

## Concentrations of Natural Radionuclides (40K, 226Ra, 232Th) at the Potash Salts Deposit

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### ABSTRACT

Specific activity of natural radionuclides 226Ra, 232Th and 40K was measured by gamma spectrometry in samples of soil and bottom sediments of rivers on the territory of potassium salt deposit development in order to collect information on radionuclide distribution. According to the results of examinations, the range of 226Ra, 232Th and 40K activity values in soils and bottom sediments of the territory of influence was <2.17–19.7 Bq/kg, <2.5–20.5 Bq/kg, 123–500 Bq/kg. Migration of the studied radionuclides is limited to 2 km zone. The results obtained provide useful information on the levels of naturally occurring radionuclides in the zone of influence of mining objects and are of interest for a comparative evaluation of the impact of salt mining on the radiation environment of the territory.

**Keywords:** soils; bottom sediments; radium; thorium; potassium;  $\gamma$ -spectrometer; Verkhnekamskoe potash salt deposit.

### INTRODUCTION

The natural radiation background of the territory is conditioned by cosmic radiation as well as by the presence of radioactive elements in the Earth's crust, water bodies and atmosphere. Among the radioactive isotopes of natural origin the most widespread and important from the ecological point of view are 238U, 232Th, 226Ra and 40K [Paschoa et al., 2010]. The last decades of civilization development have led to radical changes in ecological conditions, in particular, to intensive radioactive contamination of the main components of the natural environment (soils, natural waters, rocks, bottom sediments) [Mirzoyeva et al., 2015]. The radiation background of any territory consists of a natural background and radioactive contamination caused by anthropogenic impact. Often high levels of radiation background are associated with natural radioactivity of rocks, as well as with geochemical features of behavior of radionuclides themselves. Therefore, when characterizing the radioecological state of the territory it

is necessary to assess not only anthropogenic impact, but also the natural component and compare the data obtained with the regional background [Markelov, 2008].

Gamma spectrometric measurements of the activity of natural radionuclides in various objects of the environment are now actively used to assess the ecological state of territories. Such studies are actively conducted for soils, bottom sediments and beach sands [Abusini et al., 2008; Caridi et al., 2016; Singh et al., 2005; Yang et al., 2005; Monged et al., 2020; Dragović et al., 2012; Forkapic et al., 2017; Tahir et al., 2005; Guseva et al., 2019; Alam et al., 1999], various wastes [Beretka et al., 1985; El Afifi et al., 2009], especially for coal industry [Menshikova et al., 2019; Kozłowska et al., 2012; Sidorova et al., 2015], food salt [Caridi et al., 2019; Tahir et al., 2008; El-Bahi et al., 2003; Hançerlioğullari et al., 2020]

In the process of migration, natural radionuclides enter water bodies, accumulate in soils, grounds and bottom sediments; their high concentrations can pose danger to humans. Possible

sources of contamination of environmental objects with natural radionuclides are natural formations – rocks with increased content of natural radionuclides [Joshua et al., 2009; Khater et al., 2013; Srinivasa et al., 2019] and industrial enterprises producing and enriching certain types of minerals (coal, oil, gas, phosphates, clay, metaliferous and non-metallic ores) [Extent ..., 2003; Guerrero et al., 2020]. The attention of researchers to the radiation situation in salt deposits is associated with the promising development of speleotherapy [Calin et al., 2012]. In particular, the use of materials based on natural potassium salt for interior decoration is a known tool to create a therapeutic effect due to air ionization [Chernyi, 2019].

The study area is one of the most developed areas of the Verkhnekamskoye potash salt deposit (Ural region, Russia). Underground mining of sylvinite (raw material for the production of potash fertilizers), carnallite (production of artificial carnallite for the magnesium industry) and brines (raw material for the production of soda) is conducted here. The geological reserves of the deposit are huge and are estimated at 96.4 billion tons for carnallite rock, 113.2 billion tons for sylvinite and 4.650 billion tons for rock salt.

The purpose of the research is to estimate the activity of natural radionuclides  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in soils and bottom sediments within the territory of potassium salt deposit development, their spatial distribution and possible anthropogenic sources. In the investigated group of natural radionuclides,  $^{40}\text{K}$  is of the greatest interest for the territory under consideration. Natural potassium is a mixture of three isotopes  $^{39}\text{K}$ ,  $^{40}\text{K}$ ,  $^{41}\text{K}$ , while only  $^{40}\text{K}$  is radioactive. It is a natural radionuclide of non-serial origin that is present everywhere on the Earth. The results presented in the paper provide useful information on the content of natural radionuclides in the influence zone of mining objects and are of interest for comparative evaluation of the impact of salt mining on the formation of radiation situation of territories.

