

EVALUATION OF THE IMPACT OF DIAMOND MINING ON THE RADIO-ECOLOGICAL STATE OF THE ARCTIC ZONE ECOSYSTEMS (EXAMPLE OF ARKHANGELSK REGION, RUSSIA)

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ABSTRACT

The article was devoted to the study of the activity of natural and technogenic radionuclides in the environment components around the Lomonosov diamond deposit. This is the largest diamond mining and beneficiation complex in Europe. Samples of river sediments and surface waters were taken from the Zolotitsa River and its tributaries in the area of the diamond deposit. Samples of kimberlites and rocks were selected to assess the radioactivity parameters of the rocks extracted from the quarries. Radionuclide activity in bottom sediments and rocks was measured using low-background semiconductor gamma spectrometry with HPGe high-purity germanium detector, and isotopes ²³⁴U and ²³⁸U in bottom sediments while the waters were studied using alpha spectrometry. The influence of mining and beneficiation complex on the increase in radionuclides activity in bottom sediments of Zolotitsa River was found. The parameters of radiation safety of bottom sediments were calculated regarding the human health of the personnel of the mining and beneficiation complex and the mining camp. The values of radiation parameters were found to be, on average, below the global average, and therefore, radiation parameters do not pose a significant hazard to the personnel and public. Several patterns of radionuclide accumulation have been found depending on the physicochemical parameters of river sediments. These patterns are due to the technogenic influence of the mining and beneficiation complex.

KEY WORDS : Radionuclides, Sediments, Water, Hazard parameters, Zolotitsa river, Lomonosov diamond deposit.

INTRODUCTION

Since the second half of the 20th century, the Arctic zone of Russia has been significantly affected by technogenic radioactivity. The main radiation facility in the Russian Arctic is the Novaya Zemlya test site, where from 1954 to 1990, 87 explosions were carried out in the atmosphere, 3 underwater and 42 underground (Aybulatov, 2000; Vasiliev *et al.*, 2005). Moreover, the West European radiochemical plants (Sellafield) and plants located in Siberia (Mayak, Seversk) affected significantly the radiation situation in the Russian Arctic (Aybulatov, 2000). For geological purposes, several underground nuclear explosions were set off in the Arctic (Kiselev *et al.*, 2014). The accident at the Chernobyl nuclear power

plant as a result of global precipitation also had a certain effect (Kiselev *et al.*, 2005). As a result of the combined influence of technogenic radiation objects, a significant amount of technogenic radionuclides, such as ¹³⁷Cs, ⁶⁰Co, ²³⁸Pu, ²³⁹⁺²⁴⁰Pu, ²⁴¹Am, got into the ecosystems of the Arctic (Gwynn *et al.*, 2004; Łokas *et al.*, 2014; Matishov *et al.*, 2019). However, the contribution of technogenic radionuclides to human and ecosystem radiation is not so profound, since the greatest impact on the exposure of the population is made by the natural sources of ionizing radiation that are present in all natural objects - in bottom sediments, minerals, rocks, surface and underground waters, air aerosols, construction materials (UNSCEAR, 2008; Rikhvanov, 2009; NRB - 99/2009). The main natural radionuclides are ²²⁶Ra,

^{232}Th and ^{40}K . The prime attention in radioecological studies is given to precisely these radionuclides (Cevik, 2007; Yü *et al.*, 2009; Rafique *et al.*, 2014; Ravisankar *et al.*, 2015; Uosif *et al.*, 2016; Kiselev *et al.*, 2017; Fallah, 2019; Zorer, 2019).

On the territory of the European Arctic of Russia, to which the Arkhangelsk region belongs, there is an increased radiation background due to natural causes - the geological structure of the territory (Kiselev *et al.*, 2005; Teleleikova, Evseev, 2014). Thus, the most widespread rocks of the Baltic Shield granite gneiss are enriched with natural radionuclides ^{226}Ra , ^{232}Th and ^{40}K (Smyslov *et al.*, 1974; Titaeva, 2005). In this area, many different metal mineral deposits were discovered, including uranium ore occurrences (Voitekhovskiy *et al.*, 2008). The development of mineral deposits associated with the extraction of large volumes of rocks leads to the release into the environment of significant volumes of heavy metals and radionuclides of natural origin, in concentrations uncharacteristic of the earth's surface. Therefore, the development of mineral deposits entails a significant change in the environment, including its radioactive state within the mining areas (Fedorets, 2001; Kiselev, 2005; Panteleeva, 2006; Panteleeva, 2007; Panteleeva, 2009; Huang *et al.*, 2010; Novikov, 2016; Fei *et al.*, 2017; Kiselev *et al.*, 2018₁). In the Arkhangelsk region, the rocks of the Baltic Shield (granite gneisses enriched with radionuclides) do not reach the surface, however, in some areas in the sedimentary cover rocks, increased concentrations of natural radionuclides are detected (Kiselev *et al.*, 2018_{2,3,4}). In particular, this is characteristic of the Lomonosov diamond deposit, located 90 km north of the city of Arkhangelsk (Yakovlev, 2019).

Lomonosov field is the largest industrial diamond deposit in Europe. The field was discovered relatively recently in the 80s of the last century, however, commercial development of the field began only in the early 2000s with the development of the first Arkhangelsk pipe, and in 2014 the development of the second pipe Karpinsky-1 began (Verzhak *et al.*, 1987). The main waterway flowing in the area of deposit is the Zolotitsa River, which belongs to the White Sea basin (Bednaruk, 2008). The Zolotitsa River has a conservation status, since it is the largest spawning ground for salmon in the White Sea basin (Kalyuzhin, 2003). The river valley is very poorly populated, in the upper reaches there is only a shift camp of a mining and processing factory, and in the lower reaches there are several

fishing villages of Pomors, using the waters of the Zolotitsa River for drinking. The only technogenic object in this territory that can be a source of pollution of the Zolotitsa River ecosystems is the mining and beneficiation complex. Currently, active development of the field is being carried out, involving the extraction of large volumes of ore, which includes a full beneficiation cycle, treatment of drainage water, construction of dumps and tailings, water reduction, construction of swamp-filtration fields (Soldatova, 2016). Due to the fact that the rocks extracted from the quarries are enriched with natural radionuclides, a possible change in the radioecological state of the environmental components in the area of the Lomonosov field is wary (Kiselev *et al.*, 2018_{1,2,3,4}; Yakovlev, 2019). In connection with the increase in diamond mining, the deepening of quarries, the development of new kimberlite pipes, the increase in the population of the shift community and the personnel of the processing plant, as well as in connection with the special protection status of the Zolotitsa River, it is extremely important to conduct radioecological research of the territory. The aim of the study is to determine the activities of natural radionuclides ^{226}Ra , ^{232}Th and ^{40}K and the technogenic isotope ^{137}Cs in bottom, rocks and surface waters and to assess the radiological risks for the population of the shift camp and the personnel of the concentrating plant.

