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Assessment of the radiological situation near a mothballed uranium mining facility in North-East Kazakhstan

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In this study, we present radiometric measurements near the mothballed Grachevskoye uranium mining facility (MUMF) and the settlement of Saumalkol, located in northeast Kazakhstan. A pedestrian gamma spectrometric survey was conducted at locations of elevated gamma radiation. Spots with elevated gross activity values were detected in the northern part of the settlement and on a motor road. The pathways of radionuclides from the Grachevskoye MUMF to the area around the Saumalkol settlement were addressed. The migration ability of radionuclides and the process of radionuclide transport by ambient air did not have significant effects on the progression of the radioecological situation in the village. However, even after the mine was shut down, the concentration of uranium in the potable water intake well in the village increased eightfold (up to 240 µ/L). Radiological hazards were estimated to exceed recommended parameters.

Keywords Mothballed uranium mining facility, Grachevskoye deposit, Saumalkol settlement, Uranium, Radium, Thorium, Potassium, Lead, Soil, Ground water

The development of the nuclear industry and energy, complicated by several emergencies, disasters, and nuclear tests, has led to global pollution of the Earth's biosphere and, consequently, a higher total radioactive background. An essential role in the contamination of the territory of the Republic of Kazakhstan (RK) with radioactive substances was played by activities at the former Semipalatinsk test site (STS), which still contains numerous local spots with a highly radioactive background^{1–7}. The sites of numerous uranium deposits under development are also grave sources of radioactive contamination in the RK. Radioactivity studies in areas close to mining facilities are commonly conducted worldwide^{8–11}, as these facilities may pose a significant danger to the personnel and public health in the adjacent areas.

RK ranks second in the natural uranium inventory worldwide, with 13% of all explored world reserves being concentrated in RK. The country's reserves are estimated at more than 815.2 thousand tons, while the total inventory and resources of natural uranium amount to 1.4 million tons. Currently, uranium mined in the RK is exported to other countries, primarily China, Europe, and India. Approximately 80% of uranium products are produced by five countries namely Kazakhstan, Russia, Australia, Canada, and Nigeria. A decision was made to build and commission a nuclear power plant to ensure energy safety in RK. Therefore, the study of the behaviour of radionuclides in the environment has become particularly important for RK¹².

Until the mid-90s, uranium in the RK was mined primarily in the uranium-ore rich province of North Kazakhstan Region (NKR), where the main uranium inventory of endogenous (hydrothermal) deposits was concentrated. Due to a crash in the prices of uranium, followed by the suspension of production, virtually all waste storage facilities (mines) engaged in developing uranium deposits proved to be unattended by the mid-90s. The radioactive waste from the uranium mining industry, which has produced over 80 thousand tons of uranium, amounts to 222 million tons, with a total activity of 251 thousand Ci. Most of this is waste from mining and chemical facilities¹³.

Uranium mines were mothballed by the Republican State Enterprise (RSE) 'Uranlikvidrudnik'¹⁴ under the program 'Mothballing of uranium mining facilities and remediation after the development of uranium deposits for 2001–2010' to ensure the safety of the natural environment and the population. This company was entrusted with the responsibility of observing and monitoring radiation of remediated facilities periodically and

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undertaking recovery efforts to ensure their integrity. However, due to the cessation of the activities carried out by the RSE 'Uranlikvidrudnik' in 2010, monitoring studies and industrial inspection at the industrial sites of the mines were stopped and are no longer carried out. The situation is significantly complicated by the fact that there are no fences or signs of radiation danger around the uranium mines of the NKR today, and the access roads leading to their premises are open. Because resources are not managed within former mines, groundwater contamination due to uncontrolled input into mine workings is possible. Accordingly, the aquatic environment may become another source of radioactive contamination in nearby areas.

A reconnaissance (preliminary) survey has been conducted to assess the consequences of uranium deposits under development at several mobile uranium mining facilities (Voskhod, Viktorovskoye, Molodezhnoye, Sholpak, Kasachinoe, and Grachevskoye) and in settlements (Chistopolye, Toksan Bi, Shakpak, Novoukrainka, Sarytubek, and Soumalkol) located in adjacent areas (Fig. 1) in the southern NKR¹⁵.

Based on the preliminary survey results, Grachevskoye MUMF and Saumalkol regional centres were selected for detailed research. The Grachevskoye deposit is part of the Grachevsky ore hub and, according to the existing administrative division, is located in the NKR. The extraction of uranium ore from the Grachevskoye MUMF was performed using an underground mining method. Next, the ore was partially enriched by heap leaching, after which it was transported in containers by rail to the Stepnogorsk Mining and Smelting Plant¹³.

The main purpose of this study was to assess the radioactive contamination of the main environmental compartments on the premises of Grachevskoye MUMF and within the village of Saumalkol. This study addressed several possible pathways, such as, ore transportation by motor vehicles and rail, mobility of radionuclides in soils, by ambient air, and by the input of contaminated ground waters from the premises of mine workings to the ground waters of the settlement, to assess a possible radionuclide entry from the Grachevskoye deposit to the territory of the Saumalkol settlement.

Results and discussion

The radiological situation in the study area

EDR was measured across the perimeters of the mine, railway, and along the main streets of Saumalkol settlement. Open-pit construction was also explored. Consequently, local spots were identified at two or more times the background radiation on the daylight surface. The research findings on EDR distribution are depicted in Fig. 2.

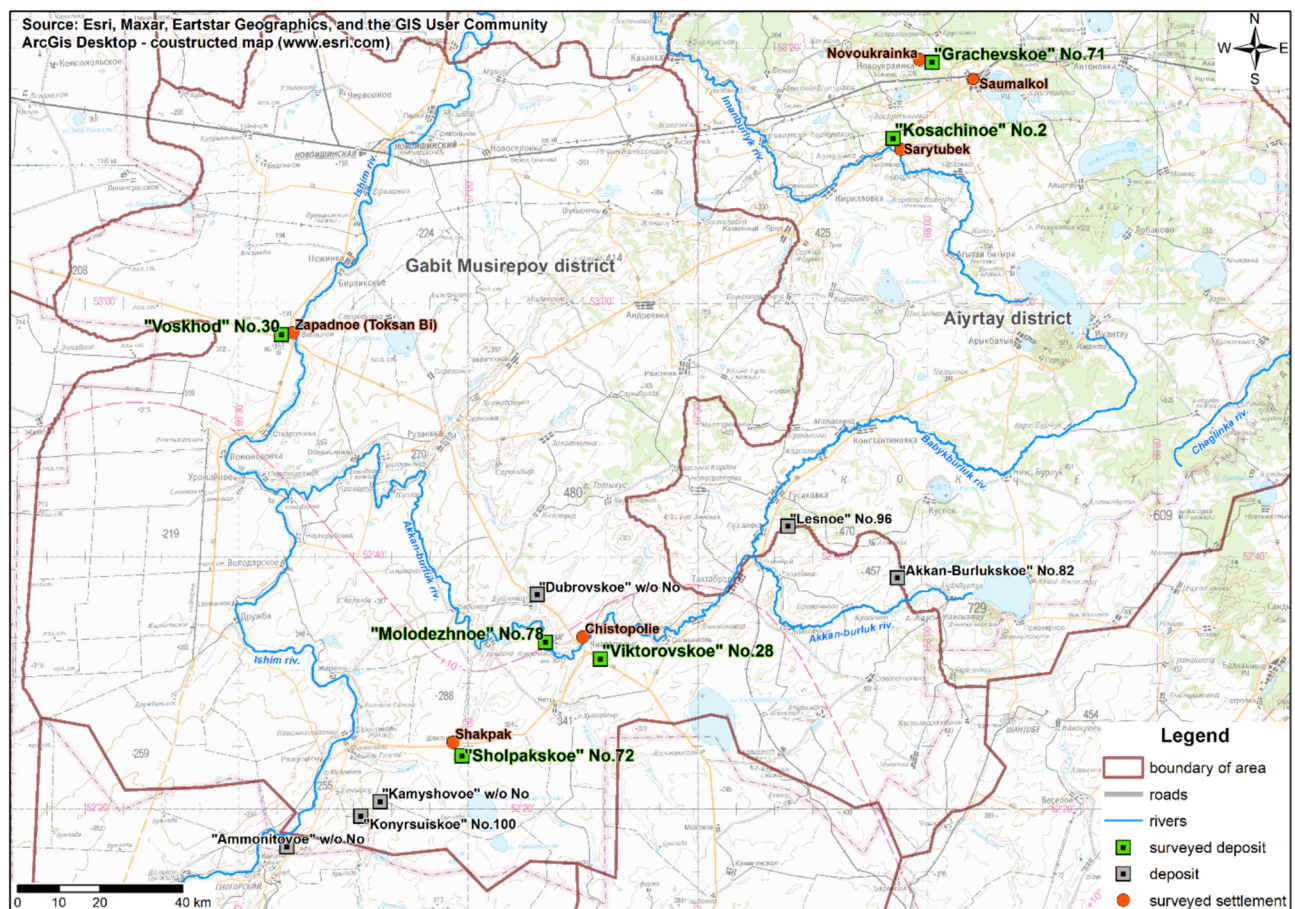


Fig. 1. Layout of explored Grachevskoye uranium mining facility (MUMF) and settlements in the southern parts of North Kazakhstan Region.