## METHODS

Soil, bottom sediments and core samples were taken from an exploration well within the study area of the potassium deposit. Soil sampling

points were relatively evenly dispersed throughout the territory adjacent to the mining operation. A total of 44 soil samples were taken by the dug-in method from the top soil layer (0–0.4 m interval). Bottom sediments (6 samples of fine-grained fraction) were taken from watercourses within the territory adjacent to the enterprise.

Radiation examination of the core was carried out on the exploration well in the depth interval of 3.5–349.0 m. Samples were taken from the minable seam and characteristic lithological types of rocks of the overlying strata. A total of 15 core samples were examined. To compare the results obtained from the core samples, the crushed rock in the underground space of the mine was examined. A total of 4 such samples were examined.

Specific activity of natural radionuclides ( $^{40}\text{K}$ ,  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ) was studied for all samples under laboratory conditions. Gamma-spectrometric measurements of samples were done on scintillation gamma-spectrometer MKS-01A “Multirad” (Russia). Gamma-spectra were processed using the programs included in the Progress package. Errors of specific activity measurements were  $\pm(20\text{--}30)\%$  of the measured value.

For the energy calibration of the spectrometer, a reference source based on  $^{137}\text{Cs}$  and  $^{40}\text{K}$  was used. The source is a 1-liter Marinelli vessel filled with KCl, closed with a lid, in the base of which there is a cylindrical recess, where the  $^{137}\text{Cs}$  source is fixed (a disc 29 mm in diameter, in the center of which there is an active spot, sealed with two polyamide films 50  $\mu\text{m}$  thick each). The source activity is 1500 Bq, and the error (at 0.95 confidence probability) is 20%. The spectrum from the gamma spectrometer’s calibration source shows maximums of  $^{137}\text{Cs}$  (energy 662 keV) and  $^{40}\text{K}$  (energy 1461 keV) total absorption peaks, count rate in a certain energy range (usually 620–700 keV). In order to control own noises of gamma-spectrometric path and take into account the impact of external gamma radiation on the hardware radiation spectrum of the investigated sample, after calibration the background measurement was made (during 1800 sec). In the investigated samples of soil and bottom sediments their organic part was removed by calcination before measurement, the samples were mixed and placed in a Marinelli vessel up to the mark of 1 L.

Rationing of exposure to natural radionuclides in accordance with the requirements of

radiation safety standards (Radiation Safety Standards, Russia) was carried out according to the calculated value of specific effective activity  $A_{\text{eff}}$ , which characterizes the total specific activity of natural radionuclides in the studied material, determined taking into account their biological impact on the human body (Bq/kg). The formula for its calculation is as follows:

$$A_{\text{eff}} = A_{\text{Ra}} + 1,31A_{\text{Th}} + 0,085A_{\text{K}}, \quad (1)$$

where:  $A_{\text{Ra}}$  and  $A_{\text{Th}}$  are the specific activities of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  (Bq/kg), which are in equilibrium with the remaining members of the uranium and thorium series,

$A_{\text{K}}$  is the specific activity of  $^{40}\text{K}$  (Bq/kg).

## RESULTS AND DISCUSSION

Soil cover, being at the intersection of migration ways of chemical elements, including radioactive elements, is the main component of the natural environment, carrying the cumulative effect of long-term anthropogenic impact [Grebenshchikova et al., 2017]. In the upper layer of soils and grounds (up to 5 cm), 95–98% of anthropogenic activity is concentrated. The natural activity changes insignificantly with depth, which is why the radiation characteristics of this particular layer are the main in the analysis of soils and grounds.

According to the results of examinations of soils and bottom sediments, the maximum values of specific effective activity of the studied natural radionuclides (value range 62–88 Bq/kg) are observed in the soils of the sites in the immediate vicinity of the operating salt mine administration and to the north-east of it (Table 1, Table 2, Fig. 1). The obtained outlines of specific effective activity distribution of the investigated natural radionuclides correspond to the outlines of  $^{40}\text{K}$  activity distribution (Fig. 2). The formed zone of higher concentrations of natural radionuclides is conditioned by the wind regime of the territory of research, where a high frequency of southern direction winds is observed during a year. Taking into account the prevailing wind direction, there is a significant removal of substances with atmospheric flows in the northern and northeastern directions.