MATERIALS AND METHODS

The general chart of the research area, the location of the sampling spots for river sediments, rocks from quarries, as well as surface waters are shown in Fig. 1.

Samples of bottom sediments and surface water were taken from the river Zolotitsa River and all its feeders in the area of the Lomonosov diamond field. Samples of rocks and ores were taken from the quarries of the Arkhangelsk and Karpinsky-1 pipes. A total of 48 samples of bottom sediments, 11 samples of river water and 55 samples of rocks and ores from quarries were collected.

Bottom sediments were collected using a Peterson hand-held sampler. After sampling, bottom sediment samples were dried in a BINDER E28 oven at a temperature of 105 °C until air-dry. For the measured activities ^{226}Ra , ^{232}Th , and ^{40}K , radiation safety parameters were calculated, such as the radium equivalent (Raeq), the external hazard index (H_{ex}), and the absorbed dose rate of gamma

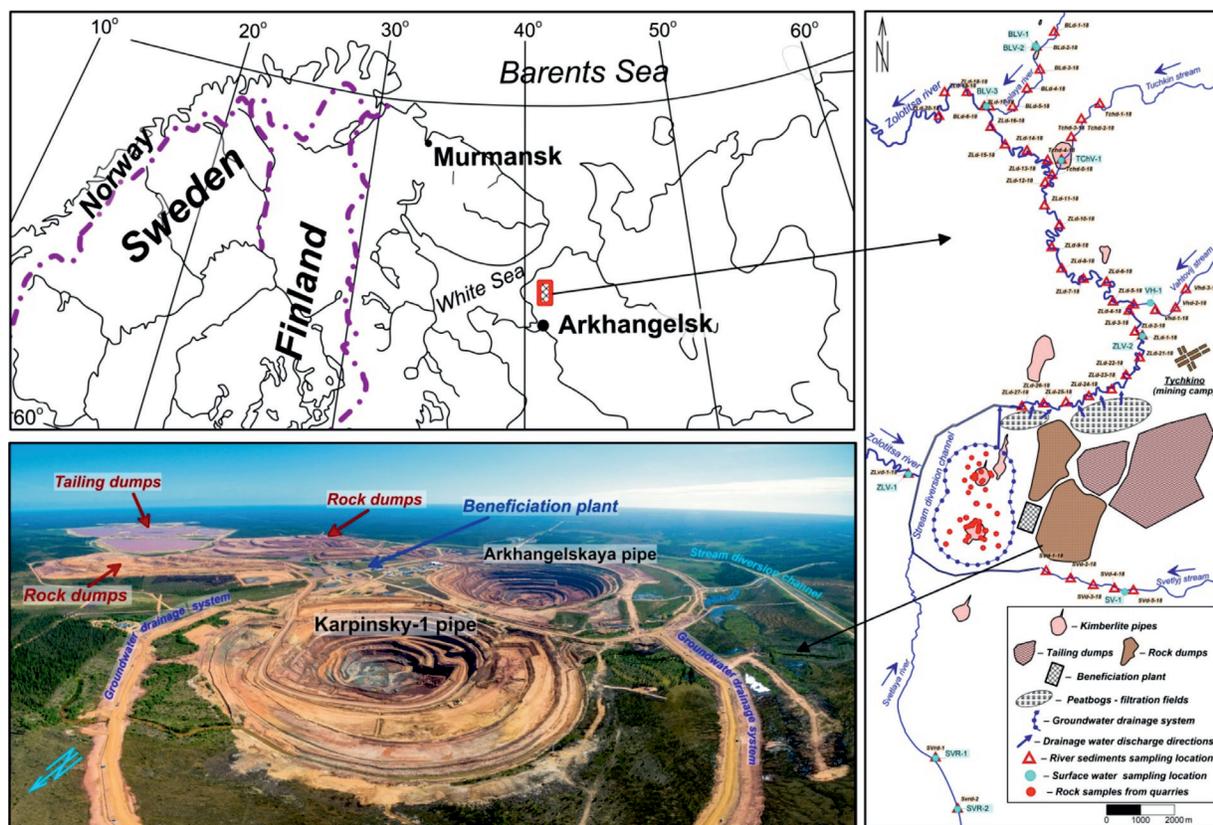


Fig. 1. Location of the studied area and sampling points (photo of deposit <http://www.severalmaz.ru/en/>)

radiation (D).

Gamma spectrometric measurements

The radionuclides ^{137}Cs , ^{226}Ra , ^{232}Th , and ^{40}K were determined using an ORTEC low-background semiconductor gamma-spectrometer (USA) based on a GEM10P4-70 coaxial detector from high-purity germanium (HPGe), a DSPEC LF digital analyzer, and MAESTRO-32 software. The resolution of gamma radiation along the 1.33 MeV (^{60}Co) line is 1.75 keV with a relative efficiency of 10%. The detector was calibrated using a standard point source KI-660 $^{137}\text{Cs} + ^{40}\text{K}$ (NPP "Doza", Russia). Measurements were performed in Marinelli geometry.