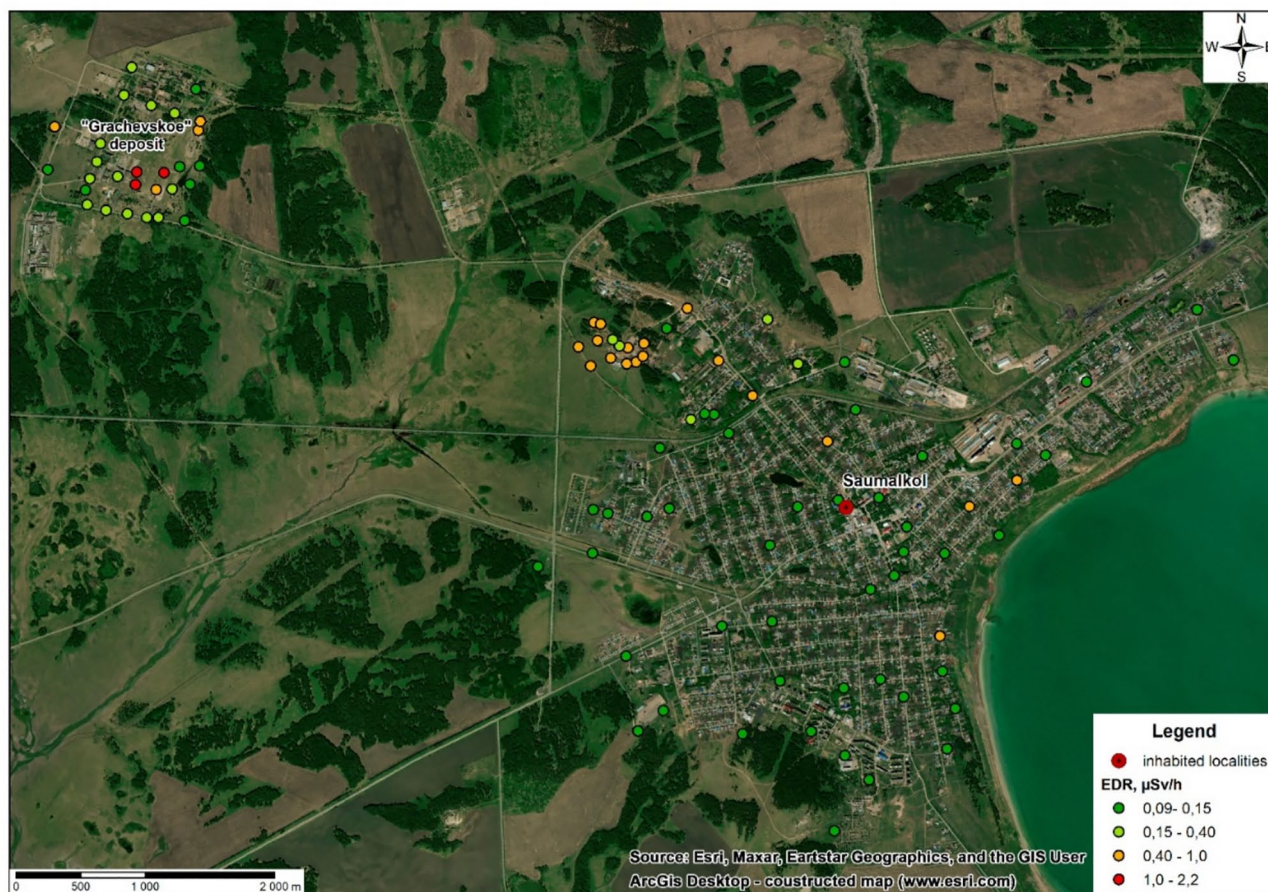


Fig. 2. Equivalent dose rate measurements.

The values of measured EDR in the territory of the settlement and the mine, except for several sites with higher than normal concentrations, varied from 0.09 to 0.80 $\mu\text{Sv/h}$, with a background of 0.09 $\mu\text{Sv/h}$ typical for this terrain. On the mine premises, outliers (from 1.13 to 14 $\mu\text{Sv/h}$) were noted in the central part.

Figure 3 shows the results of a pedestrian gamma spectrometry survey based on the premises of the Grachevskoye MUMF. The pattern of the areal distribution of gamma radiation by gross activity was established using a pedestrian gamma spectrometry survey. The values of gross activity ranged from 153 to 975 cps, with an average of 280 cps. The values were divided into five zones when plotting the map. The selected zones correspond to the following EDR values: < 200 cps for values from 0.09 to 0.12 $\mu\text{Sv/h}$; 200–300 from 0.12 to 0.20 $\mu\text{Sv/h}$; 300–500 from 0.20 to 0.40 $\mu\text{Sv/h}$; and 500–974.5 for values up to 2.0 $\mu\text{Sv/h}$. The entire surveyed site area was exposed to man-made contaminants to varying degrees. The zones of elevated values included values ranging from 300 to 500 cps. Commercial ore was dumped in a vast zone in the southern part of the site. Spots ranging from 1 × 1 m to 10 × 10 m in size with the highest values were identified in this zone. Within these spots, fragments of rocks up to 10 × 5 cm with EDR values of up to 14 $\mu\text{Sv/h}$ were found.

Figure 4 shows the results of a pedestrian gamma spectrometry survey at 10 sites in the southern part of Saumalkol Village. The gross activity values ranged from < 200 to 707 cps. The plotted map indicated that the lowest values ranging from < 200 to 250 cps were in the southwestern and central parts of the village. The highest values were observed in the northwestern part of the village near the open pit.

The results of the laboratory gamma-ray spectrometric analysis of the soil samples (Fig. S1) are listed in Table S1. The distributions of the activity concentrations of naturally occurring radionuclides ^{40}K , ^{238}U , ^{232}Th , ^{226}Ra , and ^{210}Pb in the topsoil are tabulated in Table S1 and depicted in Fig. 5. ^{238}U activity concentration in the topsoil of the Grachevskoye MUMF varied from 24 to 355 Bq/kg, with an average of 111 Bq/kg (Q2–120 Bq/kg). In the Saumalkol settlement, ^{238}U activity varied from 20 to 410 Bq/kg. The distribution pattern of ^{238}U activity concentration in the soil was similar at both research sites. The values of the content of ^{238}U in the soil of Grachevskoye MUMF and Saumalkol Village exceeded the maxima for Kazakhstani soils. The content of ^{232}Th and ^{226}Ra in the topsoil of Grachevskoye MUMF varied from 30 to 115 Bq/kg and from 11 to 960 Bq/kg, respectively. Within the Saumalkol settlement, ^{226}Ra ranged from 20 to 270 Bq/kg and ^{232}Th ranged from 50 to 820 Bq/kg. The maxima of ^{226}Ra activity concentration were well below those of Grachevskoye MUMF, but more than 2 times in RK, which is most likely caused by the close proximity of the settlement to the MUMF and its impact. Thus, the maxima of ^{232}Th activity concentration were essentially higher than those for the MUMF and nearly four times higher in the RK, which is an apparent geochemical anomaly. The mean of ^{40}K activity

