}$	$9.0*10^{-9}$
Pu-240	M	$4.7*10^{-5}$	$3.2*10^{-5}$	$5*10^{-4}$	$2.5*10^{-7}$
	S	$1.5*10^{-5}$	$8.3*10^{-6}$	$1*10^{-5}$	$9.0*10^{-9}$
Pu-241	M	$8.5*10^{-7}$	$5.8*10^{-7}$	$5*10^{-4}$	$4.7*10^{-9}$
	S	$1.6*10^{-7}$	$8.4*10^{-8}$	$1*10^{-5}$	$1.1*10^{-10}$

2) Choice of m(t) depends on the intake form, time elapsed after intake and type of radionuclide absorption (Table 3).

Table3.

Excretion proportion of radionuclide $^{239+240}$Pu in the body with urine m (t) depending on the time elapsed since intake (Publication 37, IAEA)

Time elapsed since intake, days	m(t) - fraction of the radionuclide to be excreted from the body depending on time					
	Peroral intake		Inhalation			
	Transfer coefficients through intestine f_1		AMAD = 1 micron		AMAD = 5 micron	
	$1*10^{-5}$	$5*10^{-4}$	Type M	Type S	Type M	Type S
1	6.7E-07	3.4E-06	2.0E-04	2.0E-06	2.3E-04	2.3E-06
2	5.2E-07	2.6E-06	1.1E-04	1.2E-06	1.3E-04	1.4E-06
10	3.6E-08	1.8E-07	1.9E-05	3.2E-07	1.5E-05	2.3E-07
20	1.7E-08	8.7E-08	1.4E-05	2.9E-07	1.0E-05	1.8E-07
30	1.6E-08	7.8E-08	1.4E -05	2.8E-07	9.5E-06	1.7E-07
40	1.4E-08	7.1E-08	1.3E-05	2.8E-07	9.0E-06	1.7E-07
50	1.3E-08	6.6E-08	1.3E-05	2.8E-07	8.5E-06	1.7E-07
60	1.2E-08	6.1E-08	1.2E-05	2.8E-07	8.1E-06	1.6E-07
70	1.1E-08	5.7E-08	1.2E-05	2.8E-07	7.7E-06	1.6E-07
80	1.1E-08	5.3E-08	1.1E-05	2.8E-07	7.4E-06	1.6E-07
90	1.0E-08	5.0E-08	1.1E-05	2.8E-07	7.1E-06	1.6E-07
100	9.5E-09	4.7E-08	1.0E-05	2.9E-07	6.9E-06	1.6E-07
300	5.6E-09	2.8E-08	6.6E-06	3.1E-07	4.2E-06	1.6E-07
500	4.8E-09	2.4E -08	5.1E-06	3.3E-07	3.3E-06	1.7E-07
1,000	3.8E-09	1.9E-08	3.6E-06	3.4E-07	2.4E-06	1.8E-07
3,000	2.0E-09	1.0E-08	1.8E-06	2.6E-07	1.3E-06	1.4E-07
5,000	1.5E-09	7.6E-09	1.4E-06	2.1E-07	9.4E-07	1.1E-07
10,000	1.1E-09	5.5E-09	9.9E-07	1.6E-07	6.8E-07	8.2E-08
20,000	7.8E-10	3.9E-09	7.0E-07	1.1E-07	4.8E-07	5.8E-08

This table is presented as an example to illustrate the excretion proportion of radionuclide $^{239+240}$Pu from the body m(t). The publications by the ICRP 78 and 37 IAEA give the excretion coefficients of ^{238}Pu, $^{239+240}$Pu and ^{241}Pu from the body m(t) after peroral and inhalation intake, depending on the time elapsed after intake, the various transfer coefficients by intestines and various AMAD.

When no data are available a conservative estimate is used and maximum values of dose coefficients and f_1 are taken. To calculate internal dose exposure on population the IAEA recommends AMAD equal to 1 micron (for inhalation).

2. CALCULATION OF INTERNAL DOSES ON POPULATION IN THE STS INFLUENCE ZONE BY MEASUREMENTS OF $^{239+240}$PU CONTENT IN URINE

Let us consider in detail each stage of the proposed procedure applied to the calculations of internal doses from plutonium isotopes for people living in the zone of STS influence.

Scenarios of internal exposure may be different:

1. Internal radiation from radionuclides fallen into the body through the food chain.
2. Internal exposure from accidental ingestion of contaminated soil.
3. Inhalation of radionuclides from the underlying soil surface at the locations of population.
4. Inhalation of radionuclides from the most contaminated areas of STS to less contaminated areas (e.g. wind-driven transport).

Thus, for the population living in the zone of STS influence, we can distinguish two main models of radionuclides intake into body: inhalation and peroral.

Let us consider a few scenarios of human behaviour, living in the zone of STS influence, and, accordingly, several biokinetic models of plutonium isotopes intake into the organism.

2.1. Peroral intake

Let us assume that the main way of plutonium isotopes intake into the body - intake with food during the agricultural scenario, when a person eats meat and vegetables that contain plutonium isotopes.

2.1.1. 1st scenario (conservative). Parameters of the biokinetic model can be represented as follows:

1) Choice of dose coefficient:

a) Plutonium intake - peroral, therefore, must use transfer coefficient through intestine f_1. At a conservative estimate of slowly soluble compounds of plutonium (S) for the peroral intake there will be used transfer coefficient through intestine $f_1 = 1*10^{-5}$ (Table 2).

b) Type of compound - slowly soluble.

If intake of slowly soluble compounds of plutonium is peroral the dose coefficient $e(g)_j$ for $^{239+240}$Pu is equal to $9.0*10^{-9}$Sv/Bq (Table 2).

2) The area of residence - northern part of the STS. Let us assume that the intake of plutonium isotopes occurred 55 years ago (20,000 days). In this case, the excretion proportion, for example, of $^{239+240}$Pu with urine will be $7.8*10^{-11}$ (Table 3).

Thus, parameters of the biokinetic model for this scenario of plutonium isotopes intake into the organism can be represented in tabular form (Table 4).

Table4.

Parameters of the biokinetic model for plutonium isotopes

Isotope	Intake	Absorption type	Time t, elapsed after intake	Excretion proportion from the body m(t)	Content in urine (M), Bq	Dose coefficient $e(g)_j$, Sv/Bq at $f_1=1*10^{-5}$
^{238}Pu	With food	S	20 000 days	$5.1*10^{-11}$	$1*10^{-8}$	$8.8*10^{-9}$
$^{239+240}$Pu	With food	S	20 000 days	$7.8*10^{-11}$	$1*10^{-6}$	$9.0*10^{-9}$
^{241}Pu	With food	S	20 000 days	$5.6*10^{-12}$	$1*10^{-7}$	$1.1*10^{-10}$

Based on available data, as well, assuming that the content of $^{239+240}$Pu in urine is 1 μBq/l, we calculate internal exposure dose. For $^{239+240}$Pu it is:

$$E = \frac{M}{m(t)} e(g)_j = \frac{1 \cdot 10^{-6}}{7{,}8 \cdot 10^{-11}} \cdot 9{,}0 \cdot 10^{-9} = 1{,}2 \cdot 10^{-1}\, mSv$$

By the same way we calculate internal doses from isotopes of ^{238}Pu and ^{241}Pu. For ^{238}Pu dose is 1.7*10^{-3} mSv, for ^{241}Pu – 2.0*10^{-3} mSv. Total dose of all discussed plutonium isotopes is 1.2*10^{-1} mSv. Our calculations show that the isotopes of $^{239+240}$Pu give the maximum contribution to internal dose - 97% of the total dose.

In these calculations we used a conservative scenario with the worst conditions, i.e. compounds of plutonium with long half-period (S) and slow elimination from the body (low value of m (t)).

2nd scenario ("soft"). When considering the compounds with a moderate half-period (M) the following parameters of biokinetic model were used (Table 5):

Table 5.

Parameters of the biokinetic model for plutonium isotopes

Isotope	Intake	Absorption type	Time t, elapsed after intake	Excretion proportion from the body m(t)	Content in urine (M), Bq	Dose coefficient $e(g)_j$, Sv/Bq at f_1=1*10^{-4}
^{238}Pu	With food	M	20 000 days	2.5*10^{-9}	1*10^{-8}	2.3*10^{-7}
$^{239+240}$Pu	With food	M	20 000 days	3.9*10^{-9}	1*10^{-6}	2.5*10^{-7}
^{241}Pu	With food	M	20 000 days	2.8*10^{-10}	1*10^{-7}	4.7*10^{-9}

Calculation of internal doses with parameters of this model, the dose of ^{238}Pu will be 9.2*10^{-4} mSv, $^{239+240}$Pu – 6.4*10^{-2} mSv, ^{241}Pu – 1.7*10^{-3} mSv. The total dose of all the isotopes will be equal to 6.7*10^{-2} mSv. Contribution of isotopes of $^{239+240}$Pu in a dose will be 96% of the total dose.

Thus, under the worst conditions the dose loads will be almost twice as high. If we assume that about 50-55 years have passed since the surface and air tests at the test site, and during the time elapsed after these tests, people have been living in the zone of STS influence, running a farm and consuming the food grown in contaminated areas, the dose of internal radiation from plutonium isotopes when $^{239+240}$Pu content is in urine at 1 μBq/l will be n*10^{-1} – n*10^{-2} mSv.

These calculations are presented for the calculation of internal doses of the population living in the northern territories of STS. Having applied a similar scenario to the people living in other areas of STS, in particular, in Degelen and "Balapan" we got the following:

Table 6.

Total dose from plutonium radionuclides under various scenarios

Time elapsed after intake	Dose, mSv					
	1st scenario (conservative)			2nd scenario (″soft″)		
	Northern territories	Degelen	"Balapan"	Northern territories	Degelen	"Balapan"
10 years	5.4*10^{-2}	5.5*10^{-2}	7.6*10^{-2}	3.0*10^{-2}	3.1*10^{-2}	4.2*10^{-2}
30 years	8.3*10^{-2}	8.7*10^{-2}	12*10^{-2}	4.6*10^{-2}	4.9*10^{-2}	6.9*10^{-2}
55 years	12*10^{-2}	14*10^{-2}	20*10^{-2}	6.7*10^{-2}	8.1*10^{-2}	11*10^{-2}

Thus, in different territories of STS doses on population differ due to different ratios of isotopes of ^{238}Pu, $^{239+240}Pu$ and ^{241}Pu. When performing the calculations, it is necessary to take into account the specific territory in which the scenario of plutonium isotopes intake into the organism is considered.

2.2. Inhalation intake

Let us assume that the main way of plutonium isotopes intake into the body is trough inhalation that occurred 50-55 years ago.

On the recommendations of the ICRP, if the particle size is unknown, when calculating dose loads for the population AMAD equal to 1 micron is used. Consequently, parameters of used biokinetic model for the conservative scenario can be represented in tabular form (Table 7):

Table 7.

Parameters of biokinetic model for plutonium isotopes

Isotope	Intake	Absorption type	Time t, elapsed after intake	Excretion proportion from the body m(t)	Content in urine (M), Bq	Dose coefficient $e(g)_j$, Sv/Bq at AMAD=1 micron
^{238}Pu	Inhalation	S	20 000 days	$7.1*10^{-8}$	$1*10^{-8}$	$1.5*10^{-5}$
$^{239+240}Pu$	Inhalation	S	20 000 days	$1.1*10^{-7}$	$1*10^{-6}$	$1.5*10^{-5}$
^{241}Pu	Inhalation	S	20 000 days	$7.8*10^{-9}$	$1*10^{-7}$	$1.6*10^{-7}$

Based on available data, we can calculate internal dose from inhalation of $^{239+240}Pu$ into the body:

$$E = \frac{M}{m(t)} e(g)_j = \frac{1 \cdot 10^{-6}}{1{,}1 \cdot 10^{-7}} \cdot 1{,}5 \cdot 10^{-5} = 1{,}4 \cdot 10^{-1} mSv$$

Similarly, we calculate internal doses from isotopes of ^{238}Pu and ^{241}Pu. For ^{238}Pu the dose is $2.1*10^{-3}$ mSv, for ^{241}Pu – $2.1*10^{-3}$ mSv. The total dose of plutonium isotopes considered will be $1.4*10^{-1}$ mSv. Contribution of isotopes of $^{239+240}Pu$ in the internal dose is 97% of the total dose.

When using the scenario for the compounds of plutonium with a moderate half-period the parameters of biokinetic model can be represented in tabular form (Table 8):

Table 8.