Measurement of the isotopic composition of uranium in surface waters

Preliminary radiochemical preparation was carried out directly in the field by concentrating a water sample with a volume of 20-30 L using coal BAU-A (Chalov, 1991; VIMS, 2013). To assess the degree of uranium yield, a tracer, an artificial ^{232}U isotope, was added to water samples. Separation from other interfering radionuclides with close alpha-radiation

energies was carried out by tributyl phosphate extraction. Sources for alpha spectrometric detection were prepared by electrolytic deposition of uranium on stainless steel disks with a diameter of 34 mm. The measurements were made on a "Progress" semiconductor alpha spectrometer with a 400 mm² silicon surface-barrier detector. The relative measurement error is about 10%.

RESULTS AND DISCUSSION

Activity concentrations of radionuclides in the sediments

The results of the studies of the concentration of radionuclides in bottom sediments of the Zolotitsa River and its feeders in the area of the Lomonosov deposit (Belaya River, Svetlaya River, Tuchkin Stream, Svetly Stream, Vakhtovy Stream) are shown in Table 1. The average radionuclide concentrations decrease in the order of $^{40}\text{K} > ^{232}\text{Th} > ^{226}\text{Ra} > ^{137}\text{Cs}$. The average activities of ^{137}Cs , ^{226}Ra , ^{232}Th , ^{40}K radionuclides in general for the studied bottom sediments are 5.4, 9.0, 11.3, 321.6 Bq · kg⁻¹, respectively.

Table 1. Concentration of ^{137}Cs , ^{232}Th , ^{226}Ra , ^{40}K and the radium equivalent, absorbed dose rate and external hazard index of sediment samples from the Zolotitsa River and its tributaries

Sample No	^{137}Cs ($\text{Bq}\cdot\text{kg}^{-1}$)	^{226}Ra ($\text{Bq}\cdot\text{kg}^{-1}$)	^{232}Th ($\text{Bq}\cdot\text{kg}^{-1}$)	^{40}K ($\text{Bq}\cdot\text{kg}^{-1}$)	Raeq ($\text{Bq}\cdot\text{kg}^{-1}$)	Dose rate ($\text{nGy}\cdot\text{h}^{-1}$)	H_{ex}
ZLvd-1-18	2.3±0.6	7.0±0.8	5.8±0.8	261±27.4	34	18	0.10
ZLd-27-18	5.2±1.0	7.9±0.9	10.0±1.3	278±19.5	42	21	0.12
ZLd-26-18	9.5±1.9	10.0±1.2	11.0±1.8	334±30.1	49	25	0.14
ZLd-25-18	11.0±2.1	9.6±1.2	15.6±1.9	294±23.5	52	26	0.15
ZLd-24-18	15±2.3	29±3.5	37±3.3	471±37.7	115	56	0.32
ZLd-23-18	14.3±2.9	12.8±1.5	13.2±1.6	293±26.4	52	26	0.15
ZLd-22-18	12.6±2.5	7.4±1.0	17.3±1.9	370±33.3	58	29	0.16
ZLd-21-18	11.7±2.3	21±2.7	25±2.8	418±42.0	86	42	0.24
ZLd-1-18	2.5±0.6	4.7±0.8	5.7±0.8	307±33.8	34	19	0.10
ZLd-2-18	1.5±0.3	3.0±0.6	7.8±0.9	219±17.3	29	15	0.08
ZLd-3-18	2.7±0.5	2.4±0.5	5.9±0.8	219±20.2	26	14	0.07
ZLd-4-18	11.4±2.3	15.0±1.8	16±2.4	373±32.4	64	32	0.18
ZLd-5-18	1.5±0.3	3.8±0.7	8.0±1.1	221±24.3	31	16	0.09
ZLd-6-18	2.4±0.6	4.2±0.6	7.5±1.1	267±23.8	34	18	0.10
ZLd-7-18	0.8±0.3	12.0±1.6	13±1.6	331±36.4	54	27	0.15
ZLd-8-18	2.2±0.6	3.4±0.7	8.4±1.1	218±19.7	31	16	0.09
ZLd-9-18	2.4±0.5	3.0±0.5	8.3±1.0	407±41.5	43	23	0.12
ZLd-10-18	2.2±0.4	9.6±1.3	7.1±0.9	341±30.7	44	23	0.12
ZLd-11-18	3.3±0.7	13.0±1.4	15±2.1	409±38.9	63	32	0.18
ZLd-12-18	5.2±1.0	19.0±2.3	27±3.2	514±36.0	94	47	0.26
ZLd-13-18	5.3±1.1	4.8±0.8	8.2±0.9	289±28.9	37	19	0.10
ZLd-14-18	2.4±0.7	11.6±1.3	12.1±1.2	428±38.5	59	31	0.17
ZLd-15-18	7.4±1.4	6.5±0.8	9.5±1.3	334±30.1	43	23	0.12
ZLd-16-18	1.9±0.7	3.0±0.4	6.6±0.9	262±28.8	31	16	0.09
ZLd-17-18	3.1±0.6	5.8±0.6	5.2±0.7	187±20.6	26	14	0.07
ZLd-18-18	6.3±1.3	37±4.4	59±4.7	999±69.9	191	95	0.54
ZLd-19-18	1.5±0.5	7.9±1.1	8.6±1.0	175±15.8	32	16	0.09
ZLd-20-18	1.5±0.6	7.6±1.1	5.5±0.8	232±18.6	32	17	0.09
Vhd-1-18	1.5±0.4	9.4±1.2	4.8±0.7	185±20.4	29	15	0.08
Vhd-2-18	4.7±0.9	3±0.5	6.6±1.0	228±25.2	28	15	0.08
Vhd-3-18	1.8±0.4	5.4±1.0	2.8±0.4	234±16.4	26	14	0.07
Tchd-0-18	3.5±0.7	6.1±1.2	24.8±3.5	533±53.3	79	40	0.22
Tchd-1-18	4.6±1.2	6.8±0.9	4.8±0.6	230±23.0	30	16	0.08
Tchd-2-18	2.4±0.7	3±0.5	9.4±0.8	344±37.8	41	22	0.12
Tchd-3-18	5.4±1.0	3.5±0.5	2.7±0.3	251±25.1	25	14	0.07
Tchd-4-18	3.5±0.7	4.7±0.6	4.1±0.4	287±31.6	31	17	0.09
BLd-1-18	18.4±3.7	18.3±2.6	13±1.4	343±27.4	61	31	0.17
BLd-2-18	4.3±0.9	8.5±1.5	20.7±2.9	405±36.5	66	33	0.19
BLd-3-18	7.5±1.4	5.5±0.8	4.9±0.6	382±34.4	39	22	0.11
BLd-4-18	10.4±2.1	18±2.0	13.6±1.9	363±39.8	63	32	0.18
BLd-5-18	1.5±0.3	28.6±3.4	13±1.8	264±23.8	66	32	0.18
BLd-6-18	7.9±1.2	16±2.6	17.9±2.3	424±29.7	71	36	0.20
SVd-1-18	11.1±2.3	3.6±0.6	8.6±1.4	195±18.5	30	15	0.08
SVd-2-18	4.7±0.9	2.4±0.3	3.6±0.5	263±26.9	26	14	0.08
SVd-3-18	4.5±0.9	1.9±0.3	4.5±0.4	258±19.1	26	14	0.08
SVd-4-18	4.1±1.0	2.2±0.5	3.4±0.5	257±23.1	25	14	0.07
SVd-5-18	3.9±1.1	2.6±0.4	3.1±0.4	256±22.0	25	14	0.07
SVrd-1-18	4.1±0.8	2.3±0.3	3.3±0.5	256±21.4	25	14	0.07
SVrd-2-18	4.0±0.8	5.9±0.9	<1	211±18.2	21	12	0.06
Min	0.8	1.9	<1	175.0	21	12	0.06
Max	18.4	37.0	59.0	999.0	191	95	0.54
Average	5.4	9.0	11.2	318.8	47.3	24.4	0.13