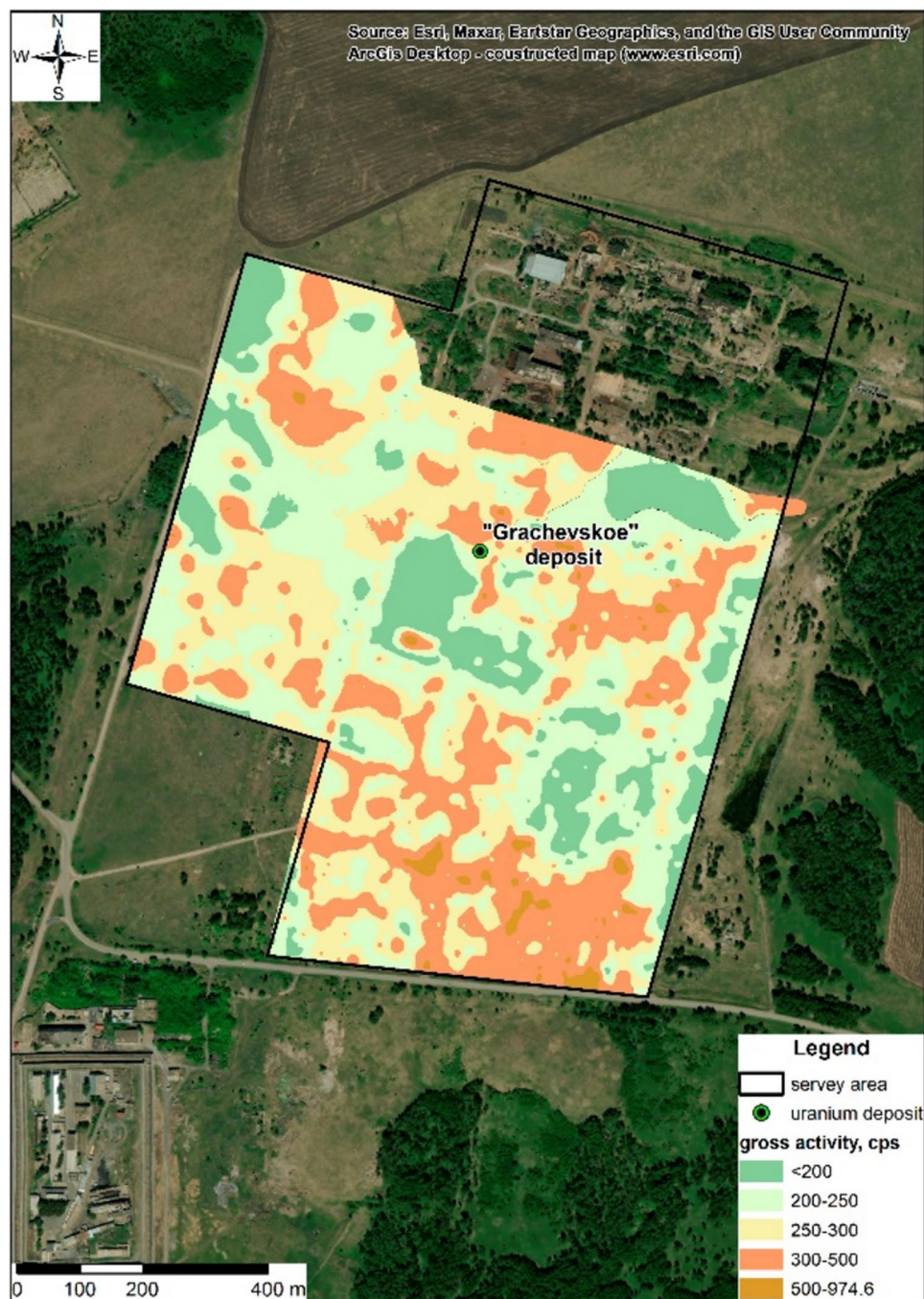


Fig. 3. Results of the pedestrian gamma spectrometry survey at Grachevskoye uranium mining facility (MUMF).

concentration in the topsoil of the MUMF was 587 Bq/kg, and that of Saumalkol was 689 Bq/kg, which is higher than the means for the soils of Kazakhstan. This was most probably caused by the different geochemical type of soil containing more radionuclides. Additionally, the means of ^{210}Pb activity concentration in the topsoil of the research sites were virtually identical, as indicated in the Fig. 5.

Assessment of possible contamination paths of the territory

Assessment of possible contamination of the territory resulted from ore transportation

The results of the pedestrian gamma spectrometry survey along the three routes of the ore transportation section from the Grachevskoye MUMF to the Saumalkol railway station and along the railway are depicted in Fig. 6.

The first route was on the right side of the motor road from Grachevskoye MUMF to the railway station in Saumalkol. Three road intervals with elevated values were observed along this route. The first interval was 1 km long on a road section from the Grachevskoye MUMF, with gross activity values ranging from 200 to



Fig. 4. Results of the pedestrian gamma spectrometry survey at 10 sites in Saumalkol village.

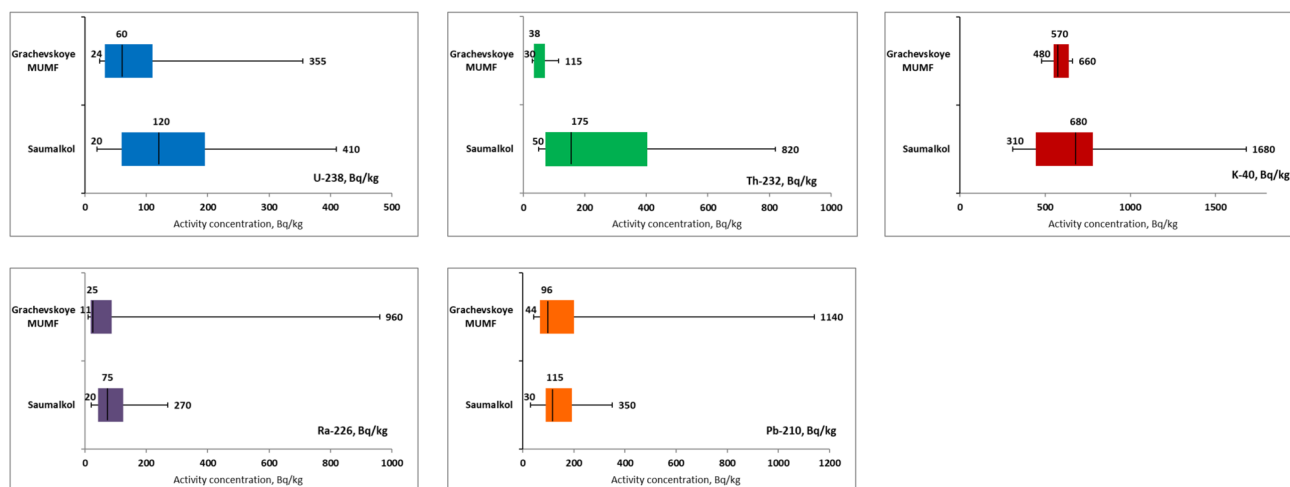


Fig. 5. Activity of ^{238}U , ^{232}Th , ^{226}Ra , ^{40}K , and ^{210}Pb in surface soils.

