UDC 621.039.9 (571.56)

Radioecological Situation within the Area of Pacific Underground Nuclear Explosion "Kraton-3" (1978, Northwestern Yakutia)

S. YU. ARTAMONOVA

Institute of Geology and Mineralogy, Siberian Branch of the Russian Academy of Sciences, Pr. Akademika Koptyuga 3, Novosibirsk 630090 (Russia)

E-mail: artam@igm.nsc.ru

(Received May 13, 2011; revised October 10, 2011)

Abstract

Novel data are discussed concerning tritium content in surface, ground and underground waters in the area the accidental pacific underground nuclear explosion "Kraton-3" in August 24, 1978. Early studies on the object "Kraton-3" were mainly devoted to the determination of surface contamination in the taiga landscape at the moment of the explosion, and the further redistribution and migration of radionuclides from contaminated areas under the influence of exogenous and biogenic processes. For the first time, an issue was considered whether the underground source of radioactivity is opened. Basing on the analysis of radioecological and geochemical data, a conclusion is drawn that there occurs a significant delivery of radionuclides from the cavity of the explosion to the surface of the earth.

Key words: pacific underground nuclear explosion, Yakutia, radiogeoecology, geological environment, surface water, ground water, underground water, radionuclides and radionuclide mass transfer, tritium, environmental risk

INTRODUCTION

The problem of spreading radionuclides in the natural environment is one of the most urgent environmental challenges of our time. Within the period of 1965–1988, in the territory former Soviet Union there was 104 underground nuclear explosions performed (with no taking into account nuclear tests at special proving grounds): 72 of them were of national economic destination, whereas 32 of the explosions were aimed at seismic sounding of the Earth. Such explosions were named "pacific underground nuclear explosions" (PUNE). Owing to the worst case of wedging the radioactivity out, four of the explosions were recognized to be accidental.

The pacific underground nuclear explosion "Kraton-3" with the power of 22 kt (TNT equivalent) was performed for seismic sounding on August 24, 1978. The place of explosion was chosen at the right bank of the Markha River, 38 km to the east from the settlement of Aykhal (Western Yakutia), at a depth of 577 m in the clayey limestones and marls of Middle Cambrian age (Fig. 1, *a*). After one year, the total activity in the cavity of the explosion was equal to 240 thousand Cu, whereas after 31 years (on October 2, 2009), this value demonstrated a decrease down to 14.7 thousand Cu resulting from the natural decay process [1].

Breaking down in the technology of driving the attack borehole resulted in accidental releasing about 2 % of the total activity through the mouth of the borehole, which, according to our estimates, was equal to approximately 4800 and 294 Cu in one year and in 31 year after the explosion, respectively. The radioactive gas-dust cloud was moving in the northeastern direction (see Fig. 1, *b*), and its main

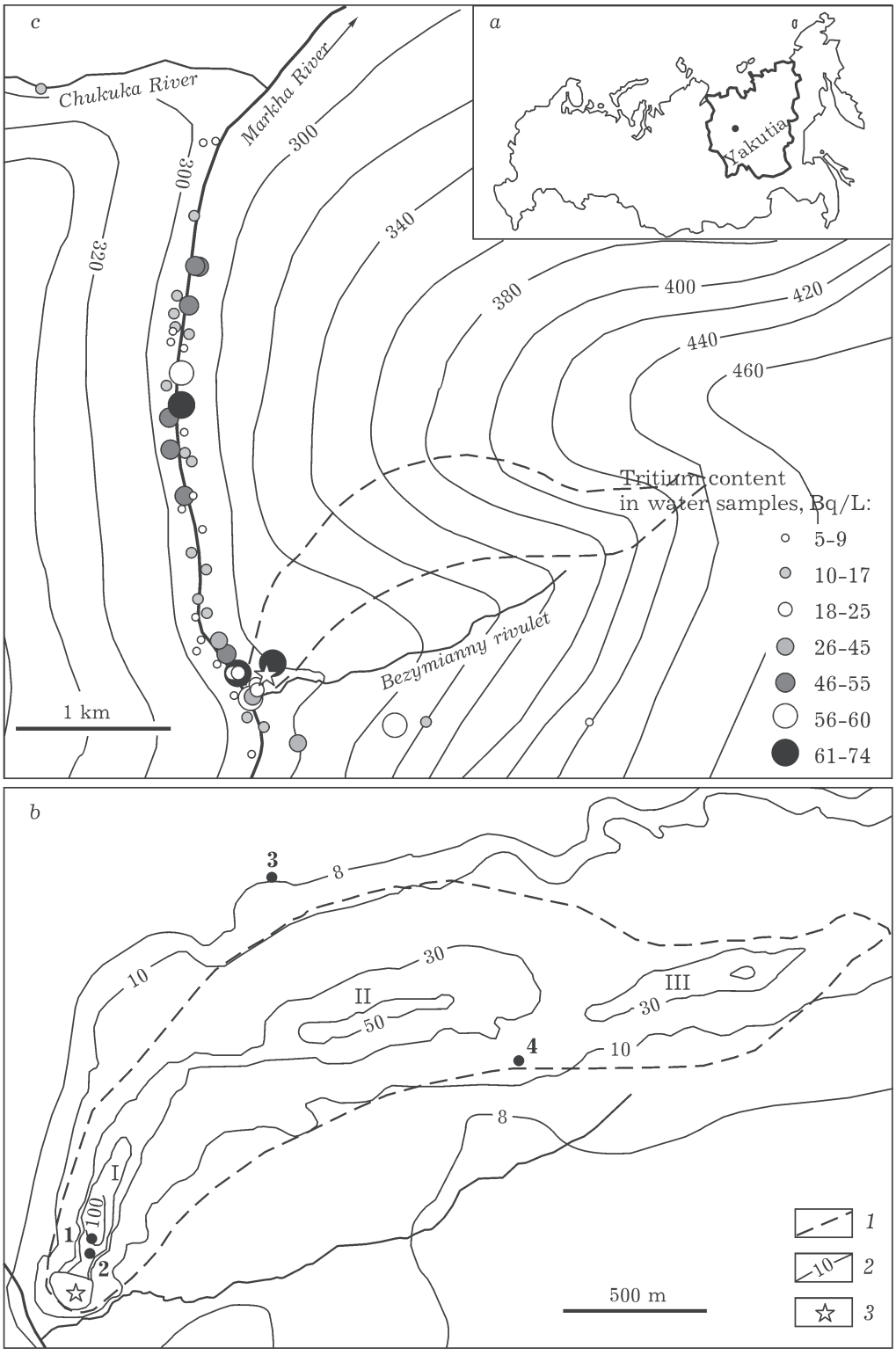

Fig. 1. Schematic map of the location of object PUNE "Kraton-3" (*a*) and its radioactive trace (*b*) on the right bank of the Markha River (*c*): *1* – boundary of the deadwood according to the results of decoding the satellite imagery Landcat (*b, c*); *2* – contour lines of γ-radiation exposure dose rate (μR/h) according to [4] (I – short-range trace, II – medium-range trace, III – long-range trace); *3* – the mouth of the attack borehole on the mound, **1–4** – soil and moss-lichen cover sampling points (Table 1).

activity fall at the first 3–4 km, which led to the death of the adjacent woodland of the northern taiga. The deceased forest, so-called deadwood, still represents a dead forest consisting of the Gmelin larch (*Larix Gmelinii*) with weakly sprouting low shrubs; There is almost everywhere moss and lichen cover developed (genera *Cladina*, race *Dicranum*, *Hylocomium*, *etc.*).