Parameters ofbiokinetic model for plutonium isotopes

Isotope	Intake	Absorption type	Time t, elapsed after intake	Excretion proportion from the body m(t)	Content in urine (M), Bq	Dose coefficient $e(g)_j$, Sv/Bq at AMAD=1 micron
^{238}Pu	Inhalation	M	20 000 days	$4.6*10^{-7}$	$1*10^{-8}$	$4.3*10^{-5}$
$^{239+240}Pu$	Inhalation	M	20 000 days	$7.0*10^{-7}$	$1*10^{-6}$	$4.7*10^{-5}$
^{241}Pu	Inhalation	M	20 000 days	$5.0*10^{-8}$	$1*10^{-7}$	$8.5*10^{-7}$

When calculating internal doses using the parameters of this model, the dose of ^{238}Pu will be 9.3*10^{-4} mSv, $^{239+240}$Pu – 6.7*10^{-2} mSv, ^{241}Pu – 1.7*10^{-3} mSv. The total dose of all the isotopes will be equal to 7.0*10^{-2} mSv. Contribution of $^{239+240}$Pu in the dose - 96%.

Thus, dose of internal radiation from isotopes of plutonium at inhalation when $^{239+240}$Pu content is in urine at 1 μBq/l will be n*10^{-1} – n*10^{-2} mSv.

These calculations were given for the population living in the northern territories of STS, given that the contents of $^{239+240}$Pu in urine are at the level of 10-6 Bq at a ratio of ^{238}Pu to the sum of ^{239}Pu and ^{240}Pu equal to 0.02 and ratio of ^{241}Pu to $^{239+240}$Pu is 0.75. Upon that the period after intake of plutonium isotopes into the organism is 55 years. When calculating the doses on the population of "Degelen" and "Balapan" territories we obtain the following:

Table 9.

Total dose from plutonium radionuclides under various scenarios

Time elapsed after intake	Dose, mSv					
	1st scenario (conservative)			2nd scenario (″soft″)		
	Northern territories	Degelen	"Balapan"	Northern territories	Degelen	"Balapan"
10 years	6.6*10^{-2}	6.7*10^{-2}	9.5*10^{-2}	3.2*10^{-2}	3.3*10^{-2}	4.4*10^{-2}
30 years	9.5*10^{-2}	9.9*10^{-2}	14*10^{-2}	4.8*10^{-2}	5.1*10^{-2}	7.1*10^{-2}
55 years	14*10^{-2}	16*10^{-2}	24*10^{-2}	7.0*10^{-2}	8.4*10^{-2}	12*10^{-2}

Thus, as in the calculation of doses from peroral intake of isotopes, the doses received by the population will differ due to different ratios of isotopes of ^{238}Pu, $^{239+240}$Pu and ^{241}Pu. This factor when performing calculations should be considered.

CONCLUSION

Calculation of radionuclides intake and internal exposure dose is performed using biokinetic models describing the behaviour of different radionuclides in the body and given in the publications of the ICRP and IAEA. To assess the radionuclides intake using analysis of excreta (urine or excrement) one uses data on the radionuclide content in the excreta and the time elapsed since intake into body. The focus should be the correct choice of dose coefficient values $e(g)_j$ and the value of m(t), that is determined by biokinetic model of radionuclides behaviour in the body and elimination from it. Correct choice of biokinetic model and then based on this selection of values $e(g)_j$, and m(t) will be the base for all of the calculations. The main factor influencing the uncertainty in the calculation of internal doses is transfer coefficient through intestine f_1, and distribution and retention of plutonium in organs and tissues. Differences in intake and chemical form of absorbed radionuclides affect the degree of their absorption in the body. In addition, a long period of elimination of plutonium isotopes from the body makes a large contribution to the calculation error.

Inhalation and peroral intake are the main ways of radionuclides intake into the organism of people living in the zone of STS influence. Our calculations show that the internal dose of plutonium isotopes when $^{239+240}$Pu content is in urine at 1 μBq/l will be n*10^{-1}- n*10^{-2} mSv. Maximum contribution to internal dose is given by isotopes of $^{239+240}$Pu – 96-97% of the total dose. When performing the calculations of internal doses, the territory of the populations must be considered, as the ratio of isotopes ^{238}Pu, $^{239+240}$Pu and ^{241}Pu will vary for different territories of STS.

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ПЛУТОНИЙ ИЗОТОПТАРЫНАНЗЕРТТЕУ НӘТИЖЕЛЕРІ БОЙЫНША ССП ӘСЕР ЕТЕТІН АЙМАҒЫНДА ӨМІР СҮРІП ЖАТҚАН ТҰРҒЫНДАРДЫҢ НЕСЕПТЕРІНДЕГІ ОНЫҢ ДОЗАЛАРЫН ЕСЕПТЕУ

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Бұл мақалада плутоний изотоптарынан тұрғындардың ішкі сәулелену дозасын есептеу үлгісі, олардың несептегі құрамына байланысты келтірілген. Есептеу үшін АТЭХАГ және РҚХК биокинетикалық үлгілері пайдаланылды. Ағзаға плутоний изотопының түсуінің түрлі сценарийі қарастырылды және бұл сценарийлер негізінде ішкі сәулелену дозасына бағалау жасалды. Плутоний изотоптарының түрлі қатынастарының дозаның шамасына әсер етуі қарастырылды. Несептегі $^{239+240}$Pu құрамының 1 мкБк/л деңгейі барысында плутоний изотопынан түскен ішкі

сәулелену дозасының бағамы $n*10^{-1}$– $n*10^{-2}$ мЗв құрайтыны көрсетілді. Ішкі сәуленеу дозасының максималды үлесін, жиынтық дозасының 96–97 %-ын $^{239+240}$Pu изотопы қосады.

Түйін сөздер: плутоний радионуклиды, ішкі сәулелену, сіңіру, шығару, ішкі сәуелену дозасы, биокинетикалықүлгі.

РАСЧЕТ ДОЗ ОТ ИЗОТОПОВ ПЛУТОНИЯ ПО РЕЗУЛЬТАТАМ ИССЛЕДОВАНИЯ ЕГО СОДЕРЖАНИЯ В МОЧЕ ДЛЯ НАСЕЛЕНИЯ, ПРОЖИВАЮЩЕГО В ЗОНЕ ВЛИЯНИЯ СИП

Н.В.Брянцева, А.В.Топорова, А.А.Жадыранова, С.Н.Лукашенко

В статье представлена модель расчета доз внутреннего облучения населения от изотопов плутония в зависимости от их содержания в моче. Для расчетов использованы биокинетические модели МАГАТЭ и МКРЗ. Рассмотрены различные сценарии поступления изотопов плутония в организм и выполнена оценка дозы внутреннего облучения на основе этих сценариев. Рассмотрено влияние различных соотношений изотопов плутония на величину дозы. Показано, что при содержании $^{239+240}$Pu в моче на уровне 1 мкБк/л доза внутреннего облучения от изотопов плутония составит оценочно $n*10^{-1}$ –$n*10^{-2}$ мЗв. Максимальный вклад в дозу внутреннего облучения дают изотопы $^{239+240}$Pu – 96–97% от суммарной дозы.

Ключевые слова: радионуклиды плутония, внутреннее облучение, поглощение, выведение, доза внутреннего облучения, биокинетическая модель.

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DEVELOPMENT OF METHODOLOGY FOR STUDY OF DOSE FIELDS IN X-RAY ROOMS

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This work has been carried out to develop a methodology for studying the dose fields in the offices for roentgen diagnostics and is based on experimental studies of the formation of fields of doses during X-ray medical examinations. The data on the formation of dose field in the offices for roentgen diagnosing will allow not only assessing the doses uptaken by medical personnel during X-ray examination but also to assess the doses gained by personnel involved in research, but not the medical staff of the X-ray room, and the population helping the patients during examination.

Key words: X-ray, medical X-ray, dose, dose field formation.

INTRODUCTION

The radiation load on population due to development of nuclear power production and nuclear tests accounts only for a small part of radiation generated by human activity, as the population is exposed to much higher doses from other sources - when undergoing the diagnostic medical procedures and treatment using the sources of ionizing radiation. At the international conference "Radiation Safety in Medicine" there were given some estimates showing that the dose load on the population of Russia following the Chernobyl accident would make 150,000 person-Sv over the period of 50 years, while the doses of radiation from medical exposures would equal to the same values – 150 000 person-Sv only within 2001 [1]. In Kazakhstan the problem of the registration of the doses uptaken during X-ray examination both by the medical personnel and population is also very urgent. X-ray examination decreases considerably the time required to diagnose the disease, which makes this diagnosis method more and more popular and aggravates the problem.

Objective of the present work is to develop a methodology for studying the dose fields in the X-ray rooms. The data on the dose fields formation in the X-ray rooms will allow estimating the doses received by the healthcare personnel when performing examinations with the use of various roentgen apparatus. The results of the dose fields studies will also allow assessing the doses uptaken by specialists involved in research, excluding the staff of the X-ray room, and other members of the public who help the patients in examinations.

1. GENERAL PRINCIPLES

The major part of X-ray diagnostics equipment is represented by fixed installations operating pulse sources of x-ray radiation (energy range from 0.1 to 250 keV). For these devices, the minimal exposure dose claimed per one roentgenogram in the x-ray radiation receiver plane comprises 50 μSv. Even if one takes into consideration absorption in the air, the dose will decrease to 12.5 μSv, therefore, for the investigation of dose fields either the instruments with a low threshold of dose registration or integral-dose instruments are required.

In the course of the development of the methodology for studying the dose fields the principles of spatial distribution of roentgen radiation were considered. To study the dose fields, in the vertical and horizontal planes of the X-ray room on the main expected radiation pass wayswe placed dosimeters at the heights of 1, 1.5 and 1.8 mabove the floor level, which correspond to the gonads, chest and head. This choice of the dosimeters positions in the vertical plane was accepted from the given points of radiation control at working places [2]. In the horizontal plane the dosimeters were placed in the directions of the most probable distribution of X-ray radiation produced by the roentgen apparatus and evenly between these areas. In order to avoid the impact of the radiation background of the premises on the dose fields formed by the roentgen apparatus, prior to taking measurements, it is necessary to determine the dose rate of the radiation background when the apparatus is switched off [3] and subtract the contribution of that from the measured dose values.

Taking into account the task set, it was decided to use dosimeters accumulating the dose and not requiring the continuous control by operator - individual thermoluminescent (TL) dosimeters or direct-reading dosimeters. To study dose fields in X-ray diagnostics rooms, the use of TL dosimeters is the most appropriate, as direct-reading dosimeters do not allow registration of the whole range of the x-ray radiation since the energy range for this type of dosimeters starts from 60 keV, while the energy range of the TL dosimeters starts from 1keV. Besides, TL dosimeters are easy to maintain and energy-independent.

The investigations were conducted in-situ in normal functioning of the X-ray room. On the basis of the measurement results of effective dose formed by n-number of "shots' the spatial distribution of doses for one conditional "shot" is calculated ("shot" means a single examination or one scan of a patient with a x-ray machine). On the basis of measurements, average dose for one conditional "shot" has been estimated.

Taking into consideration the minimum declared exposure dose per one X-ray image in the plane of the receiver, dosimeters sensitivity (100 μSv) and possible distance from the x-ray source (up to 4 meters), to obtain the representative results it is necessary to produce about 70 "shots" (as these estimations were made without taking into account absorption by patient's body and the housing of the installation, the number of "shots" was determined with a double reserve).

2. EXPERIMENTAL ASSESSMENT OF THE METHOD APPLICABILITY

To evaluate the method applicability, we performed some experimental works to investigate the dose fields.

Experimental works were conducted on two types of x-ray-diagnostic devices – photofluorographic units and computed tomography units. The choice of this equipment was based on the following:

- Photofluorographic examinations are among the most widely used types of diagnostic examinations in Kazakhstan;
- Computed tomography methods are characterized by high dose loads upon patients.

On the basis of the results of reconnaissance investigation in the room equipped with computed photofluorographic unit it was established that representative results can be achieved with the number of "shots" not less than 100.

Experimental works were conducted in the rooms equipped with (Figure 1):

- Film photofluorographic unit 12F7K – 2 studies, 400 "shots" of photofluorographic unit;
- Computed photofluorographic unit "Fluarcom" 12FK1 – 2 studies, 270 "shots" of photofluorographic unit;
- Computed tomographic scanner CTHISPEED CT/E – 3 studies, 1196 "shots".

Figure 1. Experimental works

On the basis of the results obtained in the course of studies, the averaged dose per one "shot" of photofluorographic units and a tomographic scanner was calculated (Table 1, Table 2, Table 3, Figure 5, Figure 6, Figure 7). A standard deviation was calculated for the purposes of evaluation of the spread of the measured doses relative to the averaged dose (Table 1, Table 2, Table 3).

Table 1.

Averaged dose value per 1 "shot" in the premises equipped with a film photofluorographic unit 12F7K (standard deviation), μSv

Point	1 m above the floor	1.5 m above the floor	1.8 m above the floor
1	0.30(0)	0.28(0.04)	0.35(0.11)
2	0.26(0)	0.28(0.01)	0.28(0.08)
3	0.30(0.07)	0.39(0.03)	0.36(0.07)
4	0.24(0.07)	0.38(0.04)	0.34(0.04)
5	0.41(0.04)	0.48(0.11)	0.29(0.06)
6	0.46(0.08)	0.39(0.06)	0.32(0.06)
7	0.40(0.07)	0.19(0.03)	0.37(0.06)
8	0.34(0.06)	0.36(0.03)	0.47(0.08)
9	0.24(0.01)	0.22(0.07)	0.36(0.06)

Point	1 m above the floor	1.5 m above the floor	1.8 m above the floor
10	0.38(0.03)	0.47(0.05)	0.17(0.01)
11	0.20(0.01)	0.45(0.04)	0.27(0.04)
12	0.39(0.05)	0.43(0.04)	0.27(0.19)
13	0.24(0.06)	0.29(0.01)	-

Table 2.