300 cps. The second interval was on a road section of 4–6 km, with values ranging from 200 to 480 cps. Route sections from 50 to 500 m were marked with maximum cps across the route. The third section starts 6 km from the railway station, with elevated values of 200–300 cps. These intervals of radioactive contamination could have resulted from both the loss of ore during transportation and the utilisation of local soils with a higher radionuclide content during road construction, as identified from the area survey data at the Grachevskoye MUMF site and at sites 1–5 (Fig. 6). According to the data from γ -spectrometric analysis, radionuclide activity concentrations in core samples reached the following values: <0.9 – 20 Bq/kg for ^{137}Cs , ≤ 1.2 Bq/kg for ^{241}Am ,

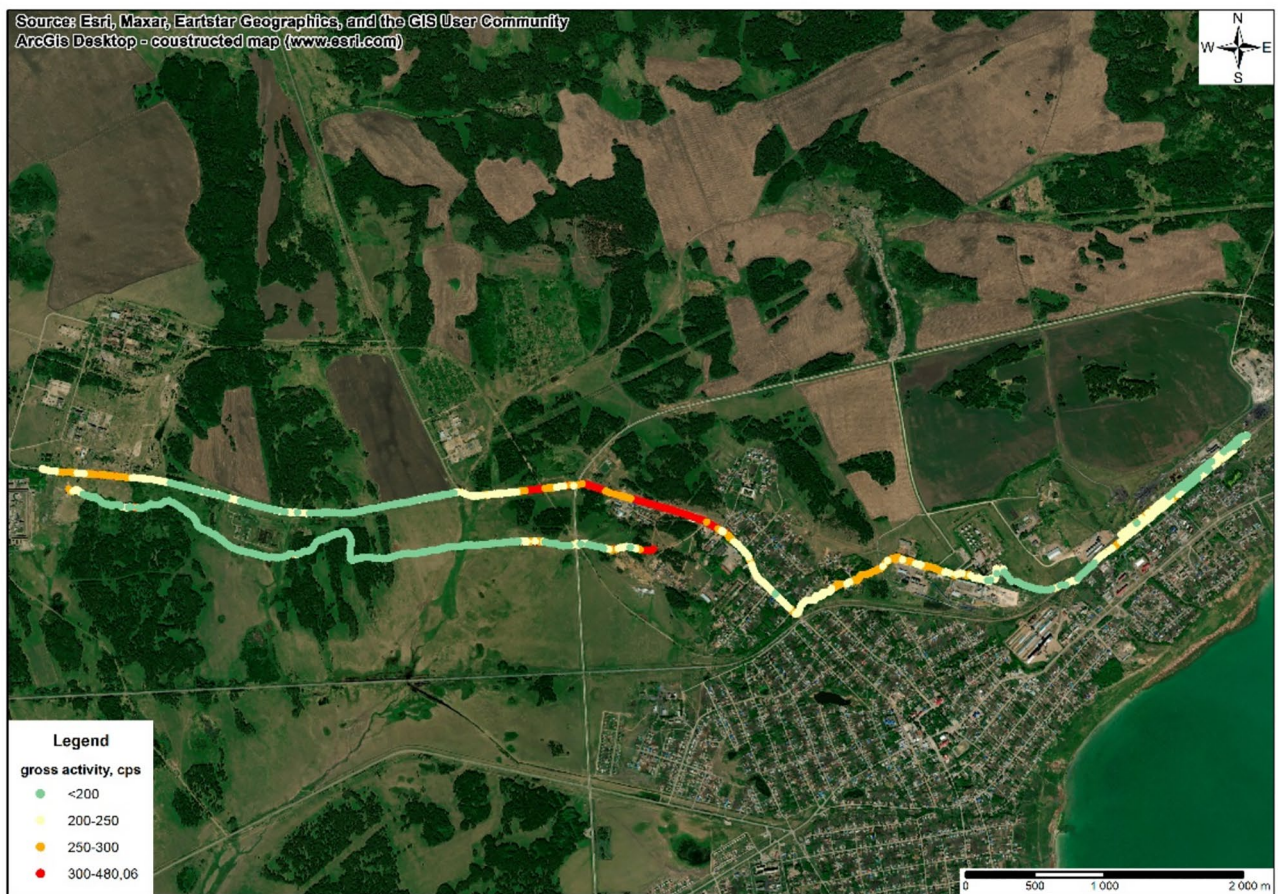


Fig. 6. Result of a route pedestrian gamma spectrometric survey.

670–1,400 Bq/kg for ^{40}K , 93–150 Bq/kg for ^{238}U , 95–160 Bq/kg for ^{210}Pb , 60–200 Bq/kg for ^{232}Th , and 63–97 Bq/kg for ^{226}Ra (Table S1).

The second route was along the first route at a distance of up to 100 m. The purpose of this route was to demonstrate background values. However, intervals with elevated cps values and peak activity concentrations of radionuclides were identified for this route. For example, at the beginning of the route, a section with values up to 300 cps was recorded near Grachevskoye MUMF, and a value of 480 cps was recorded at the end of the route, not far from the open pit. The activity concentrations of radionuclides in the soils were as follows: <5.0 –38 Bq/kg for ^{137}Cs , ≤ 1.0 Bq/kg for ^{241}Am , 510–1,100 Bq/kg for ^{40}K , 255–4,500 Bq/kg for ^{238}U , 310–2,100 Bq/kg for ^{210}Pb , 60–520 Bq/kg for ^{232}Th , and 200–2,200 Bq/kg for ^{226}Ra (Table S1).

The third route was along the railway tracks to identify possible ore losses when loaded into railway cars and further transported. A minor excess in gross activity was observed in this section, which was most likely caused by rock fragments in the ballast of the railway track. The activity concentrations of radionuclides in soil samples along this route reached the following values: <0.9 –1.7 Bq/kg for ^{137}Cs , ≤ 0.2 Bq/kg for ^{241}Am , 370–500 Bq/kg for ^{40}K , 24–170 Bq/kg for ^{238}U , 140–250 Bq/kg for ^{210}Pb , 26–45 Bq/kg for ^{232}Th , and 3.0–26 Bq/kg for ^{226}Ra (Table S1).

Overall, the pedestrian route gamma spectrometry survey indicated no loss of uranium ore material during transportation by motor vehicles and rail. The vicinity of the Grachevskoye MUMF location and the northern part of Saumalkol Village had sites with an elevated radionuclide content on the daylight surface. This feature may be associated with the weathering crust of hard rocks outcropping into the daylight surface containing elevated radionuclide values. Because no fragments of uranium ore were found during the survey of Saumalkol Village, the progression of the radioecological situation around Saumalkol was assumed to be not impacted by any contributors on the surface due to the conservation sites of radioactive waste.

Assessment of the mobility of radionuclides in the soils

The speciation of ^{226}Ra , ^{232}Th , ^{210}Pb , and ^{238}U in the soils of the Grachevskoye MUMF are indicated in Table 1.

The outputs showed that the acid-soluble (mobile) and tightly bound forms mostly contained the radionuclides ^{226}Ra , ^{232}Th , ^{210}Pb , and ^{238}U . In virtually all samples, the radionuclide content in water-soluble and exchangeable forms was less than the detection limit of the methodological hardware used. The most mobile of the studied radionuclides was the isotope ^{210}Pb , and its content in the composition of the acid-soluble (mobile) form was

Point No	Speciation							
	Water-soluble (H ₂ O)		Exchangeable (1 M CH ₃ COONH ₄)		Acid-soluble (mobile) (1 M HCl)		Tightly bound (soil after extracts)	
	Bq/kg	%	Bq/kg	%	Bq/kg	%	Bq/kg	%
²²⁶Ra								
G-1	< 6.5	< 1.4	35.0	7.7	115 ± 25	25.2	300 ± 60	65.7
G-2	< 8.5	< 3.6	< 8.5	< 3.6	< 7.0	< 3.0	210 ± 40	89.7
G-3	< 6.5	< 1.1	< 7.0	< 1.2	220 ± 45	37.7	350 ± 70	60.0
²³²Th								
G-1	< 10.0	< 10.8	< 17.5	< 18.9	25.0 ± 5.0	27.0	40.0 ± 8.0	43.2
G-2	< 15.0	< 14.5	< 14.5	< 14.0	20.0 ± 5.0	19.3	54.0 ± 11	52.2
G-3	< 10.0	< 8.1	< 10.0	< 8.1	50.0 ± 10.0	40.3	55.0 ± 11	43.5
²¹⁰Pb								
G-1	< 10.0	< 1.2	< 20.5	< 2.5	390 ± 80	48.1	390 ± 80	48.1
G-2	< 15.0	< 2.2	< 20.5	< 2.9	340 ± 70	48.9	320 ± 60	46.0
G-3	< 15.0	< 1.1	< 15.5	< 1.2	650 ± 150	48.9	650 ± 130	48.9
²³⁸U								
G-1	< 15.0	< 3.8	< 30.5	< 7.8	65.0 ± 15	16.6	280 ± 60	71.7
G-2	< 20.0	< 5.3	< 20.5	< 5.4	60.0 ± 10	15.8	280 ± 60	73.6
G-3	< 15.0	< 3.9	< 16.5	< 4.3	< 23.5	< 6.1	330 ± 70	85.7

Table 1. Results on the content of various speciation of ²²⁶Ra, ²³²Th, ²¹⁰Pb, and ²³⁸U in the soils of Grachevskoye MUMF.