In 1981, on this site decontamination works have been carried out: a 10–15 m west of the mouth of the attack borehole was constructed combat pit volume of about 4000 m^3, which buried the contaminated drilling equipment, machinery and topsoil from the work site. In 2006 the lake, formed above the mouth of the attack borehole and a burial ground there was formed mound from the alluvium of local rivulet local about 1.5 m high and with the area amounting to 0.731 hectares.

Within the period from 1981 to the 1990s, the secret object of "Kraton-3" remained without supervision. In 1984, the geologists of Yakutia "discovered" the area of taiga with the radioactive cesium anomaly of unknown origin, however further work concerning the fact were not performed. Since 1993, at the object of PUNE "Kraton-3" there began radioecological studies [2, 3]. In 2002, the Ministry of Nature Protection of the Republic Sakha (Yakutia) carried out a detailed pedestrian radiometric and gamma-ray spectrometric survey within the "dead" forest [4]. This investigation was followed by studies on spreading ^{90}Sr, ^{60}Co, ^{241}Am, ^{238}Pu, $^{239, 240}Pu$ isotopes and their distribution throughout the components of ecosystems [5–8].

During the first year after the explosion of the majority of short-lived isotopes with induced and fission-fragment activity has decayed. Further, during a long period of time (up to 100 years after the explosion) one of the main contributors to the overall activity should be presented by tritium, and only later on the total radioactivity would be determined by long-lived fission-fragment radionuclides such as ^{90}Sr, ^{137}Cs. In the course of the period under reporting the activity of tritium demonstrated an approximately five-fold decrease as to compare with the initial radioactivity value, whereas the total fission-fragment activity exhibited a several orders of magnitude decrease. There are no data available from the literature concerning the chemical species of tritium to occur in the explosion-induced cavity. Under the surface conditions, the tritium occurs predominantly in the form of water molecules $^{3}H^{1}HO$ [9], which provides a high mobility of tritium. Thus within the areas of PUNE the tritium could serve as an indicator of occurring sources and dominant aqueous mass transfer directions for radionuclides.

The purpose of the present work consisted in establishing the relationship between the tritium content in naturally occurring waters within the region of the PUNE "Kraton-3" and the radioecological situation in general.

EXPERIMENTAL

The routine sampling of naturally occurring waters within the area PUNE "Kraton-3" was performed during the period of 2008–2009: six samples were taken within the first ten days of August, 2008, whereas 59 samples were taken within the first ten days of August, 2009. Water samples of 1 L in volume were preliminary filtered through a Blue Ribbon filter paper, being further acidified with concentrated nitric acid at a ratio of 1 mL of HNO_3 per 1 L of an aqueous sample. Under the laboratory conditions the samples were passed through membrane filters with the pore diameter of 0.2 μm. The tritium activity in water samples was measured by means of liquid scintillation spectrometry technique with the use of a Tri-Carb 2800 spectrometer (the USA). Samples were mixed with a scintillation cocktail in plastic vials (V = 20 mL) at a ratio sample/cocktail = 8 : 12. The mixtures were prepared held in a cool dark place for 48 h for stabilizing, with further determining the content of tritium therein [10]. The minimum detectable ^{3}H activity was equal to 1 Bq/L; when the ^{3}H activity was higher than 40 Bq/L the relative error did not exceed 6 %, the activity of 10–40 Bq/L resulted in the determination error ranging within 10–15 %, whereas the activity lower than 10 Bq/L gave the relative average error amounting to 34 %.

Within the first ten days of 2009, we took the samples with the volume up to 80 L each in order to determine the ^{90}Sr, $^{239, 240}Pu$, ^{238}Pu

therein those were preserved with concentrated nitric acid at a ratio of 1 mL of HNO_3 per 1 L of sample. Under the laboratory conditions, the samples were previously concentrated by means of a radiochemical method. The activity of ^{90}Sr was determined according the isotope ^{90}Y by means of β-radiometry employing a RUB-01P radiometer with the use of a low background detection unit BDZhB-06P. The sensitivity of the method is equal to 0.01 Bq. The relative error did not exceed 10 %. The activity of Pu isotopes was determined by means of α-spectrometry method using a 7184 EURISYS MEASURES single-channel α-spectrometer (France) [11]. In order to register α-radiation we used high resolution solid state detectors (SSD) such as PLUS 300-15 with an active area equal to 300 mm^2 and 15 keV resolution. The detection limits for Pu isotopes are at a level of $5 \cdot 10^{-4}$ Bq at a measurement time of $2 \cdot 10^5$ s. Standard ^{42}Pu and ^{236}Pu solutions were used.

The samples of soil and vegetation, after preliminary ashing were radiochemically concentrated, with the further determination of radionuclides by means of α-spectrometric method for the isotopes of Pu and with the help of β-radiometric method for ^{90}Sr [11]. Additionally, for the solid samples we measured the activity of ^{137}Cs by means of direct semiconductor gamma-spectrometry with an EURISYS MEASURES EGPC 192-P21/SHF 00-30A CLF-FA well-type SSD detector (France). The effective volume of the detector was equal to 220 cm^3, the relative efficiency being of 47.5 %. The limit of detection for radioactive cesium was equal to 1 Bq/kg; the relative error did not exceed 10 %. Depending on the sample activity the measurement time ranged from 4 to 24 h. In the area under investigation we carried out at least 2000 ground-based measurements of γ-radiation exposure dose rate (EDR) using a SRP-68-01 radiometer. Sampling points and field measurement points were referred geographically with the help of an ETREX GPS navigator and brought us into a GIS project of the area basing on satellite imagery Landcat performed in 2001.

The ionic composition of water samples was determined in 2008: the anions were determined by means of titration; the cations were determined using mass spectrometry with inductively coupled plasma employing an Agilent mass spectrometer (the USA). The determination error for ion concentration concentrations was less than 15 %.

RESULTS AND DISCUSSION

The area of deceased forest calculated on the basis of satellite imagery Landcat interpretation and GIS technology application amounted to 1 million 600 thousand m^2 (see Fig. 1), the length was equal to 3.6 km, although earlier the area was estimated to be approximately 1 000 000 m^2 [3].