Averaged dose value per 1 "shot" in the premises equipped with a computed photofluorographic unit "Fluarcom" 12FK1(standard deviation), μSv

Point	1 m above the floor	1.5 m above the floor	1.8 m above the floor
1	0.75(0.21)	1.60(0.14)	1.20(0.07)
2	0.71(0.05)	0.84(0.08)	0.64(0.11)
3	0.69(0.16)	0.71(0.15)	0.80(0.13)
4	0.95(0.01)	0.76(0.13)	0.88(0.06)
5	-	1.20(0.23)	-
6	0.69(0.11)	-	1.30(0.14)
7	0.41(0.06)	0.84(0.17)	1.10(0.16)
8	0.60(0.08)	1.20(0.07)	0.85(0.10)
9	1.00(0.12)	0.63(0.10)	0.85(0.04)
10	0.77(0.08)	0.65(0.17)	0.81(0.09)
11	0.99(0.05)	1.07(0.11)	0.85(0.01)
12	0.92(0.29)	1.28(0.74)	2.34(1.50)

Table3.

Averaged dose value per 1 "shot" in the premises equipped with a computed tomographic scanner CTHISPEED CT/E(standard deviation), μSv

Point	1 m above the floor	1.5 m above the floor	1.8 m above the floor
1	0.36(0.09)	0.35(0.11)	0.35(0.03)
2	0.28(0.10)	0.27(0.02)	0.27(0.14)
3	0.23(0.02)	0.27(0.10)	0.24(0.04)
4	1.70(0.25)	1.30(0)	0.85(0.03)
5	0.93(0.18)	1.10(0.29)	0.96(0.23)
6	0.80(0.20)	0.85(0.12)	0.75(0.04)
7	0.84(0.05)	1.00(0.25)	0.88(0.09)
8	1.00(0.25)	0.96(0.15)	0.90(0.17)

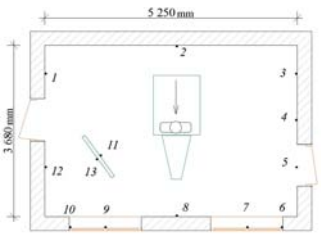


Figure 2. Arrangement of points of effective dose control in the room equipped with a film photofluorographic unit 12F7K

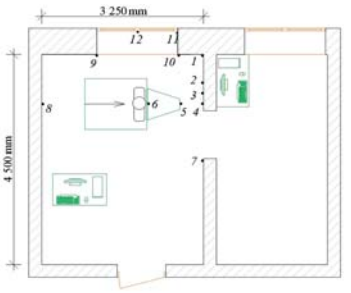


Figure 3. Arrangement of points of effective dose control in the room equipped with a computed photofluorographic unit "Fluarcom" 12FK1

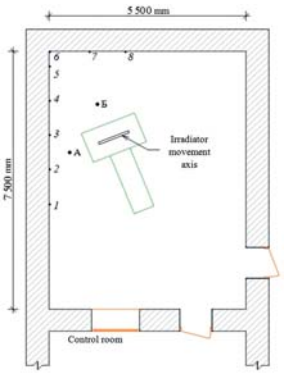
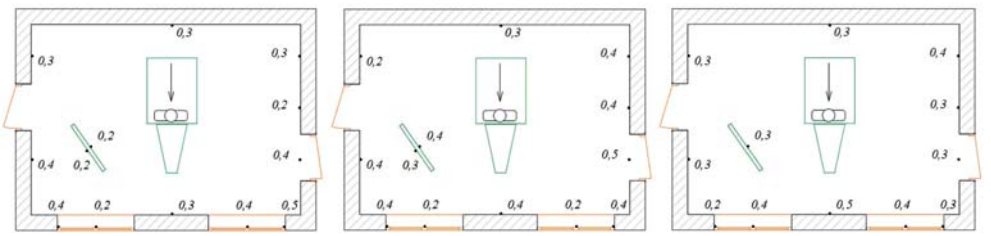


Figure4. Arrangement of points of effective dose control in the room equipped with a computed tomography scanner

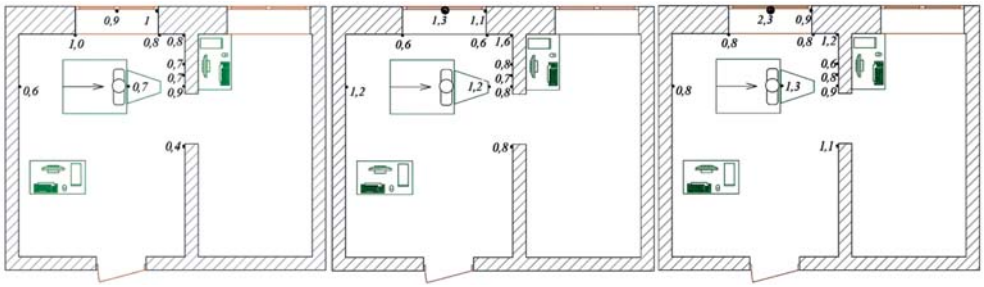
The analysis of the measurement data showed that to get a reliable picture of spatial distribution of dose fields with insignificant imprecision it is sufficient to perform two investigations with the same positions of dosimeters. In case of significant discrepancy in the results (exceeding the error of measuring instruments) it is necessary to perform the third investigation. This methodology however is based upon the assumption that in the course of the investigation the apparatus functions properly, operation parameters are within normal limits and the whole range of typical examinations is performed during the period of works. When a significant deviation of these parameters during experimental studies is observed, the obtained data on spatial distribution of doses may be either overestimated or underestimated.

When the dosimeters were arranged on the windows, the results showed significant data spread (Table 2, point 12). The data of the measurements are not suitable for the evaluation of the dose fields in the X-ray examination room, as these do not characterize the dose fields formed by the X-ray unit. It may be explained by technical peculiarities of the dosimeters used in the studies. TL dosimeters are sensitive to heating when constantly exposed to the sunlight, as it was in the case in the room with a digital photofluorographic unit and the data may be partly distorted (in the room equipped with a film photofluorographic unit the windows are located at the shady side).



1 m above the floor | 1.5 m above the floor | 1.8 m above the floor

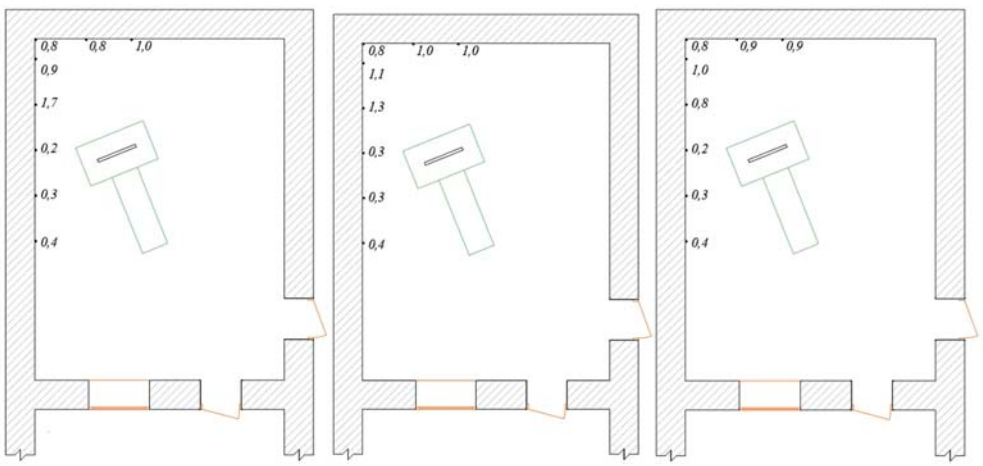
Figure 5. Dose distribution in the room equipped with a film photofluorographic unit 12F7K, μSv



1 m above the floor 1.5 m above the floor 1.8 m above the floor

– control points where unsatisfactory results were obtained

Figure 6. Dose distribution in the room equipped with a computed photofluorographic unit "Fluarcom" 12FK1, μSv



1 m above the floor 1.5 m above the floor 1.8 m above thefloor

Figure 7. Dose distribution in the room equipped with a computedtopographic scanner, μSv

Individual exposure doses were measured for all the employees of the hospitals where present studies were performed.

To check the applicability of the method for the evaluation of individual exposure doses of medical personnel, the measured individual doses of medical personnel were compared with the doses estimated on the basis of the investigated radiation fields (Table 4). The doses were estimated taking into account the background values in the premises (24-hour exposure a day), the values of the doses at working places at the moment of the "shot" of the X-ray unit (impact only in the period of the X-ray unit "shot") and the number of "shots" during the control period. The estimation included the following stages:

1. identification of the location of the medical staff during the examination of the patient;
2. calculation of the dose per one "shot" in the point of the medical staff location;

3. collection of the information on the number of examinations ("shots") per a quarter;
4. assessment of the cumulative dose on the personnel formed by the X-ray unit functioning;
5. estimation of the dose formed by the background radiation in the premises based on the results of the measured values of EDR with the switched off X- ray apparatus;
6. assessment of the dose per quarter (of the total dose formed by radiation background in the premises and the dose formed by the X-ray unit operation).

Comparison of the doses was performed only for the personnel of photofluorographic rooms since during CT scanning the personnel is out.

Table 4.

Results of individual radiation exposure control and calculated doses

Equipment	Name	Individual equivalent of effective dose $H_p(10)$ per a quarter, mSv	Estimated dose per a quarter, mSv
Filmphotofluorographicunit 12F7K	X-ray lab tech	0.52±0.15	0.85±0.25
Digitalphotofluorographicunit 12FK1	X-ray lab tech	0.34±0.10	0.45±0.14

The analyses show that the medical personnel of these medical institutions is exposed to the doses either below the estimated value, or at the estimated value level and the difference between the estimated dose values and the data obtained in individual dose rate control are in the area of uncertainty in measurement of these values. The dose values obtained by calculations in both cases exceed the measured ones as in performing the calculations it was assumed that the given personnel was present during all examinations that does not correspond to the real conditions.

Taking into consideration the results reproducibility, one may evaluate the supposed dosage on the personnel or the relatives of the patient assisting in the examination and present in the field of exposure to X-ray radiation during CT diagnostics in points A and B (Figure 4). Based on the assumption that during the examination of one patient about 30-34 "shots" are performed, it is possible to conclude that the dose of the assisting personnel present in the chosen points makes 0.04 mSv for point A and 0.23 mSv for point B.

CONCLUSIONS

This method of dose fields measurements allows simultaneous energy-independent measurements about the whole perimeter of the room in similar conditions without involvement of an operator.

Representative results require the number of "shots" to be not less than 100.

The present method does not take into account possible technical failures of the equipment and the difference in adjustments for various studies. When a significant deviation of the given parameters during experimental studies is observed, the obtained data may be either overestimated or underestimated. Since the major impact is caused by technical failures,

to decrease the error in dose fields determination it is necessary to assess the technical state of the apparatus and check the operating parameters prior to the start of the studies.

Distortions in the dosimeters' readings are possible if these are located on the windows at the sunny side or near heating facilities or devices which produce significant amount of heat in the course of their functioning (power supply sources, etc.).

The data on the radiation field studies are possible to use for the assessment of expected dosages uptaken by medical personnel or relatives of the patient assisting in examination and presenting in the area of exposure to X-ray radiation.

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РЕНТГЕН-ДИАГНОСТИКАЛЫҚ КАБИНЕТТЕРДЕГІ ДОЗАЛЫҚ АЛАҢДАРДЫ ЗЕРТТЕУ ӘДІСНАМАСЫН ӘЗІРЛЕУ

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Аталған жұмыс сәулелену диагностикасы кабинеттеріндегі доза алаңдарын зерттеу әдістемесін әзірлеу үшін жүргізілді және рентген-диагностикалық процедуралар жүргізу кезіндегі доза алаңдарының қалыптасуын тәжірибелі зерттеуге негізделген. Сәулелену диагностикасы кабинетіндегі доза алаңдарының қалыптасуы туралы деректер тек медициналық қызметкердің рентген-диагностикалық зерттеу жүргізу кезінде алған дозасын бағалауға ғана емес, сондай-ақ рентген кабинетінің қызметкер құрамына кірмейтін, зерттеу жүргізу кезінде қамтылған мамандардың және зерттеу жүргізу кезіндегі пациенттерге көмектесетін тұрғындардың дозаларына баға беруге мүмкіндік береді.

Түйін сөздер: рентген-диагностика, медициналық сәулелену, дозалар, дозалық алаңдардың қалыптасуы.

