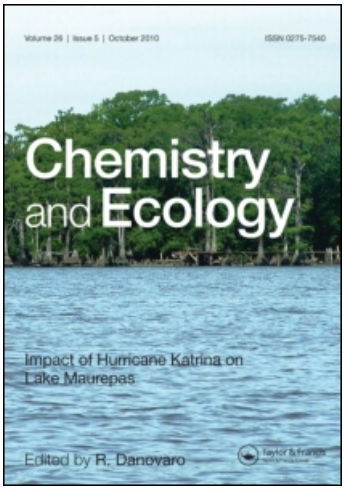

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Alexander Bolsunovsky[a]
[a] Institute of Biophysics of the Siberian Branch of Russian Academy of Sciences, Krasnoyarsk, Russia

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Artificial radionuclides in sediment of the Yenisei River

Alexander Bolsunovsky*

Institute of Biophysics of the Siberian Branch of Russian Academy of Sciences, Krasnoyarsk, Russia

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Releases from the nuclear facility Mining-and-Chemical Combine (MCC) located at Zheleznogorsk have contributed to the radionuclide contamination of the Yenisei River since operations commenced in 1958. The aim of this study was to assess the activity concentrations of artificial radionuclides and the strength of their binding in Yenisei River sediments. Investigation of Yenisei River sediment samples revealed the presence of artificial radionuclides typical of the MCC radioactive discharge: namely, isotopes of europium, caesium, ^{60}Co and transuranium elements. The concentrations of artificial radionuclides in the sediment layers remain relatively high as far as 200 km downstream of the MCC. In sediment cores collected upstream of the MCC, γ-spectrometric measurements registered only one artificial radionuclide, ^{137}Cs, with a maximal activity of 8 Bq·kg^{-1}. Sequential extraction performed on samples of the upper layers of the sediment core showed different degrees of potential environmental availability for artificial radionuclides: the highest was recorded for ^{241}Am and ^{152}Eu (up to 85% of initial activity), followed by ^{60}Co (up to 32%), and finally , ^{137}Cs (up to 15%). In a few samples, ^{241}Am was present in the unextractable form, which may be accounted for by the presence of reactor fuel microparticles.

Keywords: river sediments; artificial radionuclides; sequential extraction; Yenisei River

1. Introduction

The Yenisei is one of the world's largest rivers, 4100 km long, flowing into the Kara Sea. The Mining-and-Chemical Combine (MCC) at Zheleznogorsk is situated on the east bank of the Yenisei River, 60 km downstream of the city of Krasnoyarsk. The MCC has been producing weapons-grade plutonium in uranium–graphite reactors since 1958, when the first reactor was started up. Irradiated uranium is reprocessed at the radiochemical plant to separate uranium, plutonium and fission products. The reactor plant houses three reactors. Two of the reactors used Yenisei River water as a coolant, i.e. water was taken from the river to remove heat from the core, passed through the reactor fuel channels and then returned to the Yenisei. Both of these reactors were shut down in 1992, but the third reactor had been working until April 2010, when it was taken out of service. This reactor also used Yenisei water as a coolant for some channels and released radionuclides of activation origin into the river. Several scientific expeditions have revealed that the Yenisei River floodplain is contaminated with artificial radionuclides, including plutonium isotopes, for 2000 km downstream of the MCC [1–7]. Previously, levels

*Email: radecol@ibp.ru

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of artificial radionuclides have been mainly measured in samples of floodplain soils; only a few measurements have been carried out in sediments [3,5–8]. To study the migration of artificial radionuclides and their bioavailability, one needs know the physicochemical forms of radionuclides in sediments.

The purpose of this study was to assess the activities of artificial radionuclides and the strength of their binding in Yenisei River sediments near the MCC and at a considerable distance downstream from it.