The activity within the "deadwood" is distributed unevenly: the distribution of EDR formed three spots, so-called short-range trace, medium-range trace and long-range trace (see Fig. 1, *b*) [4].

It was found that in 2008 the EDR within the short-range trace ranges from 30 to 130 μR/h, within the medium-range trace this value ranges from 30 to 70 μR/h, ranging from 30 to 55 μR/h within the long-range trace. The short-range trace is contaminated to a greatest extent. As the distance from the borehole increases the EDR value exhibits a gradual decrease, which is confirmed by ground radiometric surveys and laboratory analyses (Table 1).

In the wood of dead trees the ^{90}Sr content is only about 6 Bq/kg, the ^{137}Cs content being equal to 10–14 Bq/kg, the $^{239,240}Pu$ content amounting to 0.041–0.064 Bq/kg [5].

The content of ^{90}Sr in the fallen bark of dead trees appeared three orders of magnitude greater to be equal to 200–490 Bq/kg, whereas the concentration of ^{137}Cs and $^{239,240}Pu$ was on the average an order of magnitude higher (120–140 and 0.6–5.0 Bq/kg, respectively). After falling the contaminated bark the major depositors of radionuclides within the area PUNE "Kraton-3" are presented by a moss-lichen covering and soils, namely the forest litter and the upper part of the humus horizon of soils (see Table 1). In the region composed by Ordovician and Cambrian carbonate rocks there are developed low-thickness permafrost sod-calcareous soils [6, 12]. The thickness of the forest litter is limited by 2–3 cm, the thickness of the humus horizon amounts to only 1–6 cm. The field of EDR is formed mainly due to the presence of radioactive cesium therein:

TABLE 1

Specific activity level of radionuclides in the samples of the object PUNE "Kraton-3", Bq/kg

Sample, sampling depth	^{90}Sr	^{137}Cs	$^{239,240}Pu$	^{238}Pu
Short-range trace, EDR 93 μR/h, 200 m to the north from the mouth of the attack borehole (see Fig. 1, point 1)				
Moss	ND	12 000±300[a]	ND	ND
Lichen	ND	201 100±2700[a]	7400±700[b]	390±40[b]
Forest litter, 0–2 cm	44 400±4600[a]	20 000±2200[a]	380±20[b]	23±2[b]
Humus horizon, 2–5 cm	5800±640[a]	5030±510[a]		
Soil, cm:				
5–10	3950±420[a]	31±4.1[a]	ND	ND
10–15	1440±160[a]	15±2.8[a]	ND	ND
Local anomaly at the skirt of the deadwood, EDR 420-460 μR/h (see Fig. 1, point 2)				
Forest litter, 0–2 cm	257790[c]	94579[c]	5900±300[b]	290±30[b]
	188 000±21 000[a]	86 700±8700[a]		
Humus horizon, cm:				
2–4	40610[c]	34089[c]	ND	ND
4–6	8715[c]	990[c]	ND	ND
6–8	2353[c]	48[c]	ND	ND
Soil, cm:				
8–14	194[c]	43[c]	ND	ND
14–21	18[c]	10[c]	ND	ND
Medium-range trace, the northern boundary of the deadwood, EDR 8 μR/h, 1.6 km from the mouth of the attack borehole (see Fig. 1, point 3)				
Moss	123.5±11.5	250±18	ND	ND
Lichen	44±3.5	135±10	ND	ND
Forest litter, 0–2 cm	98±4.9	56.2±18	5.1±0.4	<0.05
Humus horizon, 2–4 cm	46±3.3	11.5±1.5	3.8±0.15	<0.05
Soil, 4–15 cm	26±2.5	<1.5	<0.1	<0.05
Medium-range trace, the southern boundary of the deadwood, EDR 11 μR/h, 1.75 km from the mouth of the attack borehole (see Fig. 1, point 4)				
Moss	221±16	3150±220	21±1	<0.05
Lichen	102±10	1500±100	31±1.5	<0.05
Forest litter, 0–2 cm	1233±85	1030±75	15±1.0	<0.05
Humus horizon, 2–6 cm	850±70	740±60	12.5±0.8	<0.05
Soil, 6–15 cm	280±20	35±3.5	<0.1	<0.05
Background area, EDR 5 μR/h, 300 m upstream of the Markha River, right riverbank				
Lichen	75±6	90±7	2.7±0.5	<0.05
Forest litter, 0–2 cm	80±6	50±3.5	2.7±0.5	<0.05
Humus horizon, 2–5 cm	71±5.5	46±3.3	1.2±0.2	<0.05
Soil, 5–15 cm	33±3.0	<1.5	<0.1	<0.05

Note. ND – no data.

[a] Samples and 2001 [7, 8, 16].

[b] Samples of 2002 [14].

[c] Samples of 2003 [6].

the correlation coefficient between the EDR and the content of radioactive cesium in the sample was equal to 0.87 for data sample presented by 129 parallel γ-spectrometric and radiometric measurements. For the soils and the moss-lichen covering the total activity of ^{90}Sr is almost twice greater as to compare with the activity of ^{137}Cs and about two orders of magnitude higher comparing to the activity of ^{239,240}Pu. The ^{238}Pu activity amounts to approximately 5–6 % of the activity ^{239,240}Pu. According to the present classification [13], the upper horizons of soils and the moss-lichen covering in the central parts of the short-range and medium-range traces represent solid radioactive waste according to the content of radionuclides therein.

The radiometric survey performed allowed us to determine that the naturally occurring EDR in the undisturbed natural landscapes is equal to only 5–6 μR/h, which is inherent in limestone and dolomite species. The EDR values equal to 8–12 μR/h determined for the exterior borders of the deadwood indicate the presence of radioactive contamination in the edge zones: mosses exhibit the content to amount up to, Bq/kg: (123.5±11.5) for ^{90}Sr, and (250±18) for ^{137}Cs, whereas for the forest litter the content of the radionuclides is as it follows: ^{90}Sr – (98±4.9), ^{137}Cs – (56.2±18), ^{239,240}Pu – (5.1±0.4) Bq/kg (see Table 1). In addition, the same values of EDR (8–12 μR/h) registered for small areas of living forest to the south from the deadwood, those represent local contamination spots. To all appearance, they were formed from a substance, carried by a wind from the main radioactive cloud heading in the eastern direction.

All these years, under the influence of exogenous and biogenic processes there occurred the redistribution of radioactivity activity, the drift and redeposition of radionuclides, both within the deadwood, and within the neighbouring terrains, against the background of decreasing the total level of radioactivity caused by natural decay process.