Sampling location	Sampling point	Uranium content, Bk/m ³	
		U (PM2.5)	U (PM10)
Grachevskoye MUMF	1	$1.9 \times 10^{-4} \pm 3.8 \times 10^{-5}$	$1.37 \times 10^{-4} \pm 2.6 \times 10^{-5}$
Novoukrainka village	2	< d.l	Not measured
Novoukrainka village	3	$1.6 \times 10^{-4} \pm 2.9 \times 10^{-5}$	$1.81 \times 10^{-4} \pm 1.8 \times 10^{-5}$
Saumalkol village	4	$1.9 \times 10^{-4} \pm 3.5 \times 10^{-5}$	$1.27 \times 10^{-4} \pm 7.9 \times 10^{-6}$
Saumalkol village	5	< d.l	Not measured

Table 2. Results on the content of uranium in suspended aerosol particles of the ambient air.

at the same level as that of the tightly bound form. Isotopes ²²⁶Ra, ²³²Th, and ²³⁸U predominated in the tightly bound form, whereas the fraction in the acid-soluble form was smaller but still significant.

Water-soluble and exchangeable forms are represented by reactive, geochemically mobile compounds that can transform when the physicochemical environmental parameters, such as pH, Eh, salinity, and organic matter content, change. The acid-soluble and tightly bonded forms are residual (fragmental) or geochemically inert forms represented by terrigenous particles in which radionuclides are fixed in the crystal lattice of minerals. In summary, the radionuclides of interest are in a slow-mobile form and do not pose any ecological threat to the environmental compartments of deposits and settlements.

Assessment of the transport of radionuclides by the ambient air

The volumetric activities of radionuclides in ambient air are listed in Table 2.

The laboratory analyses showed that uranium concentration in the ambient air of the study area ranged from 1.6×10^{-4} to 3.0×10^{-4} Bq/m³, which is two orders of magnitude lower than the values established for the permissible annual average volumetric activity specified by the health standards¹⁶.

The assessment of the contribution by the contaminated soil to the ambient air of the adjacent areas indicated that radionuclides ²³⁸U, ²²⁶Ra, ²³²Th, and ²¹⁰Pb were distributed in particle-size fractions of soil at the Grachevskoye deposit as follows (Fig. 7). In two samples (G-1 and G-3), a similar trend in the distribution of radionuclides in fractions was defined and marked by a uniform distribution of radionuclides in fractions smaller than 63 µm and further sequential enrichment of the fractions with radionuclides as the fraction sizes increased, with a slight decrease in the coarsest fraction of 10,000 µm in the first sample (G-1). This trend was typical for all four radionuclides, including, ²³²Th, ²²⁸Ra, ²¹⁰Pb, and ²³⁸U. In the third sample (G-2), the distribution patterns of ²¹⁰Pb and ²²⁸Ra differed from each other and from those of ²³⁸U and ²³²Th. Common to all radionuclides was a decrease in the enrichment of fractions with radionuclides from 63 to 1,000 µm, a further enrichment of the 5,000 µm fraction, and a decrease in the 10,000 µm fraction. The highest enrichment of 5,000 µm fractions was common for samples G-1 and G-2. In general, enrichment in the coarse fractions was

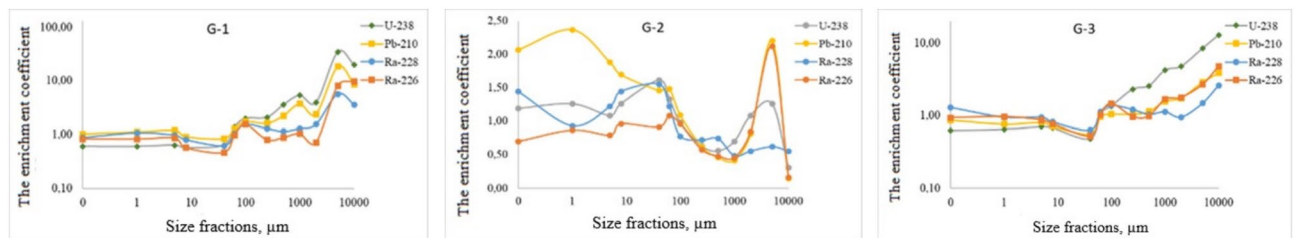


Fig. 7. Distribution of radionuclides in particle-size fractions.

No. of water sampling point	²¹⁰ Po, Bq/kg	Rn ²²² , Bq/m ³	U, µg/L	Gross activity, Bq/L		Stable isotopes	
				Alpha	Beta	¹⁸ O, ‰	² H, ‰
Water from centralised water supply sources							
1	<0,2	24	240 ± 21	8.1 ± 1.6	<0.37	− 16.54	− 117.88
6	1.6 × 10 ^{−2} ± 0.3 × 10 ^{−2}	5	22 ± 2	0.65 ± 0.12	<0.25	− 16.92	− 123.56
7	6.5 × 10 ^{−3} ± 1.0 × 10 ^{−3}	*	65 ± 5	1.45 ± 0.30	<0.26	− 16.19	− 118.72
Irrigation wells							
2	8.5 × 10 ^{−3} ± 1.9 × 10 ^{−3}	8	19 ± 2	0.69 ± 0.14	0.77 ± 0.14	− 12.95	− 94.02
3	2.0 × 10 ^{−2} ± 0.4 × 10 ^{−2}	4	20 ± 2	0.82 ± 0.16	0.38 ± 0.08	− 14.49	− 101.50
4	6.5 × 10 ^{−3} ± 1.8 × 10 ^{−3}	13	11 ± 1	0.35 ± 0.06	0.53 ± 0.08	− 14.31	− 104.12
Surface waters							
5	1.6 × 10 ^{−2} ± 1.1 × 10 ^{−2}	*	19 ± 2	0.53 ± 0.07	0.47 ± 0.08	− 7.97	− 73.61
MPC/IL _{pub}	0.11 Bq/kg	60 Bq/m ³	30 µg/L	0.2	1.0	*	*

Table 3. Research findings on the radiation characteristics of water samples.

regular because of the contribution of the parent rock. The difference in the distribution of radionuclides in the particle size fractions of the third sample (G-2) may have been due to sample disturbance during remediation.

Notably, the World Health Organization (WHO) has recommended the following concentrations of suspended particles for the ambient air: for PM_{2.5}, the average annual and daily concentrations are 10 μg/m³ and 25 μg/m³, respectively, and its excess should not last longer than 3 days per year. The average annual and daily concentrations for PM₁₀ are 20 μg/m³ and 50 μg/m³, respectively. Research on the concentrations of suspended aerosol particles under natural and artificial dusting conditions and during agricultural activities were conducted in the STS territory¹⁷. The outputs showed that under natural conditions, the average annual dust content in the air is 12 μg/m³. Peak concentrations were detected during agricultural work and artificial dusting, reaching 50 and 180 μg/m³, respectively.

During the period of air research near the Grachevskoye MUMF, no sharp climatic changes (dust storms, precipitation) were observed, the wind speed was up to 5 m/s, and the relative humidity of the ambient air varied from 50 to 60%. The temperature of the ambient air varied from 15 to 23 °C. Under these conditions, the dust content in the air can be accepted as 12 μg/m³, for which the volumetric activity of uranium in the air was 3.0×10^{-4} Bq/m³. When carrying out construction operations on the premises of the Grachevskoye MUMF, one may assume that the concentration of dust in the air will increase by a factor of 10; consequently, the volumetric activity in the air will significantly increase. It is also worth accounting for the points discovered during the route pedestrian survey, which contained as much as 4,500 Bq/kg uranium.