РАЗРАБОТКА МЕТОДОЛОГИИ ИЗУЧЕНИЯ ДОЗОВЫХ ПОЛЕЙ В РЕНТГЕНОДИАГНОСТИЧЕСКИХ КАБИНЕТАХ

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Данная работа проведена для разработки методологии изучения дозовых полей в кабинетах лучевой диагностики и основана на экспериментальных исследованиях формирования полей доз при проведении рентгенодиагностических процедур. Данные о формировании дозовых полей в кабинетах лучевой диагностики позволят не только оценить дозы, получаемые медицинским персоналам при проведении рентгенодиагностических исследований, но и дать оценку доз специалистов, задействованных при проведении исследований, не входящих в состав персонала рентген-кабинета, и населения, помогающего пациентам при проведении исследований.

Ключевые слова: рентгенодиагностика, медицинское облучение, дозы, формирование дозовых полей.

УДК 621.039.58:621.039.7:541.28:539.16

ABOUT EFFICIENCY OF PROCESSING TECHNOLOGY FOR LIQUID RADIOACTIVE WASTE FROM BN-350 REACTOR

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This article focuses on liquid radioactive waste (LRW) reprocessing technology for the fast breeder reactor unit (RU BN-350) in the city of Aktau (Kazakhstan). The work is based on theoretical calculations and experimental studies of efficiency performed for the main technological stages of liquid waste processing from BN-350 reactor. Experimental studies were done in the Laboratory of radiochemical research in the Institute of Radiation Safety and Ecology and in the Laboratory of engineering ecology of the Institute of Nuclear Physics under National Nuclear Centre RK. Samples of liquid radioactive waste taken from BN-350 units and model solutions with identical to real LRW were used in the studies. Efficiency of the proposed technology has been assessed based on the performed research.

Key words: liquid radioactive waste processing, fast breeder reactor, radionuclide composition of RU BN-350 LRW, evaluation of LRW cleaning efficiency.

INTRODUCTION

One of the key issues in the existence and future development of nuclear power production is the effective reprocessing of liquid radioactive waste and its safe utilization.

Liquid radioactive waste of reactor plants are the solutions with different salt load, contaminated with decay products, corrosive radionuclides and different substances used for water chemistry support and equipment decontamination.

Traditional LRW processing methods include evaporation to the maximum salt, cementation and bitumization. All these methods turn LRW into inert form, suitable for burying, but do not solve the problem of huge volumes of radioactive waste.

Most common technology nowadays uses the selective sorption method, which concentrates Cs tracers, bringing most of radioactivity to LRW, in the small amount of sorbent. Scientific findings [1,2] report the following technological stages of the selective sorption method of LRW rectification: pre-filtration and parent solution preparation, oxidation of the organic component of LRW, filtration and proper sorption. After oxidation and filtration the rectified solution is processed to absorb Cs on a Fe-cyanic sorbent, which is in most cases the nonorganic sorbent Termoxide-35.

The same selective sorption method was proposed for reprocessing of LRW, accumulated at the BN-350 reactor plant [3]. According to the raw data [4], salt content of the LRW is equal to 70–605 g/l, with volumetric activity of 10^7–10^8 Bq/l. Waste radioactivity is mainly produced by Cs, Co and Mn radionuclides.

In 2000–2001, the fore cited LRW reprocessing method was tested at Mangystau nuclear power plant in lab based benchmark tests on a pilot plant, consisting of ozonizing, filtration and sorption units. The pilot plant was composed of separate modules: reservoirs block, pump block, fittings, sorption columns, and ozonizer [5]. After pilot rectification of the RU BN-350 LRW, the ^{137}Cs content in LRW was reduced up to 10^{-9}–10^{-10} Cu/l (3,7–37 Bq/l),

and the ^{60}Co content - up to 10^{-8}–10^{-9} Cu/l (37–370 Bq/l) [5]. Data on the content of other radionuclides was insufficient. The available raw data was not enough for full assessment of the processed waste radiation hazard.

The raw data on composition of LRW in 2002 was the basis for pilot project of commercial plant for ion-selective refining of the RU BN-350 LRW. The projected reprocessing technology was to produce in the end a nonradioactive solution for its future evaporation, removal and storage as dry salt at the existing sanitary waste ground "Carrier 400" [4].

According to the reviewed historical data on the RU BN-350 LRW reprocessing technology and raw data records on the RU BN-350 LRW composition, we can expect the presence in the LRW of some radionuclides, the content of which shall be considered when categorizing and cleaning the waste.

1. EXPERIMENTAL PART

The following research was conducted to evaluate the efficiency of main technological processes of the RU BN-350 LRW reprocessing:

- Determination of the LRW radionuclides composition (theoretical and test study).
- Efficiency evaluation of ozonizing process for destruction of liquid radioactive waste complexing agent.
- Efficiency evaluation of LRW refining from Cs radionuclides during ion-exchange sorption.

1.1. Determination of the RU BN-350 LRW radionuclide composition

1.1.1. Theoretical evaluation

During operation, the reactor produces about 200 radionuclides (from Zn to Gd), consisting of decay products – radionuclides from the mid-range of the Mendeleev's Periodic Table, non-split Uranus and radionuclides with induced activity [6].

Except common radionuclides, produced during nuclear reactor operation ($^{137,134}Cs$, ^{60}Co, ^{54}Mn, ^{90}Sr and Pu isotopes), nuclear waste contains a crowd of short-living and long-living radionuclides , usually not accounted in the main technological methods of nuclear waste reprocessing. The article [7] describes approximate radionuclide composition of nuclear waste, just extracted from a pressurized water reactor after several years of irradiation: 1 ton of such waste contains 950–980 kg of $^{235+238}U$, 5–10 kg of Pu, 1,2–1,3 kg of ^{137}Cs, about 770 g of ^{99}Tc, about 500 g of ^{90}Sr, 500 g of ^{237}Np, 120–350 g of ^{241}Am, about 200 g of ^{129}I, about 60 g of ^{242}Cm and ^{244}Cm, 12–15 of g ^{151}Sm, etc.

The next research [8] proves the similarity of composition and activity of nuclear waste from thermal and fast breeder reactors. Still, there are certain peculiarities in the waste from fast breeder reactors, such as: volume of liquid waste is 2–2.5 times less, while volume of solid waste is 1.5–3 times higher, than from pressurized water reactors; specific solid active waste from fast breeder reactors are liquid metal primary sodium and Na-containing traps and absorbers.

Thus, basing on the foregoing approximate composition of water-cooled thermal reactor nuclear waste [7], we have evaluated possible current radionuclide composition of 1 ton of the RU BN-350 nuclear waste. As ^{137}Cs produces most part of the waste radio activity, we have calculated correlation of each long-living radionuclide to the ^{137}Cs activity. These correlations together with the quantitative data on the ^{137}Cs activity in liquid nuclear waste of the BN-350 reactor plant (10^7–10^8 Bq/kg, [3]) allowed us defining approximate radionuclide composition of the liquid nuclear waste of the RU BN-350. It being understood that:

- In passing of radionuclides from nuclear waste to liquid state, they dissolve evenly, thus preserving their correlation ratio;
- Date of fuel extraction from the reactor – 01.12.1999;
- Data on short-living radionuclides and radionuclides with induced activity was not accounted in the report;
- If several isotopes of one element were available, the activity was calculated by the radionuclide with the longer half-life.

See Table 1 for the evaluation results.

Table 1.

Evaluation of approximate radionuclide composition of nuclear waste and LRW from the RU BN-350 as of 01.01.2010

Radionuclide	Calculated specific activity in SNF, Bq/t	Ratio Radionuclide /^{137}Cs	Calculated specific activity in LRW, Bq/kg	Specific activity in decanter as for 01.09.2003, Bq/kg	IL (NRB-99), Bk/kg
^{238}U	$1.2\cdot10^{10}$	1:215 000	$2.2\cdot10^{3}$	not determined	3.1
^{239}Pu	$2.3\cdot10^{13}$	1:114	$4.2\cdot10^{6}$	not determined	$5.6\cdot10^{-1}$
^{137}Cs	$3.4\cdot10^{15}$	1:1	$4.8\cdot10^{8}$	10^7-10^8	11.0
^{99}Tc	$4.8\cdot10^{11}$	1:5400	$8.9\cdot10^{4}$	not determined	$2.2\cdot10^{2}$
^{90}Sr	$2.0\cdot10^{15}$	1:1.3	$3.8\cdot10^{8}$	not determined	5.0
^{237}Np	$1.3\cdot10^{10}$	1:200 000	$2.4\cdot10^{3}$	not determined	1.3
^{241}Am	$4.4\cdot10^{13}$	1:59	$8.1\cdot10^{6}$	not determined	$6.9\cdot10^{-1}$
^{129}I	$1.3\cdot10^{9}$	1:1 995 000	$2.4\cdot10^{2}$	not determined	1.3
^{244}Cm	$1.3\cdot10^{14}$	1:20	$2.3\cdot10^{7}$	not determined	1.2
^{151}Sm	$1.4\cdot10^{13}$	1:190	$2.5\cdot10^{6}$	not determined	$1.4\cdot10^{3}$

Our theoretical evaluations show that activity values of the main long-living radionuclides in the RU BN-350 LRW far exceed the specified levels (Radiation Safety Norms-99 doc). The following radionuclides shall be expected in the waste while establishing the technology for LRW reprocessing: ^{238}U, ^{239}Pu, ^{137}Cs, ^{99}Tc, ^{90}Sr, ^{237}Np, ^{241}Am, ^{129}I, ^{244}Cm, and ^{151}Sm. At that, most activity will be produced by ^{137}Cs and ^{90}Sr.

Test study was conducted to proof the calculated values of radionuclide composition of the RU BN-350 LRW.

1.1.2. Experimental evaluation

Subject of test were the samples of liquid radioactive waste, extracted from reservoirs B-02/2, B-02/3, B-02/5 of the RU BN-350, and standardized test solutions.

Test methodology

3 samples of LRW were selected for test determination of radionuclide composition of the RU BN-350 LRW. Lab test was done using certified methods [9,10,11,12], as well as modified ones. Content of gamma-emitting radionuclides was defined by gamma-spectrometer, containing of Canberra GX-2020 absorber diode with extended rating and relative registration efficiency of 20 %. We applied additional method of preliminary extraction of ^{137}Cs by codeposition on ammonium phosphomolybdate for more sensitive detection of ^{241}Am. For that, after consistent extraction of $^{239+240}$Pu radionuclides using extraction chromatography method, ^{134,137}Cs was two or three times codeposited from the filtrate together with ammonium phosphomolybdate. Then the filtrate was analyzed at gamma spectrometer to detect ^{241}Am, ^{60}Co, ^{54}Mn, ^{22}Na, and ^{125}Sb. After that the ^{90}Sr isotope was extracted from the filtrate by codeposition with calcium carbonate. TriCarb-2900 beta-spectrometer was used to define the content of ^{90}Sr.

Content of Pu isotopes was defined by alpha-spectrometry with preliminary radiochemical cleaning of the sample. The spectrometric sample was evaluated at Canberra 7401 alpha-spectrometer.

Lab test of LRW composition

The lab test of LRW samples from the RU BN-350 returned the quantitative data on radionuclide composition of the waste from the reservoirs B-02/2, B-02/3, B-02/5. The below table shows averaged data, obtained from analysis carried out by two laboratories of the National nuclear center of the Republic of Kazakhstan, as well as the research results, obtained while developing the design of liquid radioactive waste reprocessing complex for RU BN-350 (Table 2).

Table 2.

Radionuclide composition of LRW at RU BN-350

Radionuclide (unit)	Radionuclide content, Aspec					
	Tank B-02/2		Tank B-02/3		Tank B-02/5	
	*	**	*	**	*	**
^{241}Am, (kBq/l)	9 ± 2	no data	18 ± 1	no data	18 ± 2	no data
^{134}Cs, (kBq/l)	39 ± 6	270	20 ± 3	96	15 ± 2	260
^{137}Cs, (MBq/l)	123 ± 18	63	53 ± 8	185	113 ± 17	22
^{60}Co, (kBq/l)	15 ± 3	11	6 ± 1	37	25 ± 4	1,9
^{54}Mn, (kBq/l)	< 5	no data	< 2	1,5	< 8	no data
^{125}Sb, (kBq/l)	< 12	no data	< 13	no data	< 20	no data
^{22}Na, (kBq/l)	< 7	no data	< 3	no data	< 5	no data
$^{239+240}$Pu, (kBq/l)	< 1	no data	2.1 ± 0.5	no data	6.2 ± 1.3	nodata
^{90}Sr, (kBq/l)	65 ± 15	nodata	30 ± 10	nodata	80±20	nodata
Note: * - data of National Nuclear Center of the Republic of Kazakhstan ** - data reported in [13]						

The additional research has given proof to the estimates and historical data, testifying that Cs is the main contaminating radionuclide of the RU BN-350 LRW. Generally, it has also

proved the estimations on some other gamma-emitting radionuclides: ^{60}Co, ^{54}Mn, ^{125}Sb, ^{22}Na [14]. Still, during lab test we have also obtained data, giving evidence of significant content of such radionuclides, as ^{241}Am, ^{90}Sr and Pu isotopes.