In 2008, 145 m to the north from the mouth of the attack borehole, there was contoured a local anomaly with the EDR value ranging within 400–420 μR/h located in a small closed depression on a gentle slope near the skirt of the deadwood (see Fig. 1, *b*, point **2**). The size of the anomaly was equal to about 3 × 3 m. In 2001, as far as the forest litter and the first centimetres of the upper humus horizon are concerned, the authors of [7, 8] revealed a very high activity level (Bq/kg): ^{90}Sr 188 000, ^{137}Cs 86 700. The investigations performed in 2003 demonstrated [6] that the content of ^{90}Sr appeared 37 % higher, and that of ^{137}Cs became 9 % higher as to compare with data obtained in 2001 (see Table 1). This indicates the fact that there occurs a continued accumulation of radionuclides those are carried by surface water-flows from the territory of the deadwood. In addition to the radioactive cesium and strontium in the soils of this anomaly, in 2002 there were also Pu isotopes determined: the activity level for ^{239,240}Pu amounted to 5900 that for ^{238}Pu was equal to 290 Bq/kg [14].

There is no doubt that the abnormal area at the skirt of the deadwood could be considered an intermediate step only on the way of the further migration of radionuclides. The deadwood is located in the catchment basin of the Markha River and the Bezymyanny rivulet (see Fig. 1). The surface water flows from the medium-range and long-range traces under determining by local relief (see Fig. 1, *c*) fall into the Bezymyanny rivulet; the water flows from the most contaminated short-range trace flow directly into the Markha River, partly falling into the same Bezymianny rivulet. In the little rivulet that flows out of the skirt of the forest, 68 m to the south from the anomalous zone (Fig. 2, point 6-08) there is a high content of tritium ((68±8) Bq/L) determined, which is almost 13 times higher than the local background level. The latter, according to our data, ranges from (5±2) up (6±2) Bq/L in the Markha River upstream, and in small rivulets, which values are comparable with the global level. So, according to [9], the contemporary man-caused tritium background in surface waters is equal to about 5 Bq/L. The background content of tritium in the Yenisey River waters is of the same value [10].

For the water of the small rivulet (see Fig. 2, point 6-08) the overall β-activity amounts to (2.11±0.08) Bq/L, which more than twice exceeds the intervention level (IL). The total α-activity is equal to (0.54±0.06) Bq/L, which is almost twice greater than the IL [15].

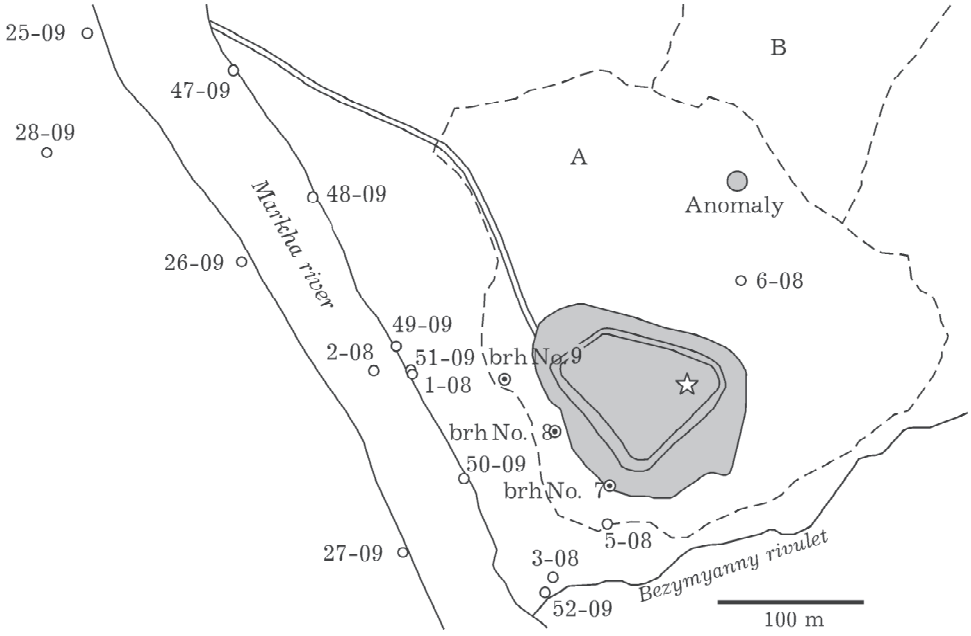

Fig. 2. Schematic map of sampling the naturally occurring waters in the region of PUNE "Kraton-3". Dashed line denotes the boundaries of working platform (A), deadwood (B); gray background denotes the mound, asterisk marks the location of the attack borehole mouth. The figures neat the circles denote the numbers of sampling points.

In May of 2002, there was a sample taken from the flood waters of the Bezymyanny Rivulet, wherein the following gross content of the main radionuclides was determined (together with the suspension) (Bq/L): ^{90}Sr (40.2±12.9), ^{137}Cs (6.2±0.6) and (5.6±0.6), ^{239,240}Pu (0.14±0.01), ^{238}Pu (0.007±0.001) [8, 16]. In August of 2009, we obtained the following data for the waters of the mentioned rivulet (filtrate), Bq/L: ^{90}Sr (0.12±0.004), ^{239,240}Pu $(4\pm0.16)\cdot10^{-4}$, ^{238}Pu $(3.9\pm0.15)\cdot10^{-3}$.

For the samples of the flood water from the Bezymyanny rivulet taken in 2002 as well as for those taken in 2009, the ratio values were obtained for the radionuclides such as ^{90}Sr/^{137}Cs ~ 6.5, ^{90}Sr/^{239,240}Pu ~ 300, which appeared higher as to compare with the ratio between these radionuclides in their major sources such as soil and moss-lichen cover within the deadwood (~2 and 100, respectively). This indicates the fact that there is a high mobility of aqueous radioactive strontium and its mass transfer observed predominantly with water flows mainly in solution, including the flood season.

The ^{137}Cs/^{239,240}Pu ratio in the samples of floodwater of 2002 amounts to about 40–44, whereas the ^{238}Pu content is equal to approximately 5 % of the ^{239,240}Pu. These data are in a good agreement with the corresponding parameters for these radionuclides in of their main sources, the components of deadwood (the ^{137}Cs/^{239,240}Pu ratio on the average is equal to 32–39, the ^{238}Pu content amounting to 5–6 % of the ^{239,240}Pu content). To all appearance, in the course of flood the mass transfer of ^{137}Cs and Pu isotopes occurs mainly *via* mechanical drift. Within the rest time, the mass transport of radionuclides occurs mainly in liquid form due to the low flow rate values waterways under the conditions of flat-and-hilly terrain. In this case, a higher mobility is inherent in ^{238}Pu. The content of ^{238}Pu in the sample of the Bezymyanny rivulet water (2009) is comparable with that in the sample taken in spring of $(3.9\pm0.15)\cdot10^{-3}$ and $(7\pm1)\cdot10^{-3}$, respectively), being almost 10 times greater than the content of ^{239,240}Pu. The latter is contained in the amount of $(4\pm0.16)\cdot10^{-4}$ Bq/L in the sample taken in summer of 2009, being contained in the amount of (0.14±0.02) Bq/L in the floodwater sample of 2002 (Table 2).