Developed potassium ores are the source of  $^{40}\text{K}$  entry into the environment. Radiation study of the exploration well core (Table 3) demonstrates high values of  $^{40}\text{K}$  activity in productive

potassium formations which is caused by their mineral composition. For the considered deposit the highest values are typical for mottled sylvinites (up to 4967 Bq/kg), which is significantly higher than the  $^{40}\text{K}$  activity for generalized data on sedimentary rocks and continental crust. All examined samples of potassium salts do not exceed the permissible value of 740 Bq/kg by the rated specific effective activity (Radiation Safety Standards, Russia).

The mine site administration monitors atmospheric air at a number of locations on the border of the sanitary protection zone of industrial facilities on a monthly basis. Potassium chloride and sodium chloride are defined among other constituents. The control results show that the maximum permissible concentrations are not exceeded (0.3 mg/m<sup>3</sup> for KCl, 0.5 mg/m<sup>3</sup> for NaCl). Taking into account the volume of emissions into the atmospheric air, the main entry of potassium chloride and sodium chloride is connected with the emissions of the drying department of the fertilizer production plant. According to the results obtained, migration of the studied radionuclides is limited to the zone of 2 km. Only insignificant part of the investigated samples (8%) exceeds the worldwide average activity of  $^{40}\text{K}$  for soils. In bottom sediments the average activity of  $^{40}\text{K}$  does not exceed the values for other territories with anthropogenic impact. The situation is different for the content of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$ . Even their maximum activities in the investigated soil samples are essentially lower than the worldwide average values.

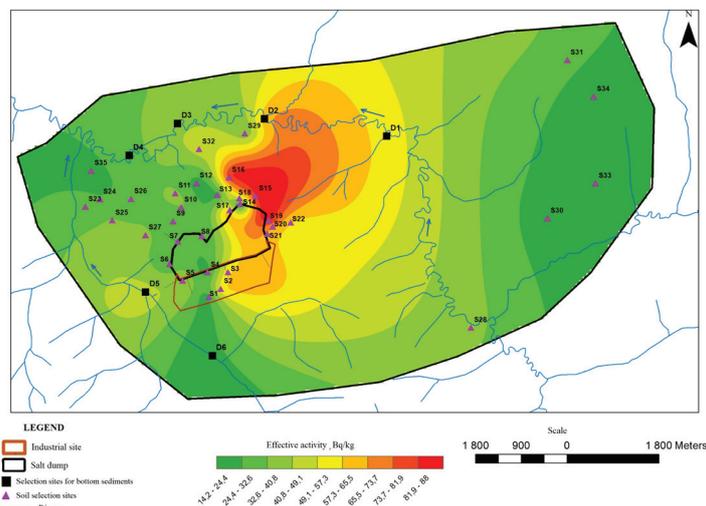
The quantitative state of radioactive equilibrium in various objects of the environment is characterized by the coefficient of radioactive equilibrium, which is the ratio of the activity of a given radionuclide to the activity of the radionuclide that originated or preceded it in the radioactive series [Vertman et al., 2002]. If the radioactive equilibrium is not disturbed, the ratio will be 1. Thus, the ratio of activity values  $^{232}\text{Th}/^{226}\text{Ra} < 1$  in soils can serve as an indicator of anthropogenic impact. The results of the conducted assessment for soils of the study area showed several zones with disturbed ratio of these natural radionuclides (Table 4, Fig. 3). The configuration of the isolated zones of imbalance indicates the absence of their connection with the activities of the mine of potassium salts.