While defining the content of ^{90}Sr by equilibrium content of ^{90}Y, we have obtained data evidencing the presence of ^{151}Sm in the RU BN-350 LRW. The tests with LRW samples showed deviation from the expected activity curve of ^{90}Y, thus evidencing the presence of some other radioactive isotope with similar chemical properties but longer half-life than Yttrium. This could be ^{151}Sm.

Radionuclides ^{99}Tc, ^{237}Np, ^{129}I, ^{244}Cm were not subject to detection during the lab test, but the theoretical evaluation expects these radionuclides in the LRW composition and calls for considering them when developing the LRW reprocessing technologies.

The researchers have tested the efficiency of main technological operations, proposed within the project of the RU BN-350 LRW cleaning from main contaminating radionuclides (Cs, ^{60}Co and ^{54}Mn isotopes): ozonizing process and ion-exchange sorption, using standardized test solutions with known radionuclide composition.

1.2. Efficiency evaluation of ozonizing process for destruction of liquid radioactive waste complexing agent

To perform this task, the researchers have conducted test evaluation of the LRW complexing agent oxidizing by ozonizing. It results in destruction of the LRW organic component and "releases" the organically bounded radionuclides, such as ^{60}Co and ^{54}Mn.

The literature sources normally propose using potassium permanganate in alkaline condition as oxidizer, as other oxidizers, like peroxide or nitric acid, are not effective in the presence of catalyst and other chemical oxidizers [15, 16, 17, 18]. High-efficiency oxidizer for versene is OH, produced for example during UV peroxide digestion. But it is also not effective for saline solutions [19], like the RU BN-350 LRW.

Ozone will be the most effective oxidizer in this case, due to a higher oxidizing ability, than most of other oxidizers. Besides, ozone will have strong effect on the organic substances in saline solutions [20].

This research was aimed to evaluate the efficiency of ozonizing for refining the RU BN-350 LRW from radionuclides. Most attention was given to the destruction of ethylenediaminetetraacetate – EDTA (versene), which the most popular nuclear decontaminant.

Versene was oxidized in an ozonizer with the capacity of 10.5 g of ozone per hour. Ozonolysis was carried out in a reactor pot of 2000 ml, with the solution temperature of 20ºC.

Test evaluation of LRW complexing agent oxidizing efficiency was done using standardized test solution with versene content of 130 g/l. Use of more concentrated solution is not practical, as then the solution over saturates, settling the sediment.

Test results

Theoretical stoichiometry of oxidizing reaction shows that oxidative degradation of 1g of versene requires 1,412 g of ozone:

$(CH_2COO\text{-})_2N\text{-}CH_2CH_2\text{-}N(CH_2COO\text{-})_2 + 9O_3 \rightarrow$ (intermediate oxidation products: carboxylic - oxalic and formic acids) $\rightarrow 2NO_3^- + 2OH^- + +2CO_2 + 5H_2O + O_2$

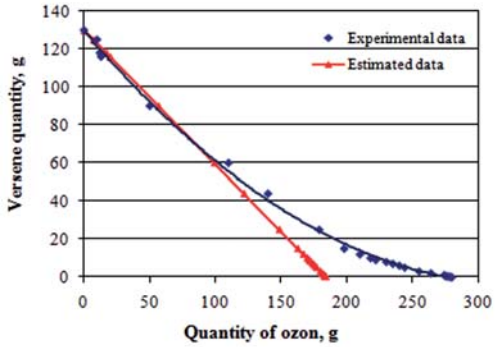


Figure 1. Change of versene component at ozonolysis

The obtained data shows that oxidative degradation of 1g of versene in solution with versene content of 130 to 60 g/l requires 1.4 g of ozone, while a lower versene content of 60 to 1 g/l will require from 1.4 to 2.2 g of ozone per 1 g of versene.

Taking into account the obtained test data (average oxidizing relationship between versene and ozone 1:2), productivity of an industrial ozonizer (2.5 kg of ozone per hour [1]), planned for use to reprocess the RU BN-350 LRW, typical concentration of versene in the LRW solution (30 g/l, [2]) and volume of the LRW decantate in one reservoir of about 1,500 m^3 (1 500 000 l), we will need 99 000 kg of ozone and 1650 days (about 4.5 years) of 24-h operation to reprocess 1,500 m^3 of the RU BN-350 LRW with one ozonizer of this type. As far as reprocessing of such LRW will take place in harmful conditions (1,700 hours per year limit), the ozonizing process using one ozonizer will take 23 years.

It shall also be considered, that we did not account other components of LRW solutions, which also have to be destroyed to clean the LRW from radionuclides, and this will also require significant ozone amounts. It is also important that when pH of the solution will go down during versene oxidation due to formation of intermediate oxidation products (oxalic and formic acids), ozone consumption in acidic media will grow.

Thus, research on oxidizing of LRW complexing agent, namely versene, show that ozone consumption depends on total amount of complexing agent in the solution. Conservatively, oxidizing of LRW complexing agent from one reservoir in harmful conditions will require 23 years of one ozonizer operation. According to the obtained results and considering the project data [13], where the estimated LRW reprocessing time is 5 years, and the volume of stored LRW is 5000 m^3, they will need at least 16 ozonizers to proceed.

1.3. Efficiency evaluation of LRW refining during ion-exchange sorption

After destruction of the LRW organic component and refining of waste from ^{60}Co and ^{54}Mn the project provides for sorption refining of LRW from main radioactive contaminant – Cs isotopes.

Project of the RU BN-350 LRW reprocessing [5] recommends to use Termoxide-35 as Fe-cyanic sorbent, that is a spherical granulated sorbent based on nickel ferrocyanide applied on nonorganic media – zirconium hydroxide.

Evaluation of LRW cleaning efficiency from Cs radionuclides during ion-exchange sorption was tested on standardized test solutions with different versene content and known activity of radioactive emitter ^{134}Cs. The standardized test solutions were filtered through fixed bed of sorbent (Termoxide-35) at a constant speed, at pH level of the medium equal to

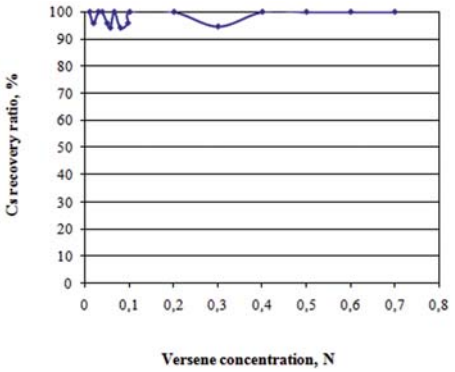


Figure 2. Dependence of ^{134}Cs removal using Termoxide-35 at pH=3÷4 from versene concentration in the solution

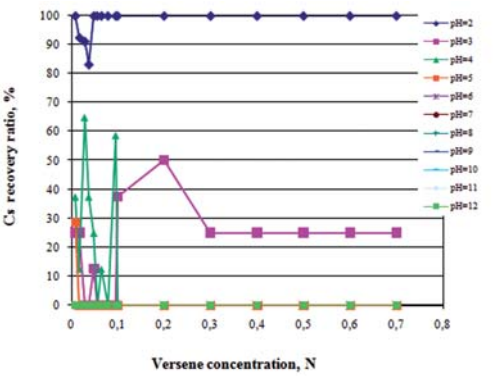


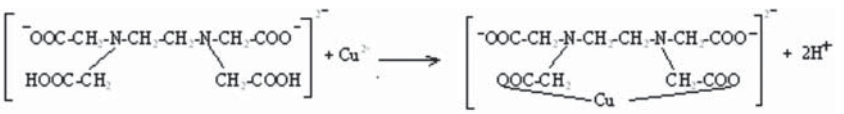
Figure 3. Dependence of ^{134}Cs removal using copper ferrocyanide at different pH levels from versene concentration in the solution

3÷4. See Figure 2 for the test results. More detailed analysis of Cs sorption by ferrocyanides required test extraction of Cs isotopes from solutions with different versene content using freshly-mixed suspension of copper ferrocyanide. See Figure 3 forthetestresults.

The obtained results show that in acidic media at pH=3÷4 cesium is well absorbed on Termoxide-35 from solutions with different concentration of versene (cleaning level 90–100%). The worst level of Cs removal (90%) at pH=3-4 was observed at versene content of 0.04 N (7 g/l) in the solution.

Cs deposition from the solution using copper ferrocyanide is most effective at versene content from 0.05 N (9 g/l) till 0.7 N (130 g/l) and at pH=2. At versene content from 0.02 till 0.05 N and pH=2, Cs extraction is below 100 %. In all other cases, the level of Cs removal from the solution in the presence of versene is below 65%; and starting from pH=6 the Cs is not sorbed from the solution. Under the given conditions the agent, absorbing the Cs from the solution (copper ferrocyanide), is dissolving.

^{134}Cs extraction ratio depends on pH level of the medium. At low pH levels (up to 3.5-4 after adding the suspension), EDTA in the versene solution dissociates, producing versene-ion and sodion. The versene-ion interacts with copper ion from copper ferrocyanide, thus dissolving the suspension, which should act as ^{134}Cs sorbent. According to reference data

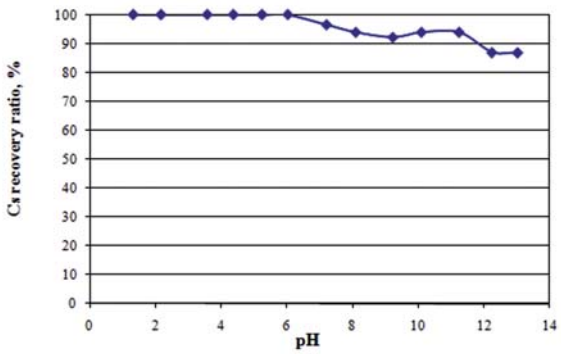


[1], compound of versene-ion with copper ion (EDTA) has stability constant 18.8, being more

stable than natriumcomplexonates, which have stability constant 1.66. Chemical reaction of the interaction is shown below:

Thus, different pH levels of the medium change chemical balance in sorbent – solution system, thus causing dissolution of the sorbent and reducing the Cs removal from LRW solutions.

As the RU BN-350 LRW reprocessing technology provides for the medium with pH level equal to 9–10 [5], we have studied this factor influence on Cs removal from standardized test solutions, cleaned by Termoxide-35. Taking into account the above results of tests on

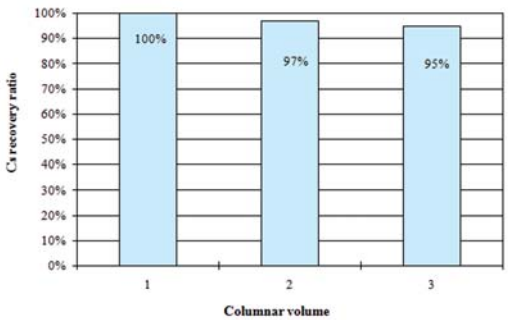


Termoxide-35, we have chosen a standardized test solution with versene content of 0.03 N (5.4 g/l), in which Cs cleaning performance reached 100 %. This solution was the basis for preparation of 13 solutions with different pH level (from 1 to 13). The solutions were filtered through ion-exchange columns with Termoxide-35. See Figure 4 for the graph with test results.

Figure 4. ^{134}Cs extraction ratio from standardized test solution with Termoxide-35 versus pH level of the solution at versene content of 5.4 g/l

The above graph shows that ^{134}Cs on Termoxide-35 absorption from versene-based standardized test solutions with pH levels of the medium from 2 to 6 is equal to 100 %. At pH levels from 7 to 13, the purification rate goes down to 87 %. Termoxide-35 reference data says that pH level above 11 destroys ferrocyanic sorbent. Due to this, preferable pH level is from 2 to 6.

Separately we have studied the case, when pH=12 and versene content = 0.03 N (5.4 g/l). As the test results show minimum level of ^{134}Cs removal by Termoxide-35 at this pH level, we carried out a test to define stages of standardized test solution cleaning by filtering portions of the test



solution through the sorbent. Total volume of the solution was equal to 120 ml, ^{134}Cs volume ac-

tivity in solution - 0.08 Bq/ml. 1 portion of the solution was equal to 1 columnar volume (40 ml). Solution throughput rate was 1 columnar volume per hour. After filtration through Termoxide-35, each columnar volume was tested on gamma-spectrometer. See Figure 5 for the test results.

Figure 5. Dependence of ^{134}Cs extraction ratio from standardized test solution using Termoxide-35 on the solution columnar volume at versene content of 5.4 g/l and pH=12

The obtained data shows that filtering of one columnar volume of standardized test solution with ^{134}Cs volume activity of 0.08 Bq/ml through the sorbent (Termoxide-35) extracts 100% of ^{134}Cs from the solution. At increasing columnar volume of solution per same sorbent, ^{134}Cs extraction ratio goes down to 95 %. At that, considering all the amount of ^{134}Cs, passing though the sorbent, the level of ^{134}Cs removal from the solution will reach around 89 %.