In 2008, for the Bezymyanny rivulet (see Table 2, point 3-08) we registered the ^{3}H activity equal to (30±5) Bq/L, which is two times

TABLE 2

Content of radionuclides in naturally occurring waters sampled near the mouth of the attack borehole of the PUNE "Kraton-3" in 1996–2009, Bq/L

Sampling place	Sample No.	Sampling date	^{3}H	Total β-activity[e] (^{90}Sr)	Total α-activity (^{239,240} Pu)	^{238}Pu
Surface water flow at the skirt of the deadwood	6-08	03.08.08	68±8	2.11±0.08 (ND)	0.54±0.06 (ND)	ND
	6a-08	03.08.08	ND	2.25±0.25 (ND)	ND	ND
Bezymyanny rivulet	–	May 2002[a]			(0.14±0.02)	$(7\pm1)\cdot10^{-3}$
	3-08	03.08.08	30±5	0.27±0.04 (ND)	0.023±0.02 (ND)	ND
	52-09	10.08.09	57±2.8	(0.12±0.004)	$(4\pm0.16)\cdot10^{-4}$	$(3.9\pm0.15)\cdot10^{-3}$
Pond near the mouth of attack borehole (before constructing the mound in 2006)	–	1996[b]	700±250	ND	ND	ND
	–	2001[b]	320±32	38±6 (ND)	ND	ND
	–	2003[c]	ND	49±18 (ND)	ND	ND
Surface water flow from under the mound of the attack borehole	5-08	03.08.08	18±5	0.398±0.028 (ND)	0.29±0.01 (ND)	ND
Ground water from borehole No. 8, depth ~1.5 m, to the west of the attack borehole mound borehole	brh No. 8	2007[d]	ND	(8.75±1.63)	ND	ND
Ground water from borehole No. 7, depth ~1.5 m, near the attack borehole No. 7	brh No.7	03.08.08	19±6	2.39±0.07 (ND)	0.16±0.04 (ND)	ND
Slope groundwater from a test pit on the riverbank of the Markha River	1-08	03.08.08	22±6	2.35±0.07 (ND)	0.15±0.04 (ND)	ND
	51-09	10.08.09	67.2±1.9	(1.99±0.07)	$(4\pm0.16)\cdot10^{-4}$	$(7.8\pm0.3)\cdot10^{-3}$
Markha River	50-09	10.08.09	13±2.0	ND	ND	ND
	2-08	03.08.08	22±5	2.82±0.03 (ND)	0.42±0.02 (ND)	ND
	49-09	10.08.09	14±1.5	ND	ND	ND
		11.08.09	48±2.4	(0.078±0.003)	$(6\pm0.24)\cdot10^{-4}$ (ND)	$(18.6\pm0.7)\cdot10^{-3}$
	48-09	10.08.09	52.1±2.6	ND	ND	ND
		11.08.09	45.2±2.1	ND	ND	ND
	47-09	11.08.09	44.3±2.2	0.011±0.010 (ND)	0.026±0.013 (ND)	ND
	27-09	09.08.09	9±1	ND	ND	ND
	26-09	09.08.09	5±2	ND	ND	ND
	25-09	09.08.09	6±2	ND	ND	ND
Water flow on the left riverbank of Markha River	28-09	09.08.09	7.5±0.5	ND	ND	ND
Water IL			7700	1 (5)	0.2 (0.56)	0.6

Notes. Sample numbers correspond to the points in Fig. 2. 2. ND – no data.

[a] According to [16].

[b] According to [8].

[c] According to [17].

[d] According to [18].

[e] Excluding the contribution of ^{3}H.

lower than that for a little rivulet at the edge of the deadwood (see Fig. 2, point 6-08). The effect could be, to all appearance, caused by a high water content of the Bezymyanny rivulet, and consequently by the dilution of the effluent water. In rainless 2009 the little rivulet at the edge of the deadwood got dry and the Bezymyanny rivulet became badly shallow thus the content of ^{3}H therein became almost two times higher as to compare with that in 2008 to amount to (57±2.8) Bq/L (see Table 2).

It is obvious that the permanent water mass transport of radionuclides from the deadwood occurs by means of surface water flows, thereby the mechanical drift of Pu isotopes and ^{137}Cs is significant only during the flood season.

For the waters of the Markha River near the object PUNE "Kraton-3", the following content of radionuclides was established (Bq/L): ^{90}Sr 0.078±0.003, $^{239,240}Pu$ (6±0.24) · 10^{-4}, ^{238}Pu (18.6±0.7) · 10^{-3}, ^{3}H (13±2.0)–(52.1±2.6) (see Fig. 2, Table 2, points 48-09–50-09). For the water of the Markha River the ratio $^{90}Sr/\ ^{239,240}Pu$ = 130, whereas the content of ^{238}Pu is 31 times greater than the content of $^{239,240}Pu$. It is a rather surprising fact that the waters of the Markha River exhibit the ^{238}Pu activity to be 10–30 times higher than the activity of $^{239,240}Pu$. This demonstrates a great aqueous mobility of the ^{238}Pu isotope (see Table 2) being fundamentally different from the data for lichens, deadwood soils (see Table 1) and for the samples of the flood waters of the Bezymyanny rivulet obtained in 2002 (see Table 2). According to our estimates, the flow rate of the Markha River is about 10–20 times greater than the flow rate of the Bezymyanny rivulet (taking into account the flow velocity, depth, watercourse width values we measured). In this regard, after mixing with the water the Bezymyanny rivulet as the main supplier of radionuclides, the tritium content should amount to only 8.4–8.8 Bq/L. However, in the waters of the Markha River set high tritium content – up to 52 Bq/L (see Table 2, points 47-09, 48-09, 49-09), which is comparable with the tritium content in the effluent water from the short-range trace in the deadwood (see Table 2, p. 6-08) and with that in the Bezymyanny rivulet (p. 52-09). Such high amounts of tritium in the full-flowing Markha River, *i. e.* the absence of any dilution effect (see Table 2), appeared an unexpected fact that requires for further studying.

Not only the water flows from the contaminated deadwood fall into the Markha River, but also water flows from the burial of contaminated equipment do, which burial is located near the mouth of the attack borehole. In 2008, on the surface of the rivulet flowing out of the mound, there were multicoloured spots of fuels and lubricants found, which could be most likely caused by washing-out from the disposal of the equipment buried. For the sample from this rivulet (see Fig. 2, p. 5-08, and Table 2), the following values of radioactivity were determined (Bq/L): tritium content 18, total β-activity level (0.398±0.028), which is almost by a factor of 2 less than the IL value [15]; α-activity level (0.29±0.01), which higher than the IL value [15].