2. Materials and methods

2.1. *Sample collection and preparation*

During the expeditions of 1997–2007, sediment samples were collected from the Yenisei River at different distances downstream of the MCC. Sediments were collected in two main areas: near the MCC (site E15, the village of B. Balchug) and at a considerable distance downstream (site E18, the village of Zakharovka) (Figure 1). The village of B. Balchug is situated ∼15 km downstream from the point of discharge from the MCC. The village of Zakharovka is situated 200 km downstream of the discharge site. For comparison purposes, we also collected sediment cores at a position upstream of the MCC, near the village of Esaulovo (site E5, 45 km downstream of Krasnoyarsk) (Figure 1). Sediments were collected from river branches with not more than 1 m of water above the sediments. The diameter of the sampler (made in Russia) was 11 cm and it was able to collect cores up to 1.5 m long. Each sediment sample was a 3 cm thick layer, except for the top sediment layer, whose thickness ranged from 5 to 10 cm, depending on the core moisture content. In the laboratory, some sediment samples were dried, but several samples were kept wet and subjected to sequential extraction.

2.2. *Radioactivity measurements*

Measurements of activity concentrations of γ-emitting nuclides (including ^{241}Am) were conducted at the Institute of Biophysics SB RAS using a Canberra γ-spectrometer (USA) coupled to a GX2320 23% hyperpure germanium (HPGe) coaxial detector. Gamma-spectra were processed using Canberra Genie-2000 software (USA). The activity concentrations of radionuclides in sediment layers were decay-corrected to the sample collection dates.

2.3. *Sediment chemical analysis*

Investigations and development of the X-ray fluorescence technique to determine the elemental composition of sediment were carried out on a VRA-20R Carl Zeiss fluorescent X-ray analyser (Jena, Germany) at the Institute of Geology and Mineralogy in Novosibirsk (Russia). The detection limits for the majority of sediment-forming components were 0.02–0.005%; MgO and Na_2O had detection limits of 0.05 and 0.1%, respectively. Results are reported in Table 1. As expected, river bottom sediments contained a large amount of SiO_2 (56–68 mass%). There was also a considerable percentage of Al_2O_3 (12.4–13.1 mass%). The percentage of Fe_2O_3 varied from 4.6 to 5.3 mass% and that of MnO constituted 0.1–0.2 mass%, as typical of freshwater sediments. Percentages of Fe and Mn in sediment samples and their vertical distribution in sediments are commonly accepted indicators of the direction and features of geochemical processes. The samples also contained MgO, CaO, Na_2O, K_2O and TiO_2, which did not exceed 2–3 mass%.

Figure 1. Diagrammatic map of the south of the Krasnoyarsk Territory (Russia), showing settlements near to which samples of sediments were collected. Scale 1 : 2,800,000.

2.4. *Sequential extraction technique*

Binding of artificial radionuclides to sediment samples was examined using a sequential extraction technique proposed by Tessier et al. [9] and modified by Klemt and colleagues [7] (Table 2). The modified technique included an additional extraction step (amorphous silicates). Sequential

Table 1. Results of X-ray phase analysis of sediments (mass%).

	Mean	Min	Max
SiO_2	63	56	68
TiO_2	0.76	0.7	0.8
Al_2O_3	12.8	12.4	13.1
Fe_2O_3	4.9	4.6	5.3
MnO	0.14	0.1	0.2
MgO	2.1	1.9	2.3
CaO	2.9	2.8	3.1
Na_2O	2.5	2.0	2.9
K_2O	1.9	1.7	2.0

Table 2. Sequential extraction scheme for sediment samples.