Assessment of the contaminated water pathway

The laboratory determinations of the radiation characteristics of the water samples from the water bodies of the Saumalkol settlement are listed in the Table 3.

According to the analysis of data on the gross alpha and beta activities, the beta activity (1.0 Bq/L) was not exceeded. However, excess values of 40, 3, and 7 maximum permissible concentrations (MPC) were observed in from borehole 1, 6, and 7, respectively, from centralised water supply sources for the alpha activity (0.2 Bq/L). The irrigation wells and surface waters showed no excess of beta activity (1.0 Bq/L). However, an excess at the level from 1 to 4 MPC was noted for the alpha activity (0.2 Bq/L). Excess uranium contents were found in drinking water: 8 MPC in borehole 1, 1 MPC for U in the water from borehole 6, and 4 MPC for U in borehole 7. No excess intervention level (IL_{pub}) was found in the water for ^{210}Po or ^{222}Rn .

A comparative analysis of data on the component composition was conducted for the drinking water of the Saumalkol settlement over a 20-year period¹⁸ after the mothballing of mine workings to assess the consequences of mining operations at the Grachevskoye deposit. According to the hydrogeological conditions, the groundwater in the study area moved from the location of the Grachevskoye MUMF mine workings to the territory of Saumalkol. The concentration of uranium in the water of the village's intake wells suggested that the input of contaminated groundwater from the flooded mine workings of the uranium mine into the village's groundwater

is possible. The histograms (Fig. 8) depict comparative data on the content of certain elements in the Saumalkol water bodies from 2005 to 2024. Figures A, B, and C present data on the water intake boreholes, where water intake is carried out from fractured waters confined to the zone of exogenous fracturing of hard rocks. Figures D and E show data from wells where water intake was carried out from pore waters common in loose sand and gravelly rocks. Figure F shows surface-water data from ponds. The origin of the ground and surface waters was confirmed by data on the content of stable isotopes in the water. For example, the levels were the lightest (from -117.88 to -123.56% for ^2H and from -16.19 to -16.92% for ^{18}O) in boreholes 1, 6, and 7. Stable isotopes were significantly higher (up to -94.02% for ^2H and up to -12.95% for ^{18}O) in the waters of wells 2 and 3. The highest levels occurred in the surface water body (point 5), where the content of stable isotopes in the water were as high as -73.61% for ^2H and -7.97% for ^{18}O .

Notably, ground water generally flows along interconnected streamlines differing in direction, speed, flow rate and component composition. Hydrogeological conditions are determined by the features of the geological structure of the territory, which are marked by the filtration properties of rocks and the presence of tectonic faults. In our case, three different streamlines occurred among the three water intake boreholes 1, 6, and 7, based on the component composition (Fig. 8). The streamline of borehole 1 was directly linked to the flooded mine workings of the Grachevskoye MUMF. This can be clearly seen from all the chemical elements noted, especially the uranium content, which increased eightfold over 20 years. The uranium content decreased markedly in the remaining boreholes and wells, whereas it remained at the same level in the surface water body. The increased uranium content in borehole 1 was accompanied by high gross alpha activity, even though the concentrations of radon and ^{210}Po were well below the guideline values.

Thus, groundwater contamination is one of the major contributors to uranium mining in the Grachevskoye deposit and to the radioecological situation around Saumalkol.

Radiological hazard assessment

The values of the activity concentrations of natural radionuclides in the soil (Table S1) were used and such indicators to assess the potential radiation risk to the public at sites of interest on the premises of the Grachevskoye MUMF and Saumalkol Village because the activity in the radium equivalent (R_{eq}), absorbed dose rate (D), internal hazard index (H_{in}), external hazard index (H_{ex}), and annual effective dose (E_{ef}) were estimated (Table 4).

Based on the average content of natural radionuclides in the soil, the equivalent radium activity (R_{eq}) was 316.3 Bq/kg in the Grachevskoye MUMF soil and 657.1 Bq/kg in the Saumalkol village. This was higher than the established value of 370 Bq/kg .

External (H_{ex}) and internal (H_{in}) hazard indices were also calculated. The H_{ex} and H_{in} values calculated with the average values of radionuclides in the soil were 0.6 and 0.9 in the Grachevskoye MUMF and 1.9 and 2.4 in the Saumalkol village, which exceeded the established value of 1 ¹⁹.

The quantity that considers human radiation risk is the total absorbed dose rate in air. The absorbed dose rate at the average levels of ^{238}U , ^{232}Th , and ^{40}K in the soil was 107.3 nGy/h and 327.0 nGy/h for the Grachevskoye MUMF and Saumalkol Village, respectively, which is higher than the global mean of 59 nGy/h ^{20,21}. The calculated potential annual effective dose for the public living at sites of interest with the mean values of the radionuclide activity concentrations in the soil is 0.7 and 2.0 mSv/year , respectively.

The findings showed that the radiological hazard parameters, namely the radium equivalent activity (R_{eq}), absorbed dose rate (D), and radiological hazard indices (H_{ex} and H_{in}) at the maxima of the activity concentrations of natural radionuclides (^{238}U , ^{232}Th , and ^{40}K), exceeded the global values²⁰ at the sites of interest.

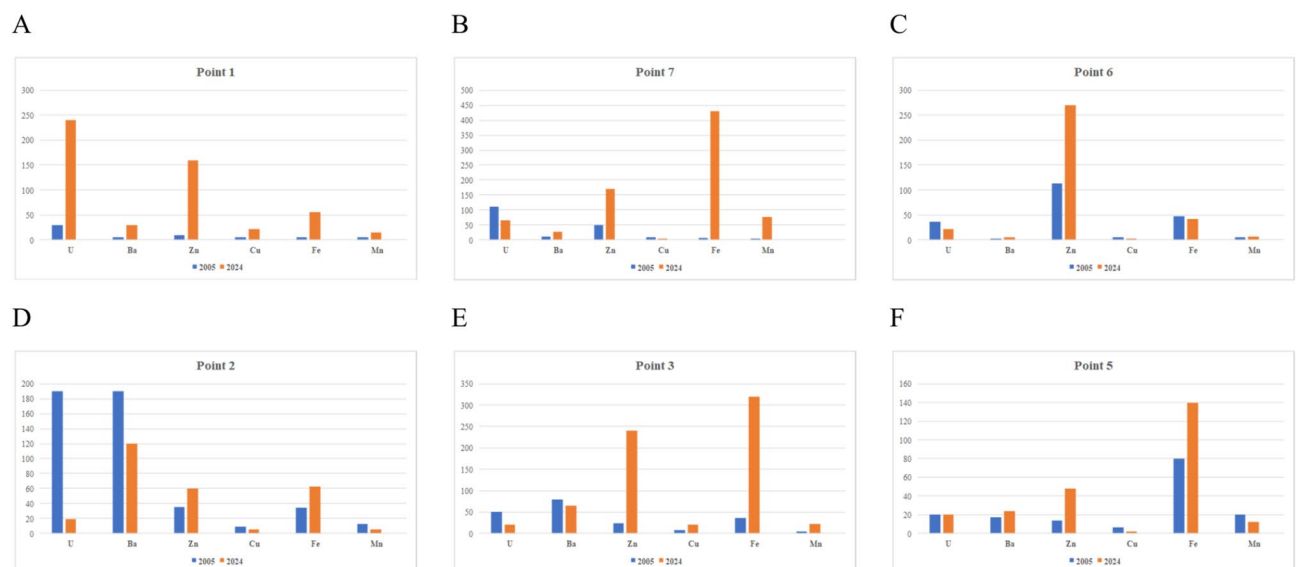


Fig. 8. Comparative data on the elemental composition of water.