TABLE 3

Total chemical composition of naturally occurring waters from the region of PUNE "Kraton-3", mg/L

Sampling place	pH	Ca^{2+}	Mg^{2+}	Na^{+}	K^{+}	HCO_3^{-}	SO_4^{2-}	Cl^{-}	*M*
Water flow on the skirt of the deadwood	8.0	63.5	27.95	<0.125	0.85	335.6	<1	3.55	432
Bezymyanny rivulet	8.22	53.3	26.7	<0.125	0.25	311	<1	1.8	393
Borehole No. 7	ND	11.5	6.4	145	4.25	98	<1	250	515
Borehole No. 8	ND	20.6	17	143	5.55	73	4.94	298	562
Water flow from under the mound	ND	51.5	25.65	2.65	0.9	311.2	<1	3.55	315
Slope runoff	ND	73.9	35	2.28	0.53	360	<1	39	511
Markha River	7.65	39	22	<0.125	0.43	229	<1	1.8	292

Note. ND – no data, *M* – water mineralization level.

For a pond formed after the explosion near the mouth of the attack borehole (which pond existed until constructing a mound in 2006), the activity of tritium was determined in 1996 to be equal to (700±250) Bq/L, whereas in 2001 the mentioned parameter already amounted to (320±32) Bq/L [8]. In 2001 and 2003, this pond water exhibited the content of ^{90}Sr equal to (38±6) [8] and (49±18) Bq/L, respectively [17], which is almost 7.5–10 times greater than the value of IL. Down the hill, for the groundwater, the radionuclide content determined in 2007 for the dissolved ^{90}Sr ((8.75±1.63) Bq/L) is almost 1.5 times higher than the IL value [18]. In 2008–2009 we determined the following content of radionuclides in groundwater (see Fig. 2, points 1-08 and 51-09) (Bq/L): ^{90}Sr (1.99±0.07), ^{3}H (22±6)–(67.2±1.9), ^{239,240}Pu (4±0.16) · 10^{-4}, ^{238}Pu (7.8±0.3) · 10^{-3}. The values obtained are lower than the IL value (see Table 2). In addition to the water flows from the deadwood and from the burial ground, there could occur a third source of radionuclides. Let us recollect that the emergency object PUNE "Kraton-3" is a complicated radiogeoecological object. In fact, the cavity of the explosion represents an uncontrolled underground point for depositing the radioactive waste wherefrom radionuclides could leak to the surface. Further, let us consider the ionic composition of naturally occurring waters.

The local surface waters represent alkalescent magnesium-calcium hydrocarbonate ones (Table 3) caused by the composition of the underlying rocks of the region such as limestones and dolomites. The Kurlov formula describes them as it follows:

$$M\,170-190\,\frac{\mathrm{HCO_3^-}\,100}{\mathrm{Ca^{2+}}\,74\,\mathrm{Mg^{2+}}\,25\,(\mathrm{Na^+},\mathrm{K^+})1}\;\mathrm{pH}\,7.2-8.0$$

In the case of the same ionic composition there is a successive decrease in the mineralization level in the order from a small water low to a greater one as it follows (mg/L): water flows from the deadwood 430, the Bezymyanny rivulet 330–390, the Markha River 290–170, which could be explained by increasing the dilution effect with increasing the flow rate of water flows in this chain.

Three monitoring boreholes near the mound of the object PUNE "Kraton-3" were drilled to a depth up to 2.5–3 m (see Fig. 2); among those only borehole Nos. 7 and 8 are able of capturing the waters of the seasonally thawed layer. The latter are mainly formed by surface and meteoric waters, mainly in the years of exceeding the mean annual precipitation level (high-water years). The third borehole (No. 9) was drilled unsuccessfully (it is dry). The groundwater of the two boreholes (see Table 3, Fig. 2) with mineralization level amounting to 515–560 mg/L demonstrated a high concentration of chlorides (up to 250–298 mg/L) and so-

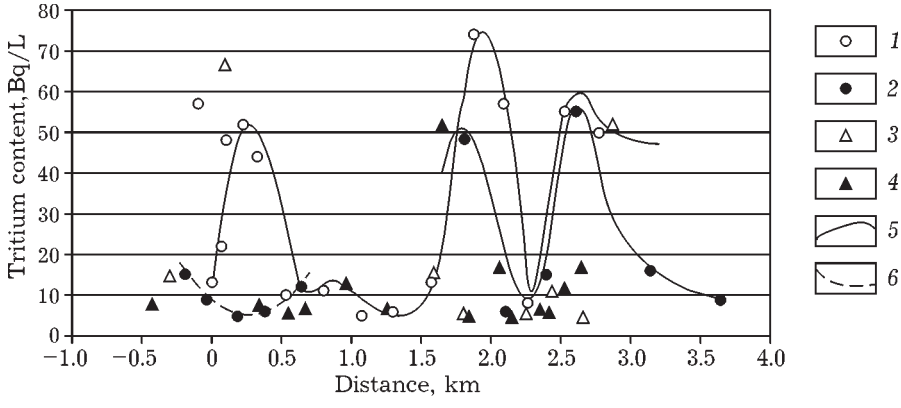

Fig. 3. Tritium distribution in naturally occurring waters within the valley of the Markha River (point 50-09 located on the slope near the mouth of the attack borehole was taken as zero point: 0–4 – downstream distance, 0…–1 – upstream distance): *1* – river water from the right riverbank; *2* – river water from the left riverbank; *3* – water of small rivulets and hollows flowing down from the right riverbank of the Markha River; *4* – water of small rivulets and hollows flowing down from the left bank of the Markha River; *5*, *6* – curves of anomalies revealed according to the content of tritium in water samples taken near the right (*5*) and left (*6*) riverbanks.

dium (up to 143–145 mg/L); they exhibit hydrocarbonate-chloride water type.

$$M\ 515\frac{Cl^-\ 60\ HCO_3^-\ 40}{Na^+\ 81\ Ca^{2+}\ 11\ Mg^{2+}\ 4\ K^+\ 4}\ \text{(Borehole No. 7)}$$

$$M\ 562\frac{Cl^-\ 68\ HCO_3^-\ 29\ SO_4^{2-}\ 3}{Na^+\ 69\ Ca^{2+}\ 17\ Mg^{2+}\ 9\ K^+\ 5}\ \text{(Borehole No. 8)}$$

It is known [19] that in the region there pressure saline water-bearing strata are developed. Data concerning the ion composition of water from the observation boreholes indicate the leakage of underground salt water along the column of the attack borehole to the surface. The authors have not another explanation for appearing sodium chloride in the groundwater under the mound of the object PUNE "Kraton-3". The presence of significant Na^+ and Cl^- concentrations in a rivulet from under the mound (see Fig. 2, point 5-08) as well as in the groundwater from a test pit at the foot of the slope on the riverbank of the Markha River (points 1-08, 51-09) also indicates the traces of leaked underground salt waters after mixing them with fresh groundwaters.