According to Table 2, the activity values of natural radionuclides are below the world average (UNSCEAR, 2000).

In the bottom sediments of the lakes of the Arkhangelsk region, the average radionuclide activity is slightly higher and amounts to for ^{137}Cs , ^{226}Ra , ^{232}Th and ^{40}K 33, 21, 23 and 300 $\text{Bq}\cdot\text{kg}^{-1}$, respectively, close to the value characteristic of the North-West of Russia, except ^{137}Cs (Kiselev *et al.*, 2017). Of the radionuclides studied in this work, the closest in activity to the values characteristic of the Arkhangelsk region and North-West Russia is activity of ^{40}K $\sim 300 \text{ Bq}\cdot\text{kg}^{-1}$ (Bykov, 2018). The prevalence of ^{40}K in the bottom sediments of the river Zolotitsa River and its tributaries compared with other radionuclides is due to the fact that ^{40}K is the most common radionuclide in rocks and prevails in many light minerals (Ergul *et al.*, 2013). In addition, the increase of ^{40}K relative to other radionuclides is explained by the mineral composition of the channel sediments of the studied territory containing weathering products of acidic igneous rocks (granites), which are present in the quaternary fluvioglacial deposits developed in the Zolotitsa River. Thus, compared with global values, the lower activity of radionuclides in river sediments of the Zolotitsa and its tributaries can be explained by the hydrological and geomorphological features of the river related to the

last glaciation (a narrow channel, relatively high flow rate, and predominance of bottom erosion); the mineral composition of the river's sediments is mostly of the same type and consists mainly of fine-grained, well-washed sand, which is a product of rock destruction as the river actively cuts its course. In relation to this, the active sedimentation processes of particles dissolved and suspended in water in the studied site of the bed of the Zolotitsa River do not occur compared to these processes in the lakes of northwest Russia.

Nevertheless, with such relatively low average activities of radionuclides in the bottom sediments of the river Zolotitsa River and its tributaries, some heterogeneities in the variations of radionuclides are still observed. Spatial analysis of the data allows us to highlight a number of patterns in the distribution of radionuclides in bottom sediments of the Zolotitsa River. Figure 2 shows graphs of changes in the activity of ^{137}Cs , ^{226}Ra , ^{232}Th , and ^{40}K radionuclides in bottom sediments of the Zolotitsa River in the direction of the river flow from south to north (from sample ZLvd-1-18 to ZLd-20-18).

Within the territory of the Lomonosov mining and beneficiation complex, the Zolotitsa riverbed was changed in connection with the development of quarries. At present, the Zolotitsa River channel in this section is a technological channel (Fig. 1).

The first sample of bottom sediments ZLvd-1-18

Table 2. Average activity concentration of radionuclides in the sediment samples worldwide

Country, locations	^{137}Cs ($\text{Bq}\cdot\text{kg}^{-1}$)	^{226}Ra ($\text{Bq}\cdot\text{kg}^{-1}$)	^{232}Th ($\text{Bq}\cdot\text{kg}^{-1}$)	^{40}K ($\text{Bq}\cdot\text{kg}^{-1}$)	References
Bangladesh, Karnaphuli river	-	39.7	65.5	272	Chowdhury <i>et al.</i> , 1999
China, Huanghe river mouth	-	28.6	57.9	542	Wu <i>et al.</i> , 2015
Egypt, Red Sea shore sediment	-	24.6	31.4	428	El-Mamony and Khater, 2004
Egypt, Brullus lake	-	14.3	20	312	El-Reefy <i>et al.</i> , 2010
Egypt, Red sea Marine sediment	-	27.4	38.5	419.4	El-TaHER and Madkour, 2011
Egypt, Safaga coast of Red Sea,	-	14.3	17.1	346.5	Uosif <i>et al.</i> , 2016
Ghana, Greater Accra, Tema Harbour	1.5	14	30	325	Botwe <i>et al.</i> , 2017
Greece, Milos volcanic island	-	67	45	691	Papaefthmiou <i>et al.</i> , 2007
Iran, Arvand river	1.96	16.10	16.47	280.9	Fallah <i>et al.</i> , 2019
Italy, Tyrrhenian sea	-	28.2	91.7	60.4	Desideri <i>et al.</i> , 2002
Malaysia, Northern peninsular	-	51	22	189	Muhammad <i>et al.</i> , 2012
Russia, largest lakes of NW	62.9	20.0	21.8	357.5	Kiselev <i>et al.</i> , 2017
Thailand, Pattani Bay,	-	4.9	55.8	183.2	Kaewtubtim <i>et al.</i> , 2017
Turkey, Lake Van basin	4.37	47.48	57.87	1021.4	Zorer, 2019
Turkey, Izmit Bay	21	18	-	568	Ergul <i>et al.</i> , 2013
USA, Reedy river	-	21.4	45.3	609	Powell <i>et al.</i> , 2007
Yugoslavia, Bga canal	-	71	49	520	Bikit <i>et al.</i> , 2005
NW Russia, Zolotitsa river	5.4	9.0	11.3	321.6	Present work
Worldwide	-	33	45.0	420	UNSCEAR., 2000