Fractions	Chemical reagent	Treatment	Phases with target ions
I	CH_3COONH_4 (1 mol·L^{-1})	24 h shaking	Exchangeable ions
II	CH_3COONH_4 (1 mol·L^{-1}) + HNO_3 (1 mol·L^{-1}) to pH 5	shaking to equilibrium	Carbonates
III	NH_2OH-HCl (0.2 mol·L^{-1}) in CH_3COOH (25%)	3 h shaking	Oxides and hydroxides of iron and manganese
IV	H_2O_2 (35%) + HNO_3 (1 mol·L^{-1}) up to 0.05 mol·L^{-1}	3 h stirring at 85 °C	Organic matter
V	NaOH (0.2 mol·L^{-1})	40 min stirring at 80 °C	Amorphous silicates
VI			Residuals

extraction was performed in fresh samples of the upper layers of sediment cores No. 1 and No. 2, collected near the village of B. Balchug (site E15).

3. Results and discussion

3.1. *The distribution of activities of artificial radionuclides in sediments of the Yenisei River*

A large number of sediment cores were collected during expeditions to the Yenisei River. In sediment layers, γ-spectrometric measurements registered artificial radionuclides typical of the MCC radioactive discharge: isotopes of europium (^{152}Eu, ^{154}Eu and ^{155}Eu), caesium (^{137}Cs and ^{134}Cs), ^{60}Co and the transuranium element ^{241}Am. Special consideration was given to the vertical distribution of artificial radionuclides with long half-lives, such as ^{241}Am (half-life 432.2 years), ^{137}Cs (half-life 30.1 years), ^{152}Eu (half-life 13.5 years) and ^{60}Co (half-life 5.3 years). The occurrence of ^{241}Am, a daughter of ^{241}Pu, can be indicative of the presence of other transuranium elements in the sediment samples. As noted above, sediment cores were collected in the two main areas: near the MCC (site E15, near the village of B. Balchug, 96 km downstream of Krasnoyarsk) and at a considerable distance downstream (site E18, near the village of Zakharovka, 280 km downstream of Krasnoyarsk). The vertical distribution of radionuclides in the sediment cores is complex and there are several concentration minima and maxima due to different amounts of radionuclides released by the MCC and to variations in global fallout. An example of the distribution of the principal artificial radionuclides in layers of sediment cores collected near the MCC at the E15 position is shown in Figure 2. Maxima for ^{137}Cs activity concentrations (1800 Bq·kg^{-1}) were registered in the middle part of the E15 core (the 13–15 cm layer) and in its lower part (25–28 cm) (Figure 2). The mean ^{137}Cs concentrations in the core layers ranged between 800 and 1000 Bq·kg^{-1}. Gamma-spectrometric analysis also registered ^{241}Am concentrations in sediment layers of the E15 position. Although in the majority of the sediment layers of the E15 position the

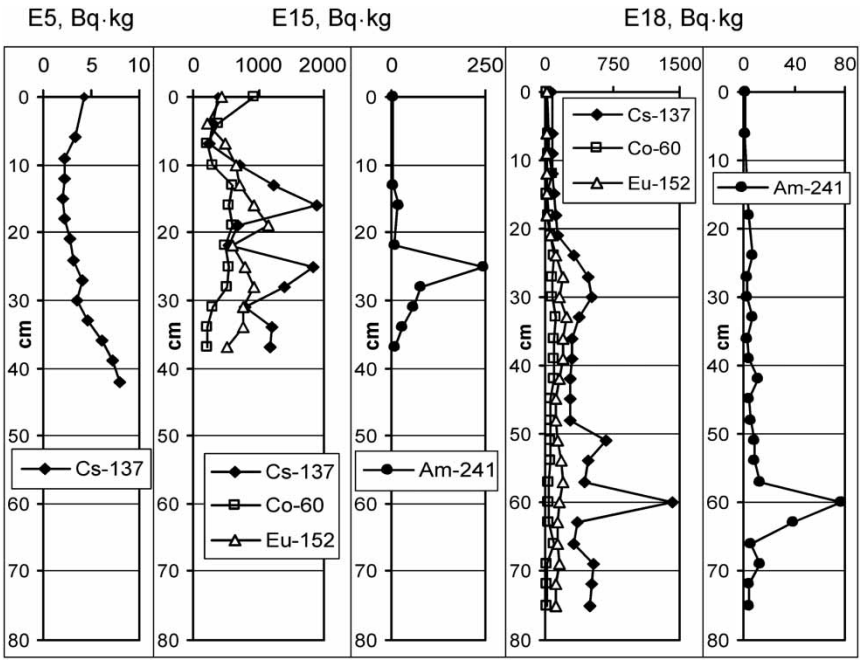

Figure 2. Vertical distribution of radionuclides in sediments (dry mass) of the Yenisei River near the villages of Esaulovo (E5), B. Balchug (E15) and Zakharovka (E18).