Thus, the analysis of data available indicates the fact that the activity level of water from the Bezymyanny rivulet and from small surface water flows near the skirt of the deadwood (upwards from the mound according to the relief) is formed due to the contamination resulting from the area of deadwood, the evident surface source of radioactivity. To all appearance, the activity level of the surface and ground water from the attack borehole and down the slope could be contributed also by other sources such as a radioactive burial ground occurring under the mound, underground saline water leaked through the borehole column (in the case when the latter are contaminated resulting from an underground source of the explosion cavity).

As far as the Markha River is concerned, whereto the ground waters flow down from the slope, the radionuclide content therein is even higher than in ground slope waters (Bq/L): $^{239,240}Pu$ $(6\pm0.24)\cdot10^{-4}$, ^{238}Pu $(18.6\pm0.7)\cdot10^{-3}$, tritium (52.1±2.6) (see Fig. 2, Table 2, points 48-09, 49-09), *i. e.*, the concentration of radionuclides does not exhibit any decrease. If the high activity level of radionuclides in the ground waters of the slope are quite expected and logically coherent, their high content in the river water could not be explained only by slope surface water-runoff. It might be assumed that there is a second way of leaking the radionuclides from the underground source such as the explosion cavity along the failure column formed resulting from the explosion and further through the fractured zone of the tectonic fissure with underground rising to the river. It is just occurring the permanent underwater rising the radioactivity under the slope the Markha River most likely determines the high content of radionuclides in the river water and an insignificant effect of dilution by the river water.

According to the authors of [20], the PUNE "Kraton-3" was performed in the immediate vicinity of a tectonic fissure line of north-northeast course (identified when interpreting aerophotogrammetric data), passing through the riverbed of the Markha River. It is known that in the course of drilling the attack borehole in 1977 at a depth of 165–186 m, a crumbling horizon was passed through that absorbed drilling fluid, which horizon, in our opinion, just represented the tectonic fracture zone. Thus, the accidental PUNE "Kraton-3" resulted in changing not only the surface landscape, but also the bowls and, in addition, the tectonic fracture could burst. The fact that the radionuclides leaked to the surface from the explosion cavity through a crumbling zone of the tectonic fissure could be revealed by means of routine sampling the naturally occurring waters in the territory of the region with further determination the tritium content therein, or with further performing so-called tritium mapping.

In 2009, we analyzed 59 samples from hollows and rivulets at the foot of the Markha River boards including the river water from both riverbanks up to 3.6 km downstream and up to 400 m upstream from the attack borehole of PUNE "Kraton-3" (see Fig. 1, curve *3*). Figure 3 demonstrates the distribution of tritium in the valley of the Markha River, at that we took point 50?09 as a zero mark on the horizontal axis, since it is located on a straight line from the mouth of the attack borehole of PUNE "Kraton-3". For the river water in point 50-09 the ^3H activity level amounts to (13±2.0)

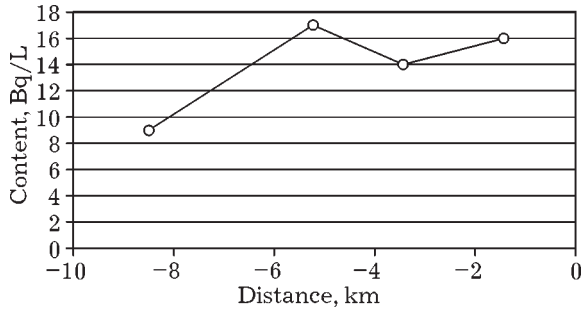

Fig. 4. Tritium content in the river water of the Chukuka River upstream from the mouth, located 4 km to the north of object PUNE "Kraton-3" taken as a distance reference point.

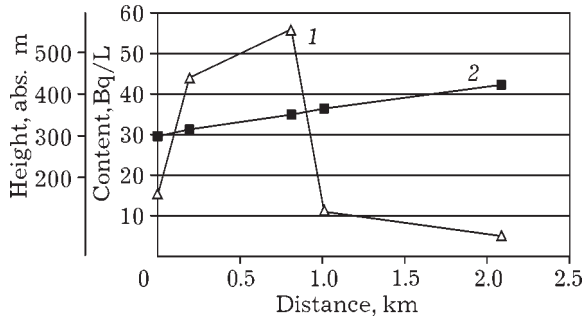

Fig. 5. Data concerning the content of tritium in the hollows along the profile "watershed-valley" for the right riverbank of the Markha River 500 m to the south of object PUNE "Kraton-3" (*1*) and the absolute height of sampling points (*2*).

Bq/L only (see Table 2 and Fig. 2). Approximately 100 m downstream from point 50–09 there was the first peak of high ^{3}H activity level (44–52 Bq/L) registered, which peak can be traced downstream for the space of another 225 m. As far as the waters near the opposite (left) riverbank at the same part of the river are concerned, we revealed therein a low level tritium activity ranging within 5–15 Bq/L, average 9.4 Bq/L. The maximum activity of ^{3}H equal to 74 Bq/L was determined for the river water near the right riverbank 1.9 km downstream from the object of PUNE. The river water near the left riverbank approximately at the same place (1.8 km downstream) also exhibits an abrupt peak of tritium activity up to 48 Bq/L with the background level amounting to 5–6 Bq/L. Nearby, in a small rivulet we registered the activity level of 52 Bq/L.

The third peak of tritium activity was registered 2.5–2.85 km downstream from point 50-09: for the river water from both riverbanks, there was an activity level determined amounting to 50–55 Bq/L. The ^{3}H activity in the hollow of the right riverbank thereat also amounted to 52 Bq/L (see Fig. 1, *a*, point **3**). The data obtained indicate the fact that there occurs permanent supplying the radionuclides from a local source, but the effluents from the deadwood in this case should be excluded due to the features of the terrain relief (see Fig. 1, *c*). In addition, elevated tritium content (up to 17 Bq/L) was revealed in the waters of the Chukuka River, the left tributary of the Markha River, which tributary flows into the latter 4 km downstream from the object of PUNE. This could indicate that there occurs a flow of radionuclides from the explosion cavity through the tectonic fissure that wedges out along the riverbed of the Markha River, which fault, to all appearance, could be connected with the Chukuka River. The level of tritium in the waters of the Chukuka River exhibits a decrease down to 9 Bq/L only at a distance 8.5 km upstream the Chukuka mouth (Fig. 4).

On the right bank of the Markha River 500 m to the south of the attack borehole within the living undisturbed forests (EDR = 5–6 μR/h, sometimes up to 8.9 μR/h), we investigated the waters from hollows along the profile of about 2 km long. It was found that the tritium activity therein also ranges over a wide range, from 5 to 56 Bq/h (Fig. 5).

CONCLUSIONS

From the data presented, it follows that the object PUNE "Kraton-3" represents a complicated radiogeological and ecological object that serves as a surface and underground source of radioactivity.