**Table 1.** Specific activity of natural radionuclides in soils  
Activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  (Bq/kg) in soil samples

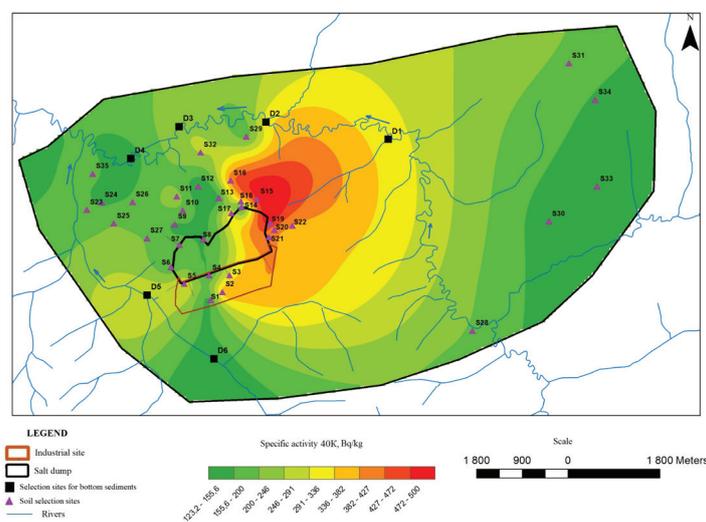
Sample code	Activity concentration (Bq/kg)			
	$^{40}\text{K}$	$^{226}\text{Ra}$	$^{232}\text{Th}$	Aeff
S1	143.30±26.00	9.10±3.00	<2,82	22.36±4.53
S2	364.50±90.40	9.70±3.80	8.62±3.73	53.70±10.20
S3	301.70±79.40	13.47±4.21	10.6±3.90	54.40±9.70
S4	157.20±27.00	3.40±2.00	4.48±2.00	22.63±3.99
S5	281.80±78.10	6.61±3.52	6.52±3.55	40.40±9.10
S6	166.10±30.00	6.76±3.00	4.08±2.00	26.22±4.70
S7	173.40±28.00	9.72±3.00	3.92±2.00	29.59±4.61
S8	154.00±27.00	6.70±3.00	4.78±2.00	26.05±4.57
S9	151.40±27.00	6.09±3.00	4.59±2.00	24.97±4.57
S10	186.50±32.00	8,99±3.00	5,75±2.00	32,37±4.79
S11	215.00±35.00	7.25±3.00	6.33±2.00	33.81±4.93
S12	123.60±23.00	8.26±3.00	<3,8	21.12±4.41
S13	155.80±27.00	6.17±3.00	3.82±2.00	24.42±4.57
S14	129.00±24.00	6.54±2.00	<3.82	19.89±3.85
S15	489.00±125.00	19.70±6.30	17.70±6.2	84.40±14.6
S16	407.00±114.00	19.10±6.70	20.50±7.20	80.60±15.00
S17	308.00±127.00	18.20±8.00	9.30±7.60	62.60±16.50
S18	432.00±51.00	15.80±9.20	12.20±9.50	68.50±19.90
S19	500.00±127.00	18.90±6.30	20.30±6.70	88.00±15.1
S20	406.00±92.00	15.60±4.20	14.70±4.17	69.30±10.35
S21	410.00±102.00	16.10±5.00	10.10±4.40	64.20±11.40
S22	385.00±109	9.30±5.50	12.90±6.20	58.90±13.30
S23	169.20±28.00	6.37±2.00	5.29±2.00	27.68±4.03
S24	151.20±26.00	5.04±2.00	5.66±2.00	25.31±3.94
S25	195.10±31.00	5.80±2.00	7.68±2.00	32.45±4.18
S26	163.50±28.00	9.17±3.00	2.89±2.00	26.85±4.61
S27	195.70±32.00	7.19±3.00	7.66±2.00	33.86±4.79
S28	195.50±35.50	9.50±3.00	6.42±3,00	34.53±5,73
S29	193.20±34.00	8.08±3.00	9.06±3.00	36.37±5.69
S30	169.3±31.00	<2.17	2.59±2.00	17.96±4.18
S31	158.8±55.00	7.09±3.27	4.53±2.98	27.30±7.10
S32	123.20±26.00	5.79±3.00	<3.00	16.26±4.53
S33	180.90±33.00	4.53±3.00	8.08±3.00	30.49±5.65
S34	141.2±28.00	4.59±3.00	<2.49	17.24±4.61
S35	144.90±27.00	<3.00	<3.06	14.60±3.98
Middle	234.91	9.14	7.43	38.55
Max	500.00±127.00	19.70±6.30	20.50±6.70	88.00±15.1
Min	123.20±26.00	<2.17	<2.49	14.60±3.98
Summary world data [UNSCEAR ..., 2000]	420	32	45	–
Average 2019 values for Moscow soils [Radiation situation ..., 2020]	442	19	24	90