^{241}Am concentration did not exceed 30–60 Bq·kg^{-1}, in the 25–28 cm layer ^{241}Am concentrations reached 240 Bq·kg^{-1}. One of the explanations for these abnormally high concentrations of ^{241}Am may be the presence of microparticles. Sukhorukov and co-workers [4,10] reported that bottom sediments and floodplain soils of the Yenisei River were found to contain radioactive particles of reactor origin, including ones with high ^{241}Am activity concentrations. For more details see Section 3.2.4.

We also collected sediment cores at the village of Zakharovka, 200 km downstream of the MCC – at site E18. The E18 cores were twice as long as the E15 cores. The distribution of the principal radionuclides in the layers of a sediment core collected at E18 is also shown in Figure 2. The maximum of ^{137}Cs activity concentration (1400 Bq·kg^{-1}) is registered in the lower part (60–63 cm) (Figure 2). The mean ^{137}Cs concentrations in the layers of the E18 core ranged between 200 and 500 Bq·kg^{-1}, i.e. values were lower than the mean values for the E15 core. Gamma-spectrometric analysis of the E18 core samples registered ^{241}Am (77 Bq·kg^{-1}) in the layers containing maximal levels of ^{137}Cs activity, similarly to E15 cores. Summing up the results obtained for sediment cores collected at the village of Zakharovka, we should note that the maximal ^{137}Cs and ^{241}Am concentrations in samples of these sediment cores were similar to activity concentrations of these radionuclides in sediments collected near the MCC discharge site, e.g. near B. Balchug (the E15 core). Thus, concentrations of radionuclides in layers of sediments collected far downstream of the MCC remain relatively high. That some sediment layers contained abnormally high ^{241}Am concentrations may be indicative of abnormally high concentrations of other transuranium elements. Our subsequent study focused on radiochemical investigations of sediment layers that, as registered by γ-spectrometry, contained maximal levels of radionuclides, including ^{241}Am. Radiochemical investigations of sediment samples collected from the Yenisei River [3–6] showed that levels of transuranium elements (^{238}Pu, ^{239,240}Pu and ^{241}Am) were at least 10 times higher than reported earlier [1] and at least 100 times higher than global fallout levels. Results of our previous measurements of transuranium elements such as ^{241}Pu and ^{237}Np in sediments showed

that their levels remained high even at a distance of 200 km downstream of the MCC [3,5]. The presence of sediment layers containing abnormally high actinide and ^{137}Cs levels at different distances downstream of the MCC [3–6] may be indicative of emergency discharges from the MCC in the past. Further investigations involving the dating of different layers of Yenisei sediments can be useful for determining the chronology of the introduction of radionuclides into sediments.

As mentioned above, we also collected sediment cores at a position upstream of the MCC, near the village of Esaulovo (45 km downstream of Krasnoyarsk), site E5. The purpose of collecting sediment cores at this position was to determine the global level of radioactive contamination of the Yenisei sediments by ^{137}Cs. In layers of these sediments, γ-spectrometric measurements recorded only one artificial radionuclide that is also present in the MCC radioactive discharge – ^{137}Cs (Figure 2). In the upper layers of sediment cores collected at Esaulovo (0–6 cm), the highest ^{137}Cs concentration amounted to 4.3 ± 0.7 Bq·kg^{-1} dry mass. With depth, the amounts of ^{137}Cs in the samples decreased more than twofold. However, from the middle layers downwards, levels of ^{137}Cs increased, and the maximal activity was registered in the lowest layers, 39–45 cm (Figure 2) – 7.9 ± 0.6 Bq·kg^{-1} dry mass. This range of ^{137}Cs (4–8 Bq·kg^{-1} dry mass) can be accepted as the background value of radioactive contamination of sediments in this part of the Yenisei, due to global fallouts of ^{137}Cs. It is quite clear that the profile of the core is incomplete, and we did not reach sediment layers containing no ^{137}Cs, so the range also needs to be viewed with caution. In our further studies, we should collect complete cores and give a more accurate estimate of the background range of ^{137}Cs contamination.