1. Moss and lichen layer, forest litter and the first few centimetres of soil humus horizon are the main depositors of surface radioactivity within the deadwood, a high content of ^{137}Cs, ^{90}Sr, $^{239,240}Pu$, ^{238}Pu therein in the near and middle traces can be attributed to solid radioactive wastes.

2. There is revealed a constant water-mediated mass transport of radionuclides presented mainly ^{90}Sr, ^{137}Cs , $^{238,239,240}Pu$, ^{3}H by means of surface water flows from the area of deadwood. Under the conditions of flat and

hilly terrain due to low water flow rate values the mass transfer of radionuclides occurs mainly in a liquid form, except for spring floods, in the course of those the mechanical drift of Pu isotopes and ^{137}Cs is significant. The content of ^{90}Sr, Pu and tritium isotopes in the samples of water from the Markha River and Bezymyanny rivulet taken in 2009 is much than the IL [15], however these values are 1–3 orders of magnitude higher than the background levels for Siberian rivers [10].

3. Shifting the ratio values for radionuclide activity allowed us to establish that the isotopes ^{137}Cs, ^{238}Pu are characterized by a high mobility in water.

4. The spatial distribution of tritium content as well as that for other radionuclides in naturally occurring waters within the region indicates the fact that there undoubtedly occurs water-flushing the radionuclides from the underground burial located near the mouth of the attack borehole, as well as permanent releasing the radionuclides from the explosion-produced cavity onto the surface of the earth, together with the pressure saline underground waters. Thereby the withdrawal of the radioactivity from the bowels occurs as it follows: 1) through a weakened part of the attack borehole of the object PUNE "Kraton-3" with releasing onto the surface near the mouth of the borehole, and 2) through the cracked zone of a tectonic fissure extended along the Markha riverbed, with wedging out along the Markha riverbed both close to the object PUNE "Kraton-3", and at the distance of 3 km. Leaking the radioactivity from the depths of the activity could, to all appearance, occur also in the valley of Chukuka River, a left tributary of the Markha River, 4 km downstream from the object of PUNE "Kraton-3", since the tritium content therein is 3.5 times higher as to compare with the global level.

REFERENCES

1 Artamonova S. Yu., Dubinin V. P., Kozhevnikov N. O., Razvorotneva L. I., Rudenko V. V., Seredovich A. V., Chernyshov A. K., Razrabotka Proekta Spetsialnogo Gornogo Otvoda na Obektakh Mirnykh Podzemnykh Yadernykh Vzryvov "Kristall" i "Kraton-3" (Mirninskiy Rayon): Otchet IGM SO RAN po Goszakazy No. 43(76-08) Ministerstva Okhrany Prirody Respubliki Sakha (Yakutia), Novosibirsk, 2009.

2 Gedeonov A. D., Kuleshova I. N., Bakushkina L. P., I Respubl. Nauch-Prakt. Konf. "Radiatsionnaya Bezopasnost Respubliki Sakha (Yakutia)" (Proceedings), Yakutsk, 1993, pp. 214–232.

3 Burtsev I. S., Kolodeznikova E. N., Radiatsionnaya Obstanovka v Almazonosnykh Rayonakh Yakutii, Yakutsk, 1997.

4 Kovalev V. K., Otchet Ministerstva Okhrany Prirody Respubliki Sakha (Yakutia), Novosibirsk, 2002.

5 Gedeonov A. D., Bulatenkov Yu. V., Shkroev V. Yu., II Respubl. Nauch-Prakt. Konf. "Radiatsionnaya Bezopasnost Respubliki Sakha (Yakutia)" (Proceedings), Yakutsk, 2004, pp. 168–180.

6 Chevychelov A. P., Sobakin P. I., Molchanova I. V., *Pochvovedeniye*, 12 (2006) 1512.

7 Ramzaev V., Mishine A., Basalaeva L, Brown J., *J. Environ. Radioact.*, 95 (2007) 107.

8 Ramzaev V., Mishine A., Golikov V., Argunova T., Ushnitski V., Zhuravskaya A., Sobakin P., Brown J., Strand P., *J. Environ. Radioact.*, 100 (2009) 1092.

9 Rikhvanov L. P., Radioaktivnye Elementy v Okruzhayushchey Srede i Problemy Radioekologii (School Book), STT, Tomsk, 2009.

10 Bolsunovsky A. Ya., Bondareva L. G., *J. Environ. Radioact.*, 55, 3 (2003) 285.

11 Sukhorukov F. V., Degermendzhi A. G., Belolipetskiy V. M., Bolsunovsky A. Ya., Kovalev S. I., Kosolapova L. G., Melgunov M. S., Raputa V. F., Zakonomernosti Raspredeleniya i Migratsii Radionuklidov v Doline Reki Yenisey, GEO, Novosibirsk, 2004.

12 Elovskaya L. G., Petrova E. I., Teterina L. V., Pochvy Severnoy Yakutii, Nauka, Novosibirsk, 1979.

13 SanPiN 2.6.1.2612–10. Osnovnye Sanitarnye Pravila Obespecheniya Radiatsionnoy Bezopasnosti (OSPORB 99/2010), Moscow, 2010.

14 Gedeonov A. D., Otsenka Urovney Radioaktivnogo Zagryazneniya Okruzhayushchey Sredy na Obyektakh "Kristall" i "Kraton-3" v Respublike Sakha (Yakutia): Otchet ZAO "Nauchno-Proizvodstvennaya Firma TreM" po Zakazy Ministerstva Okhrany Prirody Respubliki Sakha (Yakutia), St. Petersburg, 2003.

15 SanPiN 2.6.1.2523-09. Normy Radiatsionnoy Bezopasnosti (OSPORB 99/2010), Moscow, 2009.

16 Ramzaev V. P., Radiatsionnaya Bezopasnost Respubliki Sakha (Yakutia): Otchet NIR GU NIIRG Minzdrava Rossii po Goszakazu No. 2-2001 Ministerstva Okhrany Prirody Respubliki Sakha (Yakutia), St. Petersburg, 2002.

17 Ushnitskiy V. E., Otchet Ministerstva Okhrany Prirody Respubliki Sakha (Yakutia), Yakutsk, 2004.

18 Novgorodov P. G., Aleksandrov A. R., Nikashina V. S., Kats E. M., Serova I. B., Otchet Instituta Problem Nefti i Gaza SO RAN po Goszakazu No. 10 Ministerstva Okhrany Prirody Respubliki Sakha (Yakutia), Yakutsk, 2008.

19 Klimovskiy I. V., Gotovtsev S. P., Kriolitozona Yakutskoy Almazonosnoy Provintsii, Nauka, Novosibirsk, 1994.

20 Mikulenko K. I., Chomchoev A. I., Gotovtsev S. P., Geologo-Geograficheskiye Usloviya Provedeniya i Posledstviya Podzemnykh Yadernykh Vzryvov na Territorii Respubliki Sakha (Yakutia), Yakutsk, 2006.