Values	Ra _{eq}	H _{ex}	H _{in}	D _{out} (nGy/h)	E _{out}	E _{in}	E _{ext}
Grachevskoye MUMF							
1	145.7	0.5	0.6	79.5	0.1	0.4	0.5
2	119.9	0.3	0.4	63.5	0.1	0.3	0.4
3	116.7	0.3	0.4	63.1	0.1	0.3	0.4
4	115.3	0.3	0.4	58.2	0.1	0.3	0.4
5	126.5	0.6	0.8	96.7	0.1	0.5	0.6
6	113.4	0.4	0.4	65.2	0.1	0.3	0.4
7	120.2	0.4	0.5	66.3	0.1	0.3	0.4
8	118.2	0.4	0.6	72.9	0.1	0.4	0.4
9	229.5	0.7	1.0	117.0	0.1	0.6	0.7
10	104.0	0.3	0.4	60.6	0.1	0.3	0.4
11	1114.0	1.2	1.9	195.0	0.2	1.0	1.2
12	586.1	1.2	2.0	204.9	0.3	1.0	1.3
13	1103.0	1.5	2.5	252.0	0.3	1.2	1.6
Min	104.0	0.3	0.4	58.2	0.1	0.3	0.4
Mean	316.3	0.6	0.9	107.3	0.1	0.5	0.7
Max	1114.0	1.5	2.5	252.0	0.3	1.2	1.6
Saumalkol Village							
24	1458.1	4.2	5.0	717.4	0.9	3.5	4.4
25	125.4	0.4	0.4	64.8	0.1	0.3	0.4
26	340.4	1.0	1.2	173.2	0.2	0.9	1.1
27	800.7	2.3	2.9	397.0	0.5	2.0	2.4
28	146.8	0.4	0.4	64.9	0.1	0.3	0.4
29	569.7	1.7	2.1	285.5	0.4	1.4	1.8
30	600.3	1.7	2.1	292.6	0.4	1.4	1.8
31	1024.9	3.0	3.7	507.8	0.6	2.5	3.1
32	1386.3	4.1	5.2	698.8	0.9	3.4	4.3
33	1230.2	3.5	4.3	603.3	0.7	3.0	3.7
34	186.0	0.5	0.7	91.9	0.1	0.5	0.6
35	337.6	1.0	1.4	180.2	0.2	0.9	1.1
36	336.5	1.0	1.3	174.0	0.2	0.9	1.1
37	410.2	1.1	1.4	196.9	0.2	1.0	1.2
38	516.5	1.5	1.8	253.9	0.3	1.3	1.6
39	1020.5	2.9	3.5	502.2	0.6	2.5	3.1
40	493.3	1.5	1.8	252.2	0.3	1.2	1.6
41	716.8	2.1	2.6	367.9	0.5	1.8	2.3
42	860.0	2.5	3.1	435.7	0.5	2.1	2.7
43	148.9	0.4	0.5	75.3	0.1	0.4	0.5
44	170.1	0.5	0.7	88.7	0.1	0.4	0.5
45	281.3	0.8	0.9	131.5	0.2	0.6	0.8
46	134.6	0.4	0.5	68.7	0.1	0.3	0.4
47	213.8	0.6	0.8	105.3	0.1	0.5	0.6
48	150.0	0.6	0.9	100.3	0.1	0.5	0.6
49	139.2	0.4	0.5	67.0	0.1	0.3	0.4
Min	115.4	0.4	0.4	64.8	0.1	0.3	0.4
Mean	1571.9	1.9	2.4	327.0	0.4	1.6	2.0
Max	530.3	4.2	5.2	717.4	0.9	3.5	4.4
World average ^{19,21}	370	1	1	59	0.07	0.05	1

Table 4. Radiological parameters of the Grachevskoye MUMF and Saumalkol village areas.

Conclusions

According to the results of a radiation survey near Grachevskoye MUMF and in the northern part of Saumalkol village, spots of elevated radionuclide content were discovered on the daylight surface. The hotspots of radioactive contamination in the Grachevskoye MUMF were associated with mining and uranium ore conservation. Spots with an elevated radionuclide content in the northern part of Saumalkol village near the crushed rock open pit were attributed to the exposure of the weathering crust of hard rocks at the surface. The presence of motor

road sections with cps is attributed to the utilisation of local soils with elevated radionuclide values during road construction. Notably, no ore fragments containing high radionuclide activities were found during the inspection of the motor road, railway, and vicinity of the open pit. This suggested that no loss of uranium ore material occurred during transportation by motor vehicles and rail, and the spots with high levels of radionuclides outside the Grachevskoye MUMF were not linked to the radioactive waste disposal site.

Natural radionuclides in Grachevskoye MUMF soils are characterised by low mobility. The main radionuclide content is in a tightly bound form, which makes it impossible for it to migrate with surface waters from the mine premises towards the settlement.

The process of radionuclide transport by ambient air under normal climatic conditions did not affect the progression of the radioecological situation around the settlement because the volumetric activity of radionuclides in the air is two orders of magnitude lower than the established value of the permissible average annual volumetric activity established by health standards. However, when performing agricultural or construction work at spots with high uranium content in the soil, the dust content in the air may increase by more than a factor of 10, which can lead to an excess of the guideline values of the radionuclide volumetric activity in the air.

Significantly increased concentrations of ^{238}U in the drinking water of the village were associated with the input of contaminated groundwater from the flooded mine workings of the Grachevskoye MUMF. In summary, groundwater contamination is one of the main contributors to uranium mining in the Grachevskoye deposit in the radioecological environment around the Saumalkol settlement.

Radiological hazards were estimated using the means and maxima of ^{226}Ra , ^{238}U , ^{232}Th , and ^{40}K activity concentrations in the soil of the sites of interest that exceeded the recommended values. The results showed that the topsoil at the sites of interest containing elevated concentrations of natural radionuclides may pose a radiological hazard to the public.

Despite its valuable insights, this study has several limitations that should be addressed. A major limitation of this study was the limited range of samples, which may not fully represent the broader environmental variability in the region. Further research should broaden the scope to include a wider range of samples (e.g., plant and food samples) to provide a more comprehensive view of environmental contamination.

Materials and methods

Fieldwork

The fieldwork involved a preliminary radiological survey, a pedestrian gamma spectrometric survey, and environmental sampling.

A preliminary radiological survey was conducted during the summer and fall. Field radiological measurements were taken with instruments Inspector 1000 and RadEye B20 in order to reveal sites of elevated gamma radiation. A detector was moved above the daylight surface of the soil in search mode during the survey. The equivalent dose rate (EDR) and β -emission were measured using survey meters MKS-AT 1117 M and MKS-AT 6130 at fixed points with elevated values of gamma radiation. The detector was held 5 cm and 1 m above the ground when measuring the EDR, and a mean of five measurements was taken.

Based on the preliminary survey results, a pedestrian gamma spectrometric survey was conducted, in which the EDR values were elevated. This evaluation technique is based on the continuous accumulation of gamma spectra as the operator moves along a specified route across the study area at a certain speed²². The measurements were performed using an equipment system consisting of a lanthanum bromide crystal-based ($\text{LaBr}_3(\text{Ce})$) scintillation gamma spectrometer and a portable PC equipped with a USB-interfaced GPS module. The measurements were processed using specialised software on a PC, which facilitated the detection and accumulation of gamma-ray spectra. The scintillation gamma spectrometer was held at a height of 50 cm relative to the measured surface. The gamma spectra were automatically recorded every 10 s and saved on a portable PC. Next, the spectra were processed and maps of the radioactively contaminated ground were plotted using the processed results. Using this technique, an area survey of the Grachevskoye MUMF was conducted (0.73 km²). This included 10 individual sites within the Saumalkol settlement, as well as three routes: route 1 passed along the right motor road verge leading from the Grachevskoye MUMF to the railway station of Saumalkol, route 2 passed along the motor road from the Grachevskoye MUMF to the Saumalkol settlement on the right side of the road with a distance of up to 100 m between the routes, and route 3 passed along the railway route of 500 m each both ways from the railway station on either side of the railway track. A route pedestrian gamma spectrometric survey was conducted to assess possible losses of ore material during transportation on motor roads and railways.

Within the scope of the radiological survey, 49 soil samples were collected: 13 from the premises of the Grachevskoye MUMF, 10 along the routes, and 26 from Saumalkol settlement (Fig. S1). The sampling depth was 0–5 cm over an area of 100 cm²³. Eight ambient air samples were collected from Grachevskoye MUMF, Saumalkol, and Novoukrainka, and seven water samples²⁴ were collected from Saumalkol Village. Six ground water samples were collected from three boreholes (p.1, p.6, and p.7) that were used as potable water sources and three wells (p.2, p.3, and p.4) that were used as process water sources. One sample was collected from the surface water of a water body (p.5) (Fig. S2).