was taken at the spot before the river flows into the channel. As seen from Table 1 and Fig. 2. ZLvd-1-18 sample is characterized by the lowest activities of ^{137}Cs , ^{226}Ra , ^{232}Th and ^{40}K - 2.3, 7.0, 5.8 and 261 $\text{Bq}\cdot\text{kg}^{-1}$, respectively, in comparison with bottom sediments of the channel section from the northern boundary of mining and beneficiation complex (ZLd-27-18) to the section of the channel in the area of the Tuchkino camp (ZLd-21-18). In this section, bottom sediments, represented by 7 samples, are characterized by the highest average values of ^{137}Cs , ^{226}Ra , and ^{232}Th relative to other studied sections located downstream of the Zolotitsa River. On the rest of the segment of the river Zolotitsa peaks of increased activity of radionuclides have a pronounced confinement to the river entry parts of tributaries: Vakhtoviy stream, Tuchkin stream and the Belaya River (Fig. 2). Increased activity of radionuclides in the river entry parts of the tributaries of the river Zolotitsa can be associated with the removal of radionuclides from the catchment areas of these tributaries as a part of finely dispersed material, characterized by increased sorption properties and their accumulation near the river entry parts. As for the wide zone with elevated concentrations of ^{137}Cs , ^{226}Ra and ^{232}Th radionuclides in the section from the northern border of the Lomonosov MPP to the section of the Zolotitsa River channel in the area of the Tuchkino camp, the explanation of the observed values requires a separate consideration. Due to the fact that in this section of the riverbed there are no any tributaries of Zolotitsa River, the influence of mining and beneficiation complex on the change in the radionuclide composition of river sediments is

probable. To this end, we studied the activity of radionuclides in ores and rocks of extracted and two quarries of the deposit as possible sources of radionuclide pollution of river sediments (Table 3).

Sandy-clay deposits of the Vendian, Carboniferous and Quaternary ages extracted from quarries are stored in dumps (Fig. 1). A light fraction of processed kimberlites containing saponites is stored in tailings (Fig. 1). As can be seen from Table 3, the enclosing Vendian rocks extracted from quarries and transported into dumps contain on average almost twice as much ^{226}Ra and three times as much as ^{232}Th than the sediments of Zolotitsa River. The concentration of ^{226}Ra in the overlapping Quaternary and Carboniferous sediments is comparable with the activity of radium in river sediments, but ^{232}Th in them is twice as much. Kimberlites are also enriched in ^{226}Ra and ^{232}Th , relative to river sediments, but are depleted in ^{40}K . For other types of rocks, the average activity of ^{40}K are at the same level of $\sim 300 \text{ Bq}\cdot\text{kg}^{-1}$. Technogenic ^{137}Cs were not detected in the rocks and ores of the deposit, as expected. Rock dumps containing increased activity of natural radionuclides are subject to weathering and erosion by atmospheric precipitation. As a result, the transfer of radionuclides by temporary streams in the Zolotitsa River is possible and their accumulation in river sediments in the near zone of the Lomonosov mining and beneficiation complex.

Another source of increased activity of radionuclides in sediments of the Zolotitsa River in the near zone of influence of mining and beneficiation complex can serve as drainage water.

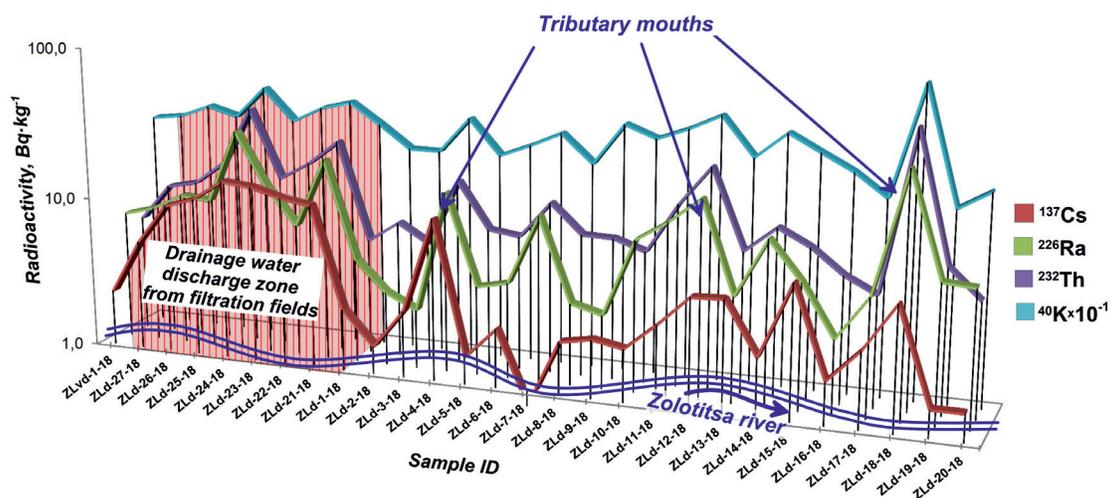


Fig. 2. Graphs of changes in the activity of ^{137}Cs , ^{226}Ra , ^{232}Th , and ^{40}K radionuclides in bottom sediments of the Zolotitsa River in the direction of the river flow from south to north.