Summarizing the test results, we defined that for the versene content in solution below 20 g/l, Cs isotopes extraction ratio (by Termoxide-35) is not a constant and reaches about 90%. Considering the average activity ratio of waste, stored currently at the RU BN-350, equal to 10^7–10^8 MBq by ^{137}Cs, then 10% of ^{137}Cs activity, remaining in the solution, will emit about 10^6–10^7MBq. It means that sorbent-treated liquid waste will still be categorized as Liquid Radioactive Waste.

The conducted research shows that pH 2-6 returns the best results of the LRW cleaning from Cs isotopes. The project materials on the RU BN-350 LRW reprocessing technology provide for using the alkaline medium (9–10), what is not proper in comparison to test results. Such pH value does not allow for complete LRW treatment, as about 10 % of Cs activity remains in the solution.

CONCLUSIONS

1. The theoretical and test evaluations of radionuclide composition of the RU BN-350 LRW have proved the project data saying that Cs is the main contaminating radionuclide of the RU BN-350 LRW. In general, they have also proved the presence of other radionuclides in the LRW, such as: ^{60}Co, ^{54}Mn, ^{125}Sb, and ^{22}Na. The research returned quantitative data on such radionuclides, presented in the RU BN-350 LRW, as: ^{241}Am, Pu and ^{90}Sr isotopes, which were not considered during project development on LRW reprocessing. The tests did not define content of ^{99}Tc, ^{237}Np, ^{129}I, ^{244}Cm, and ^{151}Sm. Still, the theoretical evaluation expects these radionuclides in the RU BN-350 LRW, and this shall be considered in the LRW reprocessing technology.
2. The conservative evaluation of conducted test results shows that oxidizing of complexing agent of 5000 m^3 of the RU BN-350 LRW using ozonizers with 2.5 kg ozone per hour capacity will require 5-years' operation of 16 such ozonizers (taking into account the working hours' limitation in harmful conditions). Optimistic evaluation, which considers continuous running of ozonizers during 5 years, requires using at least 3 ozonizers.
3. Versene content in the waste and pH level of the medium are the important test parameters, effecting the removal of Cs isotopes from the RU BN-350 LRW by ion-selective sorption. At versene content in LRW equal to 20 g/l, about 10 % of Cs isotopes will remain in the solution, bringing the activity of 10^6–10^7MBq. The best performance cleaning of the LRW solutions from Cs isotopes at ion-selective

sorption stage was observed at pH from 2 to 6. At pH above 9–10 about 10 % of Cs activity remains in the solution.

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ЖН-350 РЕАКТОРЛЫҚ ҚОНДЫРҒЫСЫН СҰЙЫҚ РАДИОАКТИВТІ ҚАЛДЫҚТАРМЕН ҚАЙТА ӨҢДЕУ ТЕХНОЛОГИЯСЫНЫҢ ТИІМДІЛІГІ ЖАЙЛЫ МӘСЕЛЕГЕ

О.Ю.Коровина, С.Н.Лукашенко, В.В.Каширский

Бұл мақала Ақтау қ. (Қазақстан) орналасқан жылдам нейтрондардағы реакторлық қондырғының (ЖН-350 РҚ) қызметі нәтижесінде пайда болған сұйық радиоактивті қалдықтарды (СРҚ) қайта өңдеу технологиясына арналған. Зерттеулердің мақсаты: ұсынылатын технологияның тиімділігін бағалау.

Түйін сөздер: сұйық радиоактивті қалдықтарды қайта өңдеу, жылдам нейтрондардағы реактор, ЖН-350 РҚ радионуклидті құрамы, СРҚ тазалаудың тиімділігін бағалау.

К ВОПРОСУ ОБ ЭФФЕКТИВНОСТИ ТЕХНОЛОГИИ ПЕРЕРАБОТКИ ЖИДКИХ РАДИОАКТИВНЫХ ОТХОДОВ РЕАКТОРНОЙ УСТАНОВКИ БН-350

О.Ю.Коровина, С.Н.Лукашенко, В.В.Каширский

Данная статья посвящена технологии переработки жидких радиоактивных отходов (ЖРО), образовавшихся в результате деятельности реакторной установки на быстрых нейтронах (РУ БН-350), расположенной в г. Актау (Казахстан). Работа основана на теоретических расчетах и экспериментальных исследованиях эффективности основных технологических этапов переработки ЖРО РУ БН-350. Экспериментальные исследования проведены на базе лаборатории радиохимических исследований Института радиационной безопасности и экологии и лаборатории инженерной экологии Института ядерной физики Национального ядерного центра Республики Казахстан. Для исследований использовались образцы жидких радиоактивных отходов, отобранные из емкостей на РУ БН-350, а также модельные растворы, имитирующие химический состав ЖРО. В результате работы были сделаны выводы об эффективности технологии переработки ЖРО РУ БН-350.

Ключевые слова: переработка жидких радиоактивных отходов, реактор на быстрых нейтронах, радионуклидный состав ЖРО БН-350, оценка эффективности очистки ЖРО.

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UTILIZATION OF SOURCES OF RADIOACTIVE CONTAMINATION AT FORMER MILITARY ARSENAL IN TOKRAU SETTLEMENT

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In 2001, the military base "Arsenal" located in Aktogay area, Karaganda region, suffered from a fire. As a result the ammunition depots were damaged. The sources of high radioactive danger were detected in two storages of the arsenal. In 2006, the decision was made to carry out the elimination-conservation activities including utilization of generated radioactive wastes and their disposal. These activities were implemented by National Nuclear Center of the Republic of Kazakhstan and specialists of the Ministry of Defense of the Republic of Kazakhstan. As a result, the danger of radioactive contamination distribution was eliminated. All the radioactive wastes located at the site were utilized and placed for long-term storage at the complex "Baikal". Final survey of the site confirmed safety of the conducted activities and elimination of negative man-made influence on the existing ecosystem. During implementation of the activities the personnel involved didn't receive radiation doses exceeding the prescribed limits.

Key words: Tokrau settl., "Arsenal", ammunition made of depleted uranium, radioactive contamination, radioactive wastes, utilization.

INTRODUCTION

The military base "Arsenal" is located in Aktogay area, Karaganda region. Geographically, the base is situated at northern coast of Balkhash lake, less than 10 km from the coast.

In 2001, the military base "Arsenal" and its ammunition depots suffered from a fire. According to the data of Ministry of Defense of RK (MD RK) total ammunition stored in Tokrau settlement was several ton. There were projectiles and mines placed for storage by the soviet army during the war in Afghanistan and removed from Armenia after 1988 earthquake in Spitak. All the ammunition was meant for further utilization.

After the fire the sources of high radioactivity were detected in two facilities of the arsenal. Specialists of the National Nuclear Center of the Republic of Kazakhstan (NNC RK) were recruited to solve the problems of utilization of the radioactive contamination sources and to carry out nature-conservative measures in the territory of the facilities. In September 2006, specialists of NNC RK and MD RK conducted joint radiation survey of storage facilities # 9 and #18. The survey showed the following:

- Storage facilities contained parts of ammunition, packages of ammunition of various level of integrity and fragmentation;
- The storage facilities #9 and #18 posed hazard to surrounding territory, since some radioactive materials were in fine-dispersed state and could be easily spread by wind.

Radiation inside the storage facilities significantly (in hundred times) exceeded natural radiation background. Density of beta-particle flow exceeded permissible levels of surface contamination established in Radiation Safety Norms-99 (NRB-99) [1].

According to the survey results it was recommended to implement elimination-conservation activities including utilization of generated radioactive wastes and their disposal. These activities were implemented by joint efforts of NNC RK and MD RK specialists.

1. DESCRIPTION OF THE FACILITY

1.1. General information

The ammunition storages were reinforced concrete constructions partially banked with soil. Reinforced concrete arches were used as support walls and ceiling. The storages had concrete floor. Length of the storage was 70 m; inner width – 12 m; height – 7 m. Telpher paths and fire extinguishing system were mounted at the ceiling of each storage by using massive l-beams. During the fire the l-beams and water supply pipes were noticeably deformed and collapsed here and there (Figure 1).

Figure 1. Ceiling metal structures

Also the both storages had suffered from local collapse of the ceiling joists. During survey of the storage territories there were discovered the ammunition caps with radioactive materials and great number of the equipped ammunition.

1.2. Characteristics of radioactive contamination

1.2.1. Radiation distribution in the territory of the storages

The following radiation parameters were measured during a radiation survey: equivalent dose rate (EDR), density of β-particle flow, equivalent equilibrium volume activity (EEVA) of decay products of radon and thoron inside the storages. The measurement results were as follows:

- EDR was in range of 0.13-3,2 μSv/h;
- density of β-particles flow – from 40 to 4000 particles/(min*cm^2);
- EEVA of radon decay products – from 10 to 28 Bq/m^3;

- EEVA of thoron decay products didn't exceed detection limit of the used equipment (8 Bq/m^3).

Radiation survey of the adjacent territory was carried out as well. The results showed radiation contamination to cover the adjacent territory of more than 10 hectare.

The survey showed that radiation inside the storages (EDR and density of β-particle flow) exceeded radiation background for hundreds of times. Radiation was registered in the territory within 200 m from the storages.

These results allowed for classification of this site as being radio-hazard for general public and environment.

1.2.2. Analysis of radioactive sources

Assessment of radioactive contamination of the soil inside storage #9 was not carried out since preliminary survey showed that majority of the caps of 30-mm projectiles was not suffered from the fire (oxidation). Therefore, they couldn't be a source of radioactive contamination of the soil with combustion products (uranium oxides). Radioactive contamination in this storage was caused by the cores of the 30-mm projectiles being chaotically scattered in the storage. The ammunition was both intact and torn due to thermal impact.

Two samples of soil were taken to assess level of radioactive contamination inside storage #18. First sample was the remains of combustion products of 125-mm projectile located in transportation case; second sample was the soil containing combustion products of ammunition. Results of gamma-spectroscopy analysis are given in Table 1.

Table 1.

Results of gamma- spectroscopy analysis

Sample	EDR, μSv/h	Specific activity, Bq/kg	
		^{238}U	^{235}U
remains of combustion products of 125-mm projectiles located in transportation case	2	$1.7 \cdot 10^5$	$1.7 \cdot 10^3$
the soil containing combustion products of ammunition	0.5	$4.7 \cdot 10^4$	$3.2 \cdot 10^3$
Background area	0.14-0.15	–*	–*
Note: * – difficulties with identification of the radionuclides was due to their low concentration.			

The analysis results showed that remains of combustion products of 125-mm ammunition located in transportation case and the soil containing these products may pose radiation danger for personnel causing both external radiation and internal one due to inhalation. High radiation doses were called forth by uranium radionuclides. Other natural and man-caused radionuclides weren't discovered.

1.2.3. Analysis of physical-chemical parameters of radioactive contamination

1.2.3.1. Study of physical parameters of radioactive objects

Cores of the 30-mm and 125-mm projectiles were sent to a laboratory for analysis of their physical-chemical composition. Weight of 30-mm projectile's core was 235 g; weight of 125-mm core was 4.5 kg.

The samples were solid metal rods. Core of the 125-mm projectile was black with fragments of steel structures (tail unit, cap); length was 470 mm; diameter of middle part was 30 mm (see Figure 2). Core of the 30-mm projectile was grey with oxidation traces on a surface. Length of the projectile was 110 mm, diameter – 30 mm (see Figure 3 and Figure 4). Some samples had traces of thermal impact, such as: changes in metal structure (formation of black powder at the surface), oxidation traces on a surface (yellow coating).

Figure 2. Sample of the core of 125-mm projectile

Figure 3. Sample of the core of 30-mm projectile

Figure 4. Sample of the core of 30-mm projectile

Since material of the samples had heterogeneous structure then two objects were selected for study – metal rods and black powder. Powdery substance had fine-dispersed structure; particles were indistinguishable by the unaided eye; size of the particles was 50 μm at the most.

Radionuclide composition of the both materials was determined by using gamma-spectrometry analysis. The powder was studied by using the mass-spectrometry analysis to determine isotope composition of the material.

The laboratory studies of core of 30-mm projectile (weight of 235 g) and core of 125-mm projectile (weight of 4.5 kg) showed the presence of decay products of ^{238}U – ^{234m}Pa and ^{234}Th; ^{235}U was detected as well. Results of gamma-spectrometry analysis are given in Table 2.

Table 2.

Results of gamma-spectrometry analysis of 30- and 125-mm projectiles' cores

radionuclide	^{238}U	^{234}Th	^{234m}Pa	^{235}U
C, Bq/kg	$(1.5\pm0.8)\cdot10^{6}$	$(1.5\pm0.8)\cdot10^{6}$	$(1.5\pm0.8)\cdot10^{6}$	$(9.5\pm3.5)\cdot10^{4}$

Total concentration ^{235}U in the samples was 0.31% from the total quantity of uranium. Control measurements of the powder collected from 125-mm core were conducted in a laboratory of Institute of Nuclear Physics (Almaty) and showed the same radionuclide composition and similar specific activity (Table 3).