**Table 2.** Specific activity of natural radionuclides in samples of bottom sediments  
Activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  (Bq/kg) in samples of bottom sediments

Sample code	Activity concentration (Bq/kg)			
	40K	$^{226}\text{Ra}$	$^{232}\text{Th}$	Aeff
D1	313.00±86.00	7.90±4.00	14.30±4.90	53.30±10.4
D2	314.00±87.00	12.60±4.60	16.80±5.30	61.30±11.00
D3	234.30±58.00	6.61±2.63	7.85±2.66	37.90±6.80
D4	144.10±26.00	9.20±3.00	6.16±2.00	29.52±4.53
D5	283.40±70.90	6.42±2.97	8.66±3.19	43.20±8/20
D6	131.20±22.00	2.39±2.00	6.08±2.00	21.51±3.77
Middle	236.67	7.52	9.98	41.12
Summarized data on the content in the sediments of Lake Karun [Rafat, 2015]	410–1426	6,2–22,4	5,2–26,6	–
Average 2019 values for the bottom sediments of Moscow [Radiation situation ..., 2020]	279	15	20	66
Summary data on Sedimentary rocks [Paschoa et al., 2010]	<300–900	–	<8–50	–



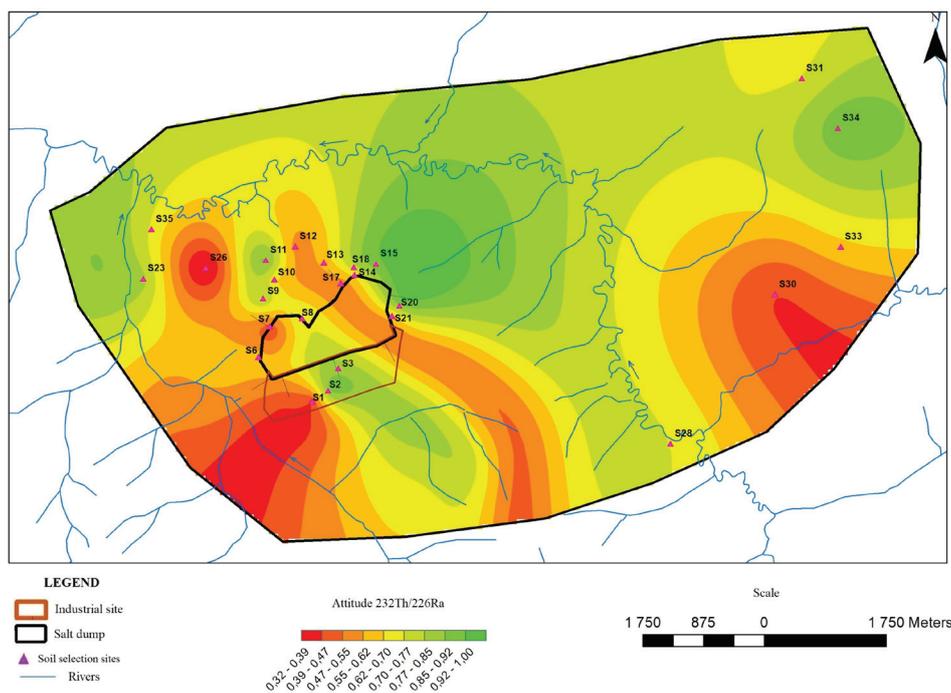
**Figure 1.** Specific effective activity of natural radionuclides in soils and bottom sediments in the study area



**Figure 2.** Specific activity of  $^{40}\text{K}$  in soils and bottom sediments in the study area

**Table 3.** Specific activity of natural radionuclides in rocks of exploration well and crushed ore Activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K (Bq/kg) in rocks from an exploration well and shredded ore