Table 3. Average concentration values of radionuclides and radiation hazard parameters in the studied quarry rocks and river sediments

Type of sediment	Number of Samples	^{137}Cs (Bq·kg ⁻¹)	^{226}Ra (Bq·kg ⁻¹)	^{232}Th (Bq·kg ⁻¹)	^{40}K (Bq·kg ⁻¹)	Raeq (Bq·kg ⁻¹)	Dose rate (nGy·h ⁻¹)	H _{ex}
Overlying rocks, Q and C ₂ *	10	-	9.2	21.0	318.0	61.5	30.3	0.17
Enclosing rocks, V ₂ *	27	-	18.2	32.7	317.5	87.2	41.5	0.24
Kimberlites, D ₃ -C ₂ *	18	-	13.6	19.1	230.3	57.1	27.5	0.16
River sediments	49	5.4	9.0	11.2	318.8	47.3	24.4	0.13

*Q – Quaternary fluvio-glacial, glacial, lacustrine, marsh and alluvial sediments (sand, loam, pebble, sandy loam),

*C₂ – Middle Carboniferous sediments (sandstones, limestones and dolomitic limestones),

*V₂ – Upper Vendian sediments (siltstones, argillites and sandstones),

*D₃-C₂ – Kimberlites of Upper Devonian–Middle Carboniferous age.

The technology for cleaning drainage water generated during drainage of quarries by pumping with surface pumps is carried out by dumping quarry water to peatbog-filtration fields located northeast of the mine near the dumps (Fig. 1). The kimberlite rocks of the deposit are characterized by a very high degree of chemical weathering, due to which almost all of the initial igneous material is represented by a high magnesian clay mineral saponite, the content of which in kimberlites is up to 75-100% (Karpenko, 2008; Posukhova *et al.*, 2013). During mining operations, the latter falls into the quarry waters, forming a finely dispersed suspension in it, which has very low sedimentation rates (~ 0.004 cm/h) and a density of the formed sediment (~ 70%) (Malov, 2004). As a result, the quarry waters pumped and discharged to the filtration fields also contain significant amounts of saponite (Malov, 2018_{1,2}). It is likely that ^{226}Ra (U) and ^{232}Th are also present in the composition of the saponite suspension entering the quarry waters, since the rocks of the exocontacts of kimberlites are significantly enriched in ^{234}U , ^{238}U , ^{226}Ra , ^{232}Th (Kiselev *et al.*, 2018_{2,3,4}; Yakovlev, 2019).

Apparently, complete purification of pumped quarry waters as a result of filtration through the swamp massif does not occur. In this case, the unloading of swamp (actually drainage) waters enriched with ^{226}Ra and ^{232}Th into the Zolotitsa riverbed occurs on a fairly wide stretch of about 3 km from the northern boundary of the mining and beneficiation complex to the Tuchkino camp (Fig. 1). In this section, the highest concentrations of technogenic ^{137}Cs are observed relative to other sections of the Zolotitsa River. The technogenic ^{137}Cs isotope formed during nuclear tests in the atmosphere and accidents at nuclear power plants and entered the earth's surface as a result of global

precipitation cannot be present in the drainage water, therefore its presence in bottom sediments is explained solely by flushing from the earth's surface. During the discharge of drainage water to the swamp, ^{137}Cs is activated, which is recorded in the upper peat layer as part of high molecular weight organic compounds (mainly humic acids), its migration from a large area of the swamp massif, and its discharge into the Zolotitsa riverbed together with ^{226}Ra and ^{232}Th .

Radium equivalent activity (Raeq)

Radium equivalent activity (Raeq) is widely used to assess the radiation safety of natural objects and materials. The calculation of the index was conducted according to the equation in the work by Beretka and Mathew (1985):

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.07C_{K}$$

Where C_{Ra} , C_{Th} and C_K are specific activities of ^{226}Ra , ^{232}Th and ^{40}K in Bq·kg⁻¹, respectively. As shown in Table 1, the radium equivalent index (Raeq) in the bottom sediments of the Zolotitsa River and its tributaries ranges from 21 to 191 Bq·kg⁻¹, with an average of 47.3 Bq·kg⁻¹. The average Raeq index in rocks and ores extracted from the quarries of the field is slightly higher than in the river sediments of the study area (Table 3). The radium equivalent index (Raeq) for rocks of Quaternary (Q) and Middle Carboniferous (C₂) is 61.5 Bq·kg⁻¹, for kimberlites (D₃-C₂) 57.1 Bq·kg⁻¹ and for Vendian rocks (V₂) 87.2 Bq·kg⁻¹, respectively. As can be seen, from a comparison of the data, the rocks extracted from the quarries can be potential sources of contamination of the adjacent territories with radionuclides. Also as shown, the Raeq index fluctuates in fairly wide aisles, but it does not exceed the maximum value of 370 Bq·kg⁻¹ established by the

Nuclear Energy Agency (NEA-OECD, 1979).

Absorbed gamma dose rate (D_R)

The absorbed dose of gamma radiation (D_R) is the fundamental dosimetric value. The calculation of the absorbed dose was conducted in accordance with (UNSCEAR, 2000):

$$D_R = 0.462C_{Ra} + 0.604C_{Th} + 0.042C_K,$$

where D_R is the absorbed dose of ionizing radiation in $\text{nGy}\cdot\text{h}^{-1}$, and C_{Ra} , C_{Th} and C_K are specific activities of ^{226}Ra , ^{232}Th and ^{40}K in $\text{Bq}\cdot\text{kg}^{-1}$. The range of changes in the value of the absorbed zone varies from 12 to 95 $\text{nGy}\cdot\text{h}^{-1}$. The average D_R value for river sediments is 24.4 $\text{nGy}\cdot\text{h}^{-1}$. For quarry rocks and ores, the average value of the absorbed dose is slightly higher, and the maximum average value is characteristic of sandy-clay rocks of the Vendian of the Padun suite (V_2), which is 41.5 $\text{nGy}\cdot\text{h}^{-1}$. It can be noted that the average absorbed dose rate of gamma radiation in the river sediments of the study area is approximately two times lower than the world average value of 55 $\text{nGy}\cdot\text{h}^{-1}$ (UNSCEAR, 2000).