Table 3.

Results of gamma- spectrometry analysis of the powder from 125-mm projectile's core

radionuclide	^{238}U	^{234}Th	^{234}Pa	^{226}Ra	^{235}U	^{223}Ra	^{228}Ac
C, Bq/kg	$(1,5\pm0.2)\cdot10^6$	$(1,5\pm0.2)\cdot10^6$	$(1,5\pm0.2)\cdot10^6$	$< 7,0\cdot10^2$	$(1,0\pm0.1)\cdot10^5$	$< 1,0\cdot10^3$	$< 1,3\cdot10^3$

Gamma-spectrometry analysis showed that black powder and cores contain mixture of uranium isotopes (^{235}U, ^{238}U) and decay daughter products of ^{238}U (^{234}Pa и ^{234}Th). Specific activity was: ^{235}U ~10^5 Bq/kg, ^{238}U ~$1,5\cdot10^6$ Bq/kg. Weight concentration of ^{235}U in the samples was 0.30%. These data was confirmed by mass-spectrometry analysis, which showed presence of the uranium isotopes and their weight concentration to be ^{238}U – 99,72%, ^{235}U – 0.28%.

Analysis results showed that the samples contained "depleted uranium" used for projectile production, since natural concentration of ^{235}U is about 0.7%.

1.2.3.2. Study of chemical parameters of radioactive objects

Preliminary studies of the samples were carried out in a laboratory of Institute of Radiation Safety and Ecology. The samples were delivered from storages #9 and #18 of the former arsenal (Tokrau settl.).

Gamma-spectrometry analysis of the soil samples taken in an area with highest radiation parameters showed insignificant concentration of ^{238}U and ^{235}U. Concentration of radionuclide ^{137}Cs in the samples didn't exceed 2 Bq/kg, that is lower that background value for the territory of the Republic of Kazakhstan.

After gamma-spectrometry studies, chemical analysis of the samples was conducted in order to define possible composition of the materials.

Powder from the burnt core was black friable substance looking like activated charcoal. The substance was easy to grind and was mainly presented by fine powder-like fraction. The following features were studied to define chemical composition of this substance:

- Solubility in water;
- Solubility in acids (nitric, etching, sulphuric and hydrochloric acid).

To define solubility in water the substance under study was placed into a glass of distilled water and shuffled. After the lapse of time the powder settled at the bottom of glass. Visible changes were not detected. Then the sample was heated during one hour; visible changes were absent. To confirm the fact of no solubility in water and possible transfer of the radionuclides into water the sediment was filter by using "white tape" filter and the solution was studied by using gamma-spectroscopy analysis. The analysis confirmed visual observations. Water extracts didn't contain the radionuclides (^{235}U, ^{238}U, ^{234}Pa и ^{234}Th). Therefore, the conclusion was done on poor solubility of the substance in water.

To study behavior of the powder in the acids the set of the experiments was carried out by using general mineral acids. Results of the experiments are given in Table 4.

Table 4.

Results of the experiments with mineral acids

Substance	HCl, concentrated	HNO_3, concentrated	H_2SO_4, concentrated		HF, concentrated
			cold	hot	
Powder	No reaction	Reacted	Poor reaction with formation of greenish-grey sediment, emission of colourless gas, exothermic reaction	Reaction with formation of greenish-grey sediment, emission of colourless gas, exothermic reaction	reaction with further dilution of sediment and formation of greenish-blue solution

This analysis allowed for preliminary conclusions on possible chemical composition of the powder. This sample contained mixture of two uranium oxides (uranium dioxide – UO_2 and uranium oxide – U_3O_8), which were formed as a result of burning of the projectiles. In accordance with the literature data uranium burns into the air with formation of exactly these oxides [2, 3]. Chemical properties of these oxides are in full compliance with description of their properties in literature [4].

To define type of metal surface contamination (removable, non-removable), the damaged case of 30-mm projectile was preliminary measured at gamma-spectrometer, then washed in soap solution, dried and again measured. First measurements before deactivation showed presence of ^{234}Pa and ^{235}U in the sample; after deactivation these radionuclides were not detected.

Study of the physical-chemical properties of the samples delivered from storages #9 and #18 showed that main danger is posed by black powder from the burnt cores. Presence of depleted uranium in a form of dioxide and oxide presented with two problems: radiation and toxicity. Fine powdery fraction was additional issue as well.

Since the contamination of metal surfaces of the damaged projectiles is removable one then it is possible to carry our decontamination of these materials, and low solubility of the uranium compounds in water allowed cleaning scourage by using filtering technique. The generated radioactive wastes will be less bulky and more compact.

Thus, the samples under study were the cores of canon projectiles. They were exposed to thermal impact; as a result the structure of material changed. The additional study object was formed – black powder. As the gamma-spectroscopy studies showed, radionuclide composition of the cores and black powder was similar and presented the mixture of the uranium isotopes. Weight concentration of ^{235}U was about 0.3% from the total quantity of uranium, which categorized the samples' material as depleted uranium.

1.3. Volume of contaminated materials and their categorization

1.3.1. Ammunition

In accordance with the data presented by Central Ammunition Department of Armed Forces of the Republic of Kazakhstan, military unit 89533 (Tokrau settl.) had 125-mm armor-piercing sub-caliber projectiles and 30-mm armor-piercing projectiles.

30-mm projectile was brownish-grey uranium metal cap with yellow-green traces of uranium oxide. Some caps had white cones. During visual observation few intact caps were detected, so the conclusion was made that the biggest part of the projectiles was damaged and uranium metal caps were scattered and mixed with burnt packaging material (metal pipes, fastenings of the cases, etc.) and soil. Caps of the various 30-mm projectiles were observed in a radius of 50 m from the storage.

The survey showed that storages #9 and #18 had great number of 30-mm projectiles both torn and intact, and their cores.

EDR of uranium 30-mm penetrators was about 6 μSv/h, β flow density was $5 \cdot 10^3$ particles/(min*cm^2).

Total quantity of 30-mm armor-piercing shots in military unit 89533 was 101936 pieces. Their tactics-technical characteristics were as follows:

- weight of shot – 0.853 kg;
- weight of missile charge– 0.127 kg;
- weight of fully equipped projectile– 0.4 kg.

Thus, total weight of 30-mm armor-piercing shots was 12946 kg.

Using EDR categorization, 30-mm shots were related to low-level RW of group I in accordance with SRCPSRW-2003 [5] and SPORO-97 [6] (according to the reference [5] low-level RW are the substances of a dose rate exceeding a background from 0.001 mGr/h to 0.3 mGr/h – for this territory background EDR is about 0.13-0.15 μSv/h; according to the reference [6] group I of the wastes are the substances of EDR up to 0.3 mSv/h). Using categorization by β-flow density, this ammunition was related to low-level RW in accordance with SRCPSRW-2003 [5] (low-level RW are the substances of β-flow density from $5 \cdot 10^2$ to 10^4 particles/(min·cm^2)).

125-mm armor-piercing ammunition was stored at storage 18. Some transportation cases contained burnt ammunition cores; the cores were in the main lump of metal items as well.

Unlike 30-mm projectiles, the 125-mm projectiles were subjected to strong thermal impact. As a result the cores were partially or completely destroyed due to oxidation processes during the fire. Core's burning result was mixture of the uranium oxides (uranium dioxide – UO_2 and uranium oxide – U_3O_8) in a form of black powder.

EDR at the core's surface was 14 μSv/h, density of β-particle flow – $5 \cdot 10^3$ particles/(min·cm^2).

Total number of 125-mm armor-piercing ammunition in military unit 89533 was 6245 items. Their tactics-technical characteristics were as follows:

- Weight of shot – 20.43 kg;
- weight of missile charge– 8.86 kg;
- weight of fully equipped projectile– 7.05 kg.

Thus, total weight of 125-mm armor-piercing ammunition was 44027 kg.

Using EDR categorization, 125-mm shots are related to low-level RW of group I in accordance with SRCPSRW-2003 [5] and SPORO-97 [6]. Using categorization by the density of β-particle flow, this ammunition is related to low-level RW in accordance with SRCPSRW-2003 [5].

Summary information on the ammunition is given in Table 5.

Table 5.

Summary information on ammunition

Ammunition type	Q-ty	Weight, kg			Activity, Ci	
		Projectile	^{235}U*	^{238}U*	^{235}U	^{238}U
30-mm armor-piercing projectile	101936	0.127	40.13	12905,74	0.09	4,34
125-mm armor-piercing projectile	6245	7.05	136.48	43890.77	0.29	14,76
Total		57000	176.6	56796.51	0.38	19,1
note: * – in accordance with the laboratory studies ration of uranium isotopes in ammunition is about 99,69% of ^{238}U and 0.31% of ^{235}U.						

Total weight of 30-mm and 25-mm armor-piercing shots was ~57000 kg. RW volume including preliminary packages was about 85 m^3.

1.3.2. Contaminated soil

Storage # 9

Soil volume in western part of the storage was about 250 m^3, in eastern part – 370 m^3. Total volume of soil in the storage was about 720 m^3.

In accordance with gamma-spectroscopy studies concentration of ^{238}U in soil didn't exceed 10^4 Bq/kg, which is not RW but limited use material (LUM) according to SanR&S [6]. In accordance with preliminary assessments, 10% of total soil was considered as LUM, i.e. about ~70 m^3.

Storage #18

Soil volume in western part of the storage was about 80 m^3, in eastern part– 540 m^3. Total volume of soil in the storage was about 620 m^3.

Storage #18 contained both 30-mm and 125-mm projectiles, which combustion and destruction resulted in contamination of surface layer of the soil. This soil was not considered as RW taking into account measured values of EDR. But taking into account ^{238}U concentration it was radioactive wastes, since activity was 4 times bigger than minimal permissible specific activity established in NRB-99 [1] (10000 Bq/kg). In accordance with preliminary assessments, 10% of total soil was considered as RW, i.e. about ~120 m^3 (top layer of soil mixed with ammunition combustion products), and 25% of soil was considered as LUM, i.e. about ~150 m^3.

1.3.3. Metal items

In accordance with preliminary estimations, volume of metal items (transportation cases, ammunition fragments, etc.) was about 370 m^3 in storage # 18, and 180 m^3 in storage #9. Studies showed that after cleaning procedures, majority of the items can be categorized as unlimited use materials and can be handed over to a customer for utilization. Part of contaminated metal fragments shall not exceed 5% and is categorized as LUM. Their estimated volume shall not exceed 30 m^3.

2. ACTIVITIES ON UTILIZATION OF RW

The activities were carried out in accordance with the developed project. The project included environmental impact assessment report, which underwent an independent ecological examination and was approved by supervisory bodies: Ministry of Environment Protection and Committee of Energy of the Republic of Kazakhstan.

To implement the activities a field camp with the entire appropriate infrastructure was located at the site (see Figure 5 and Figure 6). The works were implemented in the course of 2 years. Firstly, collection and utilization of the radioactive ammunition were carried out for the adjacent territory. Then collection and clean-up of the territory of storages #18 and #9 were carried out correspondingly. After completion of the works final, radioecology survey was carried out for both storages and adjacent territory. All the activities were carried out together with the specialists of Ministry of Defense, which was responsible for providing for explosion safety while implementing the activities, clearing away obstructions with heavy machinery and providing the operation of the object's infrastructure. It is necessary to note that all the works on removal and utilization of RW were carried out under constant supervision of field engineers due to presence of great number of fully equipped ammunition at the site (Figure 7).

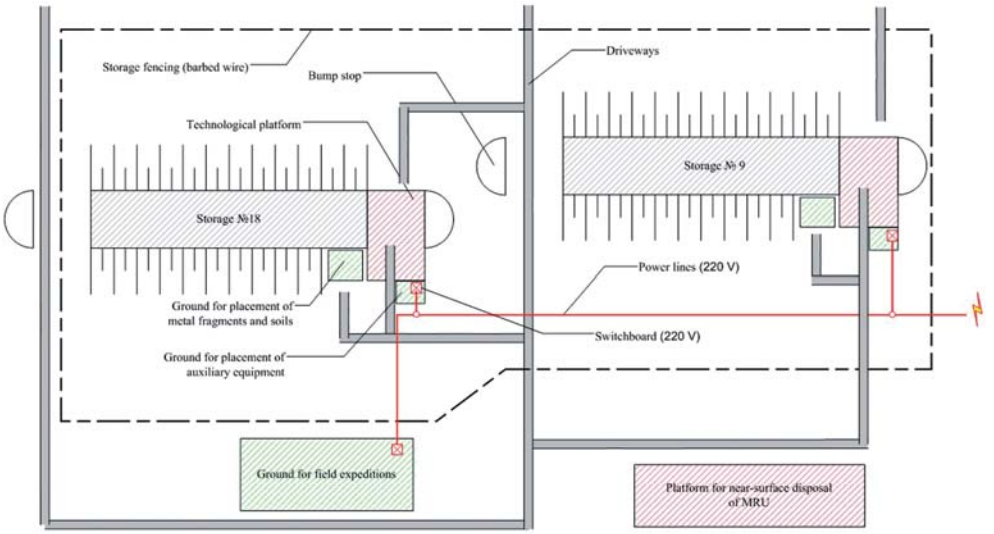

Figure 5. Layout of the technological sites and the field camp

Figure 6. Technological sites and the field camp

Figure 7. Work of army field engineer

2.1. RW removal technology

Technology for removal of radioactive wastes was developed on the basis of conducted studies. RW removal was carried out in four stages in accordance with the following scheme (see Figure 8).