3.2. *Radionuclide speciation in sediments of the Yenisei River*

Sequential extraction techniques are the most common method of studying radionuclide speciation in soils and sediments. Sequential extraction was performed on samples of the upper layers of sediment cores No. 1 and No. 2, collected near the village of B. Balchug (E15). Results of sequential extraction are listed in Table 3 and shown in Figure 3. Below is a detailed description of the distribution of radionuclides among chemical fractions of the samples from the two sediment cores.

3.2.1. *Fractionation of ^{137}Cs*

Although core No. 2 contained half as much ^{137}Cs activity as core No. 1, the percentages of the extracted radionuclide were similar (13–15%). The highest percentage of ^{137}Cs was extracted from

Table 3. Sequential extraction results of artificial radionuclides in the uppermost layers of sediment samples from cores No. 1 and No. 2 collected at the village of B. Balchug (E15).

	^{60}Co		^{137}Cs		^{152}Eu		^{241}Am	
Fractions	No. 1	No. 2	No. 1	No. 2	No. 1	No. 2	No. 1	No. 2
I (%)	2.2 ± 0.1	<MDA[a]	3.8 ± 0.3	2.5 ± 0.3	<MDA	<MDA	<MDA	<MDA
II (%)	5.8 ± 0.3	1.2 ± 0.2	1.9 ± 0.2	1.6 ± 0.2	7 ± 0.3	4 ± 0.3	27 ± 4	<MDA
III (%)	6 ± 0.3	1.8 ± 0.2	1 ± 0.2	0.8 ± 0.1	6 ± 0.3	26 ± 2	12 ± 2	<MDA
IV (%)	18 ± 1	4 ± 0.3	8.6 ± 0.8	7.4 ± 0.6	68 ± 3	52 ± 3	46 ± 6	<MDA
V (%)	<MDA	<MDA	0.7 ± 0.1	0.7 ± 0.1	<MDA	1 ± 0.2	<MDA	<MDA
VI – mineral residue (%)	68 ± 4	93 ± 7	84 ± 7	87 ± 8	19 ± 1	17 ± 1	15 ± 2	100
Initial radionuclide activity in the sample, Bq·kg^{-1} FW (100%)	168 ± 19	486 ± 12	486 ± 44	168 ± 10	314 ± 13	683 ± 13	16 ± 3	5.0 ± 1.5

Note: [a]MDA, minimum detectable activity.