External hazard index (H_{ex})

The external hazard index (H_{ex}) quantifies the risk of natural gamma radiation (Rafique *et al.*, 2014). This index is used to assess the radiation suitability of various materials and is expressed by the following ratio (Ibrahim, 1999; Al-Hamarneh and Awadallah, 2009):

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1$$

The main purpose of this index is to limit the level of equivalent radiation dose to a permissible value of 1.0 $\text{mSv}\cdot\text{y}^{-1}$ (International..., 1993). The calculated values of the H_{ex} index for river

sediments range from 0.06 to 0.54 (Table 1); the average H_{ex} for these precipitations is 0.13. The average values of H_{ex} rocks extracted from quarries are 0.17 for Quaternary and Middle Carboniferous sediments, 0.16 for kimberlites, and 0.24 for Vendian sediments of the Padun suite, respectively (Table 3). Thus, it can be concluded that, for all studied samples of river sediments and rocks extracted from quarries, the value of the external hazard index (H_{ex}) < 1 .

Uranium isotopes in surface waters

Uranium isotopes are the main radionuclides that determine the radiation quality of surface waters (WHO, 2004; NRB 99/2009; Malov *et al.*, 2014). Due to its ability to form readily soluble compounds, uranium is widely distributed in natural waters (Osmond *et al.*, 1968; Rikhvanov, 2009; Malov, Kiselev, 2008).

The results of the study of the isotopic composition of uranium in the waters of the river Zolotitsa and its tributaries are presented in Table 4, sampling locations are shown in Fig. 1. Three samples were taken from the Zolotitsa River: the first was upstream before flowing into the channel (ZLV-1), the second was taken near the village Tuchkino (ZLV-3) and the third 2.5 km downstream from the mouth of the river Belaya from the river Svetlaya (cipher SVR) and the river Belaya (BLV) was taken in two samples, and from Vakhtovy (VH), Tuchkin (TChV) and Svetly (SV) streams one sample each was taken. In addition, groundwater was tested from a self-draining hydrogeological well on the river Belaya (BLV-1).

As seen from Table 4, the $^{234}\text{U}/^{238}\text{U}$ isotope ratio in surface waters varies from 1.57 to 3.29, and the uranium concentration is from 0.2 to 2.29 $\mu\text{g}/\text{L}$. For groundwater wells on the river Belaya value of $^{234}\text{U}/$

Table 4. The uranium isotope activity in waters of the Zolotitsa River and its tributaries

Sample No	Sampling location	$^{234}\text{U}/^{238}\text{U}$	U, $\mu\text{g}/\text{L}$	α -activity, $\text{Bq}\cdot\text{L}^{-1}$
VH-1	Vahtovij stream	2.23±0.32	0.59±0.07	0.02
TChV-1	Tuchkin stream	2.18±0.30	0.73±0.08	0.04
SV-1	Svetlyj stream	1.83±0.25	0.20±0.02	0.01
SVR-1	Svetlaya River -1	2.45±0.34	0.26±0.03	0.01
SVR-2	Svetlaya River -2	2.93±0.41	0.29±0.03	0.01
BLV-2	Belaya River, upstream of the well	1.61±0.22	0.39±0.04	0.01
BLV-1	Flowing well, bank of Belaya River	1.67±0.19	2.66±0.28	0.09
BLV-3	Belaya River, mouth	1.63±0.20	0.73±0.08	0.02
ZLV-1	Zolotitsa River, upstream of the mining complex	1.57±0.18	0.61±0.07	0.01
ZLV-3	Zolotitsa River, opposite the Tychkino camp	2.66±0.33	1.16±0.12	0.06
ZLV-2	Zolotitsa River, downstream from Belaya River mouth	3.29±0.42	2.29±0.27	0.13

^{238}U is 1.67, the concentration of uranium is $2.66 \mu\text{g/L}$. The average value of $^{234}\text{U}/^{238}\text{U}$ in the water of the Zolotitsa River and its tributaries is 2.24, which is slightly higher than the values of surface runoff characteristic for waters 1.1 - 1.4. The minimum value for the research area is $^{234}\text{U}/^{238}\text{U}$ detected in the Zolotitsa River before flowing into the channel - 1.57, the uranium concentration for this section is $0.61 \mu\text{g/L}$. At the next test spot of the Zolotitsa River in the area of the village Tuchkino value $^{234}\text{U}/^{238}\text{U}$ increases to 2.66, and the concentration of uranium to $1.16 \mu\text{g/L}$. At the third spot of observation, 2.5 km downstream of the Belaya river entry, the value of $^{234}\text{U}/^{238}\text{U}$ increases to 3.29, and the uranium concentration increases to $2.29 \mu\text{g/L}$. Thus, as you move from south to north along the Zolotitsa River isotope ratio $^{234}\text{U}/^{238}\text{U}$ increases more than 2 times, and the uranium concentration increases almost 4 times (Fig. 3).

The observed significant increase in both the value of $^{234}\text{U}/^{238}\text{U}$ and the concentration of uranium in the water of the river Zolotitsa indicates a significant contribution of other sources to the water balance of the river. The main natural sources that contribute to the water balance of Zolotitsa in the studied area are its tributaries of the Svetlaya River, Svetly stream, Vakhtovy stream, Tuchkin stream and the river Belaya. However, in the Svetly stream, the Svetlaya River and the Zolotitsa River upstream of the mining complex the $^{234}\text{U}/^{238}\text{U}$ varies between 1.57 - 2.45, and the uranium concentration is $0.20 - 0.61 \mu\text{g/L}$. Considering several times lower discharge values of the Svetlaya river and the Svetly stream compared to Zolotitsa itself, it is impossible to explain the increase in the water of Zolotitsa in the area of the Tuchkino camp ratio of $^{234}\text{U}/^{238}\text{U}$ to 2.66 and uranium content to $1.16 \mu\text{g/L}$. Obviously, in this segment other sources play a significant role in the