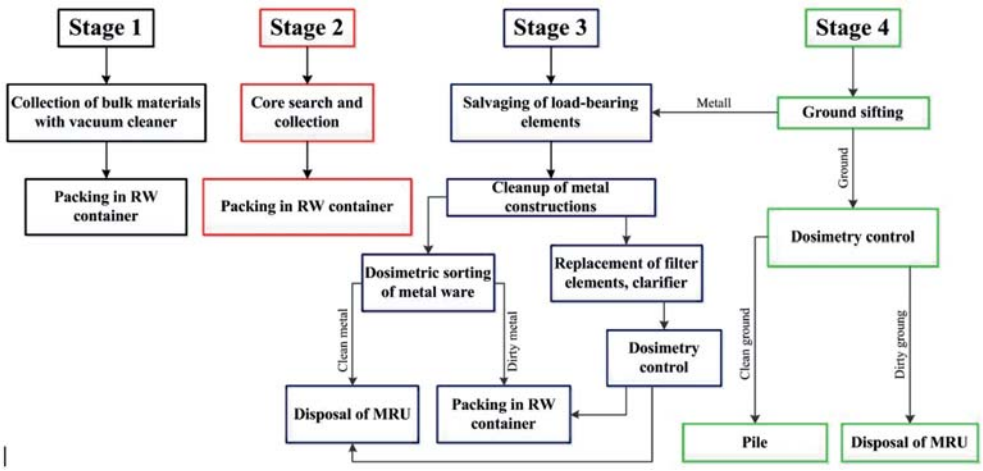

Figure 8. Scheme of step-by-step removal of RW

Stage 1 – Collection of loose RW

Collection of the loose radioactive materials was carried out by using the industrial vacuum cleaner simultaneously with the activities on cleaning of the storages. Filled filtering packages were placed into RW collection containers.

Stage 2 – Search and Collection of Radioactive Fragments

While clearing away a bulk in the storages, radioactive fragments (ammunition cores) were searched and collected by using metal handling devices (tongs). Transportation to the site of dosimetry control and loading into RW containers was realized in metal buckets. RW-related radioactive fragments were packed into initial packages and placed into RW collection containers. LUM-related fragments were placed at the LUM disposal site.

Stage 3 – Collection of Non-Radioactive Metal Fragments

This stage included several steps:

- Collection of transportation cases of 125-mm armor-piercing ammunition;
- Collection of other non-radioactive fragments;
- Cleaning of contaminated metal fragments.

While clearing away a bulk in the storages, transportation cases containing radioactive material (remains of burnt ammunition) were collected. Radioactive materials from the cases were packed into initial packages and placed into RW collection containers. Other non-radioactive fragments (fragments of storage buildings, broken non-radioactive projectiles, etc.) were transported to the dosimetry control site.

At the dosimetry control site the metal constructions were observes as for their contamination level. If they were clean then the constructions were utilized in usual fashion, otherwise they were sent for decontamination. If decontamination was impossible, RW-

related radioactive fragments were packed into initial packages and placed into RW collection containers. LUM-related fragments were placed at the LUM disposal site.

Metal fragments and constructions with radioactive contamination were cleaned at a specially designed and equipped site. Collected metal elements were loaded into special washing facility (Figure 9) and cleaned by using high pressure apparatus.

Figure 9. Decontamination of metal elements

A special metal frame with plastic curtains was placed over the washing facility to prevent sprinkling and leakage of water. Water after decontamination of the metal fragments was cleaned by using filtering substances (crushed stone, sand or other filtering material) and was sent back into washing tank after passing final fine filter. So closed cycle of water circulation was provided. Specific radioactivity of water was periodically measured to control radionuclide concentration in returning liquid. If the filtering and sedimentation tanks were clogged up, the cleaning was carried out and filtering elements were replaced. Sediments and filtering elements underwent dosimetry control. If the materials were categorized as RW, they were packed into preliminary packages and placed into RW collection containers, otherwise they were placed at the LUM collection site.

Stage 4 – Control of presence of radioactive materials in soil

After completion of the collection of the metal items the soil was sifted by using a vibrating screen. The soil was not moved from the storages. This procedure insured soil to be free from radioactive fragments (ammunition cores). Soil was sifted by using of the vibrating screen of ~0.8-1 cm opening. Thus the metal elements of size more than >0.8-1 cm were collected. Soil was manually supplied to a receiving bin of the vibrating screen. Collected metal elements were sent to dosimetry control site to control their radioactive contamination. The soil was also controlled as for its contamination. If contamination was not detected then metal fragments were handed over to MD RK representative for following utilization in general fashion (Figure 10), otherwise the fragments were categorized. RW-related soil was packed into preliminary packages and place into RW collection containers. LUM-related wastes were placed at the LUM collection site.

Figure 10. Clean scrap metal are prepared for transfer to MD RK representative

Figure 11. Transportation packages

2.2. Packaging and transportation of RW

In the course of the works RW were packed into preliminary packages and placed into transportation packages (Figure 11).

RW was transported en route Tokrau settl.– Kurchatov by motor transport in compliance with the requirements of the RK legislation. After delivery the transportation packages filled with RW were placed for long-term storage at the complex "Baikal" of Institute of Atomic Energy NNC RK (Kurchatov). During the course of the works more than 30 trips on RW transportation were implemented. Total RW volume placed for storage was 330 m^3.

2.3. Ensuring radiation safety while conducting the works

The works on utilization of the radioactive contamination sources in the territory of storages # 9 and 18 were considered as radiation-hazardous since they posed real radiation danger to personnel. Thus the measures were developed to ensure radiation safety of personnel including continuous radiation and dosimetry control.

During the works the personnel was exposed to combined radioactive irradiation – external and internal one due to inhalation. Main sources of external danger were gamma-emitting radionuclides (^{238}U, ^{235}U, ^{234}Th, ^{234}Pa), sources of the internal danger were fine-dispersed particles and aerosols containing α- and β-emitting radionuclides (^{238}U, ^{235}U, ^{234}Th, ^{234}Pa). While carrying out the works all the types of irradiation were taken into account; the protective measures were provided to minimize total dose from all the external/internal sources up to levels lower than maximum permissible dose or a dose limit for corresponding categories of the personnel. While conducting the activities under conditions of air contamination with radioactive substances the personnel was provided with additional disposable special clothes, filtering and isolating means for respiratory protection (Figure 12). Radiation control included observation over radiation situation in the territory of the working site and in the storages.

Figure 12. Work with using the additional personal protection means

Figure 13. Determination of specific activity and dose rate

The following list of the radiation control parameters was composed taking into account a type of the works carried out:

- Measurement of gamma-radiation dose rate, flow density of α-, β- and ionizing radiation particles in the storages;
- Measurement of the levels of radioactive contamination of the surfaces, equipment, transportation vehicles, personal protection means, skin and clothes of personnel;

Determination of specific activity and dose rate of solid radioactive wastes (Figure 13).

These parameters were used as a basis to assess probable radiation burden of the personnel working in a working area. Control of radioactive contamination of the working area, equipment, accessories, and transport vehicles was realized by direct measurements by using portable devices and smear collection technique.

Individual control of personnel irradiation included:

- Radiometry control of skin and personal protection means;
- Control of the doses of external beta-/gamma-radiation by using individual accumulation-type dosimeters. While working in the contaminated areas the personnel was provided with direct reading dosimeters;
- Control of internal intake of uranium isotopes by their content in urine.

Control of the doses of external beta-/gamma-radiation showed that personnel doses were within the range from 0.3 to 2.4 mSv/year. These doses were significantly lesser (~ 8 times) than the dose limits established for personnel in NRB-99 [1].

Control of internal irradiation due to intake of uranium isotopes was carried out by analyzing the urine samples with ICP-MS technique [8]. Uranium content in the samples was in the range 0.06-1.9 mg/l. Average content was 1.4 mg/l, expected annual dose of internal irradiation– 0.2 mSv, i.e. one hundred times lesser than the dose limits established for personnel in NRB-99 [1]. Ratio $^{235}U/^{238}U$ was 0.0072. i.e. similar to the natural ratio of these isotopes. This fact showed that natural uranium was detected, which was naturally ingested (with food, liquid, etc.). Depleted uranium has isotope ratio of 0.003. The conclusion was that there was no internal intake of radionuclides while conducting the works.

Thus, measures on ensuring the radiation safety of personnel allowed to exclude high external radiation doses and to exclude inhalation intake of the radionuclides.

3. CURRENT RADIATION STATE OF THE STORAGES AND ADJACENT TERRITORY

As a result of the elimination-conservation activities including utilization of generated radioactive wastes and their disposal, as well as environment protection measures, radiation situation in the storages and at the adjacent territory was restored up to normal level. EDR distribution at the storages and adjacent territory is given in Figure 14.

EDR range at the territories under study was:

- Adjacent territory – from 0.18 to 0.24 μSv/h;
- Territory of storage # 9 – from 0.18 to 0.29 μSv/h;
- Territory of storage # 18 – from 0.15 to 0.45 μSv/h.

EDR of adjacent territories doesn't exceed 0.24 μSv/h. Higher levels of EDR at some areas of storage # 18 were caused by presence of uranium combustion products incorporated into concrete floor constructions. Constructions were handed over to MD RK representative for following dismantling and utilization in general fashion.

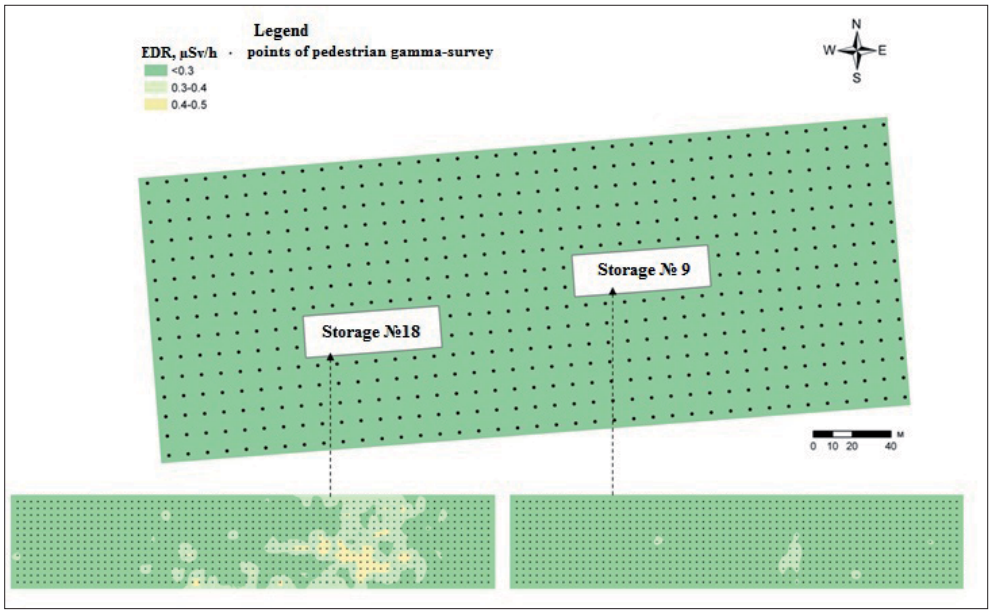

Figure 14. Distribution of EDR at the storages and adjacent territory

CONCLUSION

There were successfully completed the activities on elimination of danger of radioactive contamination formed as a result of the fire accident in the territory of military unit 89533 (Tokrau). All the existing radioactive wastes were utilized and placed for long-term storage at complex "Baikal" of Institute of Atomic Energy NNC RK (Kurchatov).

The results of radiological survey and comparative spectrometer analysis of the samples before/after treatment showed that radiation danger in storages #9 and #18 and adjacent territories was eliminated. EDR of adjacent territories didn't exceed 0.24 μSv/h. Higher levels of EDR at some areas of storage #18 are caused by presence of uranium combustion products incorporated into concrete floor constructions. Specific activity of these constructions is lower that a level of radioactive wastes and doesn't pose any danger. Final survey of the site confirmed safety of the conducted activities and elimination of negative man-made influence on the existing ecosystem. During implementation of the activities the personnel involved didn't receive radiation doses exceeding the limits established by NRB-99 [1].

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