Sample code	Rocks (sampling interval, m)	Activity concentration (Bq/kg)			
		<sup>40</sup> K	<sup>232</sup> Th	<sup>226</sup> Ra	Aeff
SI-1	Loam (3,5–4,0)	333±55	15,3±5	17,3±3	67,16±14,45
SI-2	Mergel (13,0–13,5)	177±37	7,5±4	4,9±3	30,58±11,53
SI-3	Clayey Marl (26,2–26,4)	382±60	12,9±5	7,3±3	58,45±14,90
SI-4	Clayeymergel (29,4–29,5)	346±56	17,3±5	7±3	60,63±14,54
SI-5	clayeymergel (151,9–152,1)	359±57	20,9±5	4,6±3	64,08±14,63
SI-6	Gypsummergel (173,4–173,8)	927±195	22,2±6,26	6,26±4,89	118,55±30,58
SI-7	Rocksalt (197.0–197.3)	3±0	3,2±3	3±0	7,43±3,9
SI-8	Carnallite (226.7–227.0)	2896±330	19,8±5	3±0	289,38±36,2
SI-9	Carnallite (245.5–245.9)	3031±340	17,8±5	3±0	298,93±37,1
SI-10	Carnallite (272.8–273.2)	2824±320	16,7±5	3±0	278,87±35,3
SI-11	Mottled sylvinite (282,6–283,0)	4967±540	38,3±8	3±0	499,82±59,00
SI-12	Striated sylvinite (284,9–285,4)	3393±380	41±8	3±0	361,67±44,60
SI-13	Red sylvinite (296,5–297,1)	2055±240	16,4±5	3±0	209,27±28,10
SI-14	Striated sylvinite (304,0–304,55)	4542±470	24,9±6	3±0	418,05±50,10
SI-15	Rocksalt (348.3–348.9)	3±0	3±0	3±0	7,17±0,00
Crushed ore					
GS-1	Ground sylvinite	4510 ± 730	<8	14,5 ± 4,2	398 ± 66
GS-2	Ground sylvinite	4612 ± 742	<8	<8	418 ± 67
GG-1	Halite waste	283 ± 70	<8	<8	24 ± 6
GG-2	Halite waste	297 ± 66	<8	<8	27 ± 7
Summary of salt data the Khewra Mines, Pakistan [Baloch et al., 2012]		36±20	–	–	–
Summary data on rocks [Paschoa et al., 2010]					
Sedimentary rocks		<300–900	<8–50	–	–
Continental crust		850	44	–	–



**Figure 3.** Ratio of specific activity of <sup>232</sup>Th and <sup>226</sup>Ra in soil cover

**Table 4.** Significant ratios of the value of specific activity  $^{232}\text{Th}$  and  $^{226}\text{Ra}$  in soil cover

Sample code	Activity concentration (Bq/kg)		$^{232}\text{Th}/^{226}\text{Ra}$
	$^{232}\text{Th}$	$^{226}\text{Ra}$	
S1	2,8	9,1	0,3
S2	8,6	9,7	0,9
S3	10,6	13,5	0,8
S6	4,1	6,8	0,6
S7	3,9	9,7	0,4
S8	4,8	6,7	0,7
S9	4,6	6,1	0,8
S10	5,8	9	0,6
S11	6,3	7,3	0,9
S12	3,8	8,3	0,5
S13	3,8	6,2	0,6
S14	3,8	6,5	0,6
S15	17,7	19,7	0,9
S17	9,3	18,2	0,5
S18	12,2	15,8	0,8
S20	14,7	15,6	0,9
S21	10,1	16,1	0,6
S23	5,3	6,4	0,8
S26	2,9	9,2	0,3
S28	6,4	9,5	0,7
S30	3,2	7,2	0,4
S31	5,4	7,3	0,7
S33	4,4	7,7	0,6
S34	3,0	3,8	0,9
S35	3,4	4,7	0,7
Range of indicators for the soils of the study area	2,8–17,7	3,8–19,7	0,3–0,9
Significance of indicators in soil-forming rocks	15,3±5	17,3±3	0,88

## CONCLUSIONS

Monitoring of changes in radiation background and radionuclide activity is currently an important task in order to assess the radiation situation in industrial and urbanized areas. In spite of the high activity of  $^{40}\text{K}$  in the developed potassium ores, the value of this index for the majority of investigated soil samples at the Verkhnekamskoye salt deposit does not exceed worldwide average values, for samples of bottom sediments – values of other territories with anthropogenic influence. Migration of the investigated radionuclides is limited to the zone of 2 km. Even the maximum activities of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$

in the investigated soil samples are significantly lower than the worldwide average values. All this allows us to conclude that the underground development of the deposit has no effect on the radiation situation of the territory under study. The results of the studies can be used in solving the problems of radiation monitoring of mining enterprises and environmental assessment of territories, taking into account the radiation loads.

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