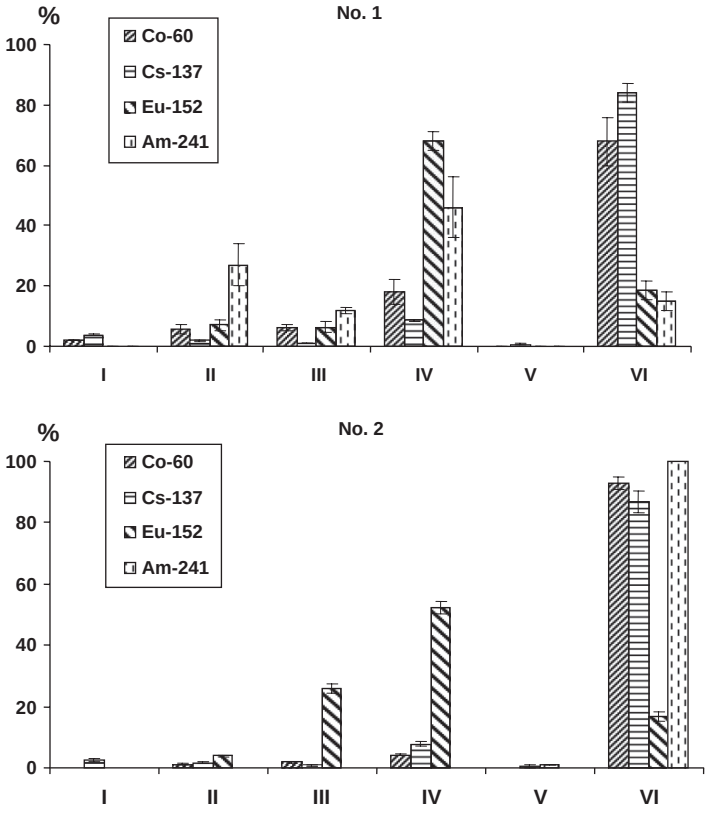

Figure 3. The distribution of radionuclides among chemical fractions (I–VI) of the uppermost layers of sediment cores No. 1 and No. 2 collected at the village of B. Balchug (E15).

organic matter (Fraction IV) – 8%, up to 3.8% was extracted from exchangeable ions (Fraction I), and up to 1.9% from sesquioxides and hydroxides (Fraction III). The lowest ^{137}Cs content was recorded in Fractions II and V, ∼1%. The results of sequential extraction of Yenisei sediments reported by Klemt [7] show that just 4% of ^{137}Cs was extractable and the highest percentage was extracted from Fraction IV and the lowest from Fraction III. This discrepancy between the data reported by Klemt and our results (4 vs. 13–15%) can be accounted for by the fact that the cores subjected to analysis had been collected from sites located at different distances downstream of the MCC. We analysed the core from site E15, located 15 km downstream of the MCC, whereas Klemt's core was collected from a site located ∼160 km downstream of the MCC. As reported by Todorov and co-workers [11], analysis of soils by six different sequential extraction techniques can yield the range of extracted ^{137}Cs from 0.8 to 13.8%, and the highest percentage of ^{137}Cs is bonded to organic matter.

3.2.2. *Fractionation of ^{60}Co*

The percentages of ^{60}Co extracted from sediment cores No. 1 and No. 2 differed considerably: 7% from core No. 2, which, taking into account the error, was similar to the percentage of extracted ^{137}Cs, and several times higher from core No. 1 – 32% of total ^{60}Co. The highest percentage of ^{60}Co (similarly to ^{137}Cs) was extracted from Fraction IV – up to 18%, and up to 6% was extracted from Fractions II and III. In Fraction V, ^{60}Co content was below the detection limit of the method. Results of sequential extraction of Yenisei sediments reported by Klemt [7] show that at least 20%

of ^{60}Co was extractable and the highest percentage was extracted from Fraction IV and the lowest from Fraction I. Thus, the data reported by Klemt are in good agreement with our results on the distribution of ^{60}Co in samples of core No. 1. Todorov and co-workers [11] report even higher percentages of extractable ^{60}Co (42–78%), which vary depending on the sequential extraction technique used to analyse soils.

3.2.3. *Fractionation of ^{152}Eu*

Similarly to the situation for ^{137}Cs, the difference in activity concentrations of ^{152}Eu in cores No. 1 and No. 2 did not affect the percentages of the extracted ^{152}Eu, which amounted to 83–85%. The highest percentage of ^{152}Eu (similarly to ^{137}Cs and ^{60}Co) was extracted from Fraction IV – up to 68%, and up to 26% was extracted from Fractions II and III. In Fractions I and V, ^{152}Eu content was below the detection limit of the method. Results of sequential extraction of Yenisei sediments reported by Klemt [7] show that ∼60% of ^{152}Eu was extractable and the highest percentage was extracted from Fraction IV and the lowest from Fractions I and II. Thus, the data reported by Klemt and our results on the distribution of ^{152}Eu in samples of cores No. 1 and No. 2 are similar, allowing for the difference in the distances from the MCC. No other data on extraction of ^{152}Eu from sediment and soil samples can be found in the available literature.