water balance of Zolotitsa River. Most likely, they are drainage waters of water-reducing wells, which are discharged into the channel in the area of the northern boundary of the mining and processing complex. The isotopic ratio in the waters of water-reducing wells varies between 1.63–5.94, and the uranium concentration is $0.13\text{--}18.5 \mu\text{g/L}$ (Malov, 2018₁). With a total discharge of water-reducing wells of $5000 \text{ m}^3/\text{h}$ and a river flow rate of $10,000 \text{ m}^3/\text{h}$ at a low water, the mixing of drainage water with river occurs in a ratio of 1:2 (Malov, 2018₂), and it seems likely that a significant contribution to the Zolotitsa's water balance of drainage water, having relatively high values of $^{234}\text{U}/^{238}\text{U}$ and uranium concentration. On a site from the Tuchkino mining camp to the northernmost point of testing contribution to the water balance of the Zolotitsa River is brought by Vakhtovy and Tuchkin streams, as well as the Belaya River. In it, the uranium isotope ratio varies from 1.61 to 2.23, and the uranium concentration is from 0.59 to $0.73 \mu\text{g/L}$. Given the low values of the uranium-isotopic characteristics in these tributaries, however, to explain such a significant change in the concentration of uranium and the isotope ratio in the water of the river Zolotitsa at the extreme point of observation (ZLV-2) seems to be difficult, since the concentration of uranium compared to the point ZLV-3 in the area of the village Tuchkino increased 2 times, and the ratio of $^{234}\text{U}/^{238}\text{U}$ was almost 1.3 times. Apparently, in the area from the Vakhtoviystream to the Belaya River, groundwater with a high ratio of $^{234}\text{U}/^{238}\text{U}$ and uranium concentration is wedged out directly into the Zolotitsa riverbed. The total alpha activity of surface water samples ranges from 0.01 to $0.13 \text{ Bq}\cdot\text{L}^{-1}$ (Table 3). Despite the noted spatial variations of the $^{234}\text{U}/^{238}\text{U}$ composition in the water of the Zolotitsa River and its tributaries, the total alpha activity in

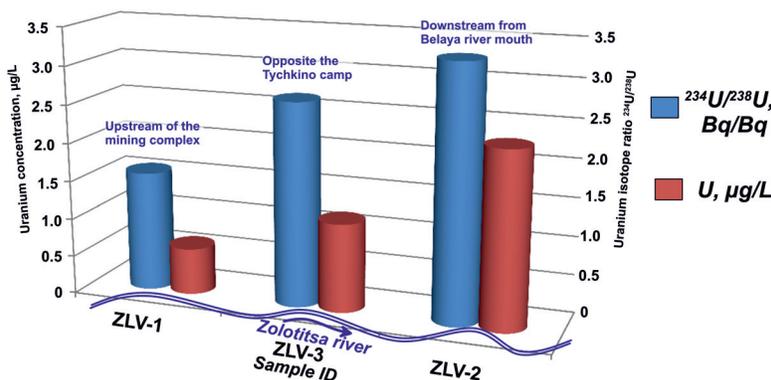


Fig. 3. Changes of uranium isotope ratio $^{234}\text{U}/^{238}\text{U}$ in the water of the Zolotitsa River

water does not exceed the established norms of 0.2 Bq·L⁻¹ for drinking water (NRB 99/2009).

CONCLUSION

Studies of the activity of natural radionuclides ²²⁶Ra, ²³²Th and ⁴⁰K, as well as technogenic ¹³⁷Cs in the bottom sediments of the river Zolotitsa River and its feeders in the area of the developed Lomonosov diamond field. In the developed quarries of the field, samples of kimberlites and rocks were selected on the Arkhangelskaya and Karpinsky-1 pipes to assess the parameters of the radioactivity of the rocks extracted from the quarries. Studies of the isotopic composition of uranium in surface water samples from the Zolotitsa River and its tributaries to assess the radiation quality of river waters. Measurements of the activity of radionuclides in bottom sediments and rocks were carried out by the method of low-background semiconductor gamma spectrometry with an HPGe detector from high-purity germanium, the study of uranium isotopes ²³⁴U, ²³⁸U in water was carried out by alpha spectrometry. The average activity concentrations of ¹³⁷Cs, ²²⁶Ra, ²³²Th, and ⁴⁰K were 5.4, 9.0, 11.3, 321.6 Bq×kg⁻¹. The influence of mining and beneficiation complex on the increase in the activity of radionuclides in the bottom sediments of the Zolotitsa River was evidenced. The maximum average values of radionuclides are confined to the area of the river Zolotitsa River, in the region of which drainage water is unloaded from the swamp-filtering fields into the riverbed.

The radiation hazard parameters of bottom sediments and rocks from quarries were calculated. The average absorbed dose rate of gamma radiation D_R of river sediments was 24.4 nGy·h⁻¹. Radium equivalent R_{aeq} for bottom sediments averages 47.3 Bq·kg⁻¹. R_{aeq} and D_R are slightly higher for rocks from quarries than for those in bottom sediments. The maximum average values of R_{aeq} and D_R are characteristic of sandy-clay Vendian deposits of the Padun suite (V_2) and are 87.2 Bq·kg⁻¹ and 41.5 nGy·h⁻¹, respectively. The calculated external hazard index H_{ex} for river sediments and rocks extracted from quarries does not exceed 1. This indicates that the dose rate is at a level below the permissible value of 1 mSv·y⁻¹ (ICRP, 1993). The total alpha activity in water does not exceed the established values of 0.2 Bq·l⁻¹ for drinking water (NRB 99/2009). The values of the radiation parameters are on average below the world average and do not pose a significant

danger to the personnel of mining and beneficiation complex and the population of the mining camp. We can conclude that at the moment in the field area there is a relatively low level of natural radioactivity. However, in the future, with the deepening of quarries and an increase in diamond ore production, an increase in the volume of pumping and mineralization of drainage water, and with an even greater decrease in the sorption properties of the peatbog-filtration fields, an increase in radioactivity in the bottom sediments of the Zolotitsa River should be expected.

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