Air was sampled in the summer using an MVS-6 sampler at a flow rate of 2.3 m³/h. The air sampler's inlet duct is fitted with calibrated nozzles for sampling airborne aerosols by fractions ranging from 0.01 to 10.0 μm (PM10.0). A quartz filter QMA is a filter element with a capture efficiency of 99.5%. Airborne aerosols were sampled from a height of 1.5 m for 2 h. The airflow rate during sampling was 2.3 m³/h. When assessing ambient air contamination with radioactive aerosols, one has to consider the human hazard level of particular fractions of suspended airborne particles. It is common international practice²⁵ to determine concentrations of suspended aerosol particles <2.5 μm (PM2.5) and <10 μm (PM10) while assessing the air quality because suspended

1–10 μm in size are inhaled. Therefore, high levels can pose a great hazard to man. In this regard, PM_{2.5} and PM₁₀-size suspended aerosol particles were collected at each point.

Laboratory work

Determination of radionuclide speciation

A study on radionuclide speciation and the parameters of mobility and bioavailability of radionuclides in soils was conducted to assess the possible migration of radionuclides in surface waters. Radionuclide speciation in three soil samples (G-1, G-2, and G-3) collected from the premises of the Grachevskoye MUMF was determined using sequential leaching using the following extractants: distilled water (water-soluble forms), 1 M $\text{CH}_3\text{COONH}_4$ (pH 6.8) (exchangeable forms), 0.1 N NaOH (organic form), and 1 N HCl (mobile or acid-soluble)²⁶. The soil: leaching solution ratio was 1:5 (g/mL). Each soil sample weighed 150.0 g each. The soil suspension was shaken for 1 h on a rotator and filtered through a blue ribbon filter the following day. The average extraction time was 16 h. Extraction was performed at room temperature without heating. The water extracts were acidified to pH 1–2 for stabilisation. The radionuclide residue that could not be extracted from the soil was collected in a tightly bound form. The gamma-emitting radionuclide content was determined in the soil extract and soil samples after leaching.

Particle-size analysis

A particle size analysis of the three most contaminated soil samples (G-1, G-2, and G-3) from the premises of the Grachevskoye MUMF was conducted to assess the ability of contaminated soil to rise in dust and be further transported from the atmospheric air to adjacent areas. The particle size fractionation of the soil was accomplished through sieving and sedimentation. Soil fractions of 40–1000 μm were isolated using a 'wet sieving' method²⁷. The dust fractions (<40 μm) were separated using sedimentation. The pipette method, which consists of separating particles by grain size when they fall in a stationary liquid, was used as a basis for separating the dust fractions of the soil through sedimentation to determine the particle size composition of the soil. In total, 13 fractions ranging from 10,000 to <1 μm were isolated from each test soil sample. Each isolated fraction was air-dried, weighed, and analysed for radionuclides. In addition, the possibility that atmospheric air contamination might increase due to radioactive aerosols was theoretically assessed based on previous work performed at the STS^{17,28}. The enrichment coefficient, a parameter indicating the enrichment or depletion rate of particle size fractions relative to the average radionuclide content in the soil, was applied to analyse the findings. The enrichment coefficient was determined from the ratio of a radionuclide activity concentration in a particle-size fraction (A_{ac}^{fr}) to that of the same one in a respective original sample (A_{ac}^{soil}) (1).

$$K_o = \frac{A_{ac}^{fr}}{A_{ac}^{soil}} \quad (1)$$

Radionuclide analysis

The concentrations of ^{137}Cs , ^{241}Am , ^{40}K , ^{238}U , ^{210}Pb , ^{232}Th , and ^{226}Ra were determined using an ORTEC gamma-ray spectrometer fitted with a solid-state ultrapure germanium detector GER-FX7025P4 in accordance with the standard procedure²⁹. ^{210}Po content in water was determined using an Alpha Analyst7200 Alpha spectrometer (Canberra) consisting of 12 vacuum cells with PIPS alpha-ray detectors and GENIE 2000 software. ^{209}Po tracer was used to control the chemical yield of ^{210}Po during analysis. The total alpha–beta activity of the water samples was determined by evaporation of a dry residue, from which a counting sample was prepared using a low-background alpha–beta radiometer UMF-2000³⁰. Alpharad plus-AR was used to determine the concentration of radon in the water.

Elemental analysis

Inductively coupled plasma-atomic emission spectrometry (ICP-AES) with an Elan 9000 quadrupole mass spectrometer (Perkin Elmer SCIEX) and an iCAP 6300 Duo ICP-AES (Thermo Scientific) was used to determine the elemental composition of water³¹.

Measurement of stable isotopes

The ratios of stable isotopes $^2\text{H}/^1\text{H}$ and $^{18}\text{O}/^{16}\text{O}$ in the analyte and standard were measured with a highly sensitive laser spectrometer LGR 912-0008^{32–34}. Water samples calibrated against the international standard VSMOW (IAEA) were used as internal standards.

Assessment of the radiological hazard

The absorbed dose rate was calculated using a coefficient to convert the activity concentrations of A_U , A_{Th} , and A_K to the absorbed dose rate (D) at a height of 1 m above the underlying surface according to the formula used globally^{19,20}:

$$D = 0.462A_U + 0.604A_{Th} + 0.0417A_K, \text{ nGy/h} \quad (2)$$

where A_U , A_{Th} , and A_K are the activity concentrations (Bq/kg) of the isotopes ^{238}U , ^{232}Th , and ^{40}K , respectively.

The annual effective dose (E_{ext}) for adult residents was determined using the following formula^{19–35}:

$$E_{ext} = E_{out} + E_{in}, \text{ mSv/year} \quad (3)$$

$$E_{out} = D \times 0.7 \times 0.2 \times 8800, \text{ mSv/year} \quad (4)$$

$$E_{in} = D \times 0.7 \times 0.8 \times 8800, \text{ mSv/year} \quad (5)$$

where: E_{out} , E_{in} : annual effective external dose outside and inside, mSv/year, respectively; D : absorbed dose rate, nGy/h; 8800: exposure time throughout the year, h; 0.7: transfer factor from the value of the absorbed dose in the air 1 high to that of the effective dose (Sv/Gy); 0.8 and 0.2: time fractions spent by people indoors or outdoors, respectively.

The equivalent activity of radium (Ra_{eq}) was computed using the following formula²⁰ to assess the radiological hazards:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K, \text{ Bq/kg} \quad (6)$$

where A_{Ra} , A_{Th} , and A_K were the activity concentrations (Bq/kg) of ^{226}Ra , ^{232}Th , and ^{40}K , respectively.

The parameters of the external (H_{ex}) (Bq/kg) and internal hazard (H_{in}) (Bq/kg) were used to assess the potential severity of exposure to radiation and subsequent effects related to this risk. The value of this parameter was assumed to be less than that of unity; therefore, the radiological hazard was minor. The indices of external and internal hazards were calculated using the following formula²⁰:

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}, \quad (7)$$

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}, \quad (8)$$

where A_U , A_{Th} , and A_K were the activity concentrations (Bq/kg) of ^{238}U , ^{232}Th , and ^{40}K , respectively.

Data availability

Data is provided within the manuscript or supplementary information files.

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Author contributions

Conceptualization, S.S., N.L.; data curation, S.S., P.K., A.T., A.K.; funding acquisition, S.S.; methodology, S.S., P.K., N.L., D.T.; project administration, S.S.; investigation, S.S., P.K., A.T.; resources, P.K., A.K., D.T., Zh.T.; visualization, P.K., A.T., A.K.; software, P.K., A.T.; formal analysis, A.K., D.T., Zh.T.; supervision, Zh.T., A.A.; validation. writing – original draft, S.S., A.T., A.K.; writing – review & editing, N. L., A.A.

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Declarations

Competing interests

The authors declare no competing interests.

Additional information

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