3.2.4. *Fractionation of ^{241}Am*

Results given in Table 3 and Figure 3 suggest considerable differences in the distribution of ^{241}Am among fractions in cores No. 1 and No. 2.

The distribution of ^{241}Am among fractions in samples of core No. 1 is similar to that of ^{152}Eu: 85% of ^{241}Am was extracted; the highest percentage of ^{241}Am was extracted from Fraction IV – up to 46%, and up to 27% was extracted from Fractions II and III. In Fractions I and V, ^{241}Am content (similarly to ^{152}Eu) was below the detection limit of the method. For core No. 2, ^{241}Am content in all fractions (I, II, III, IV, and V) was below the detection limit of the method, and it was only reliably recorded in the residual solid (Fraction VI). This suggests that in this sediment sample, ^{241}Am was present in the unextractable form. Klemt [7] reported similar percentages of extracted ^{241}Am and ^{152}Eu. Skipperud and co-workers [8] studied the distribution of ^{239,240}Pu isotopes among chemical fractions of sediments collected from different sections of the Yenisei River, at increasing distances downstream of the MCC. They found that 70–80% of the ^{239,240}Pu was released in the H_2O_2fraction, i.e. Fraction IV in our study. Thus, our results on the distribution of ^{241}Am in sediment samples of core No. 1 are in good agreement with the literature data on the distribution of ^{241}Am and ^{239,240}Pu in sediment samples from the Yenisei River [7,8]. The unusual distribution of ^{241}Am in samples of core No. 2 may be accounted for by the presence of microparticles of the reactor fuel. Sukhorukov and co-workers [4,10] reported that sediments and floodplain soils of the Yenisei River contained radioactive particles of reactor origin, including ones with high ^{241}Am concentrations. These particles, similarly to radioactive particles from the Chernobyl accident, have a uranium–graphite matrix and a very long dissolution time (several decades).

It is well-known that radionuclides in the first two fractions (Exchangeable and Carbonates) of soil (sediments) are all potentially available to plants and mobile in the environment [12]. Radionuclides in Fractions III and IV (Fe–Mn Oxides and Organic Matter) can also be potentially bioavailable as under certain conditions, Fe–Mn oxides can be unstable, and organic matter can be degraded [9,13]. Thus, assessment of potential environmental availability of radionuclides can be based on the data for the first four fractions. In the analysed sediments from the Yenisei River, ^{241}Am and ^{152}Eu exhibited the highest potential environmental availability – up to 85% of the initial activity, then followed ^{60}Co (up to 32%), and, last, ^{137}Cs (up to 15%). Different sediment

properties or forms in which radionuclides are introduced into the freshwater environment can affect their degrees of mobility.

4. Conclusions

Investigations of the Yenisei River sediment samples revealed the presence of artificial radionuclides typical of the MCC radioactive discharge: isotopes of europium (^{152}Eu, ^{154}Eu and ^{155}Eu), caesium (^{137}Cs and ^{134}Cs), ^{60}Co and transuranium elements. Concentrations of artificial radionuclides in sediment layers remain relatively high as far as 200 km downstream of the MCC. In layers of sediment cores collected at a position upstream of the MCC, the range of ^{137}Cs (4–8 Bq·kg^{-1} dry mass) can be accepted as the background value of radioactive contamination of sediments in this section of the Yenisei, due to global fallouts of ^{137}Cs.

Assessment of the potential environmental availability of artificial radionuclides in the analysed sediments from the Yenisei River was based on the data for the first four fractions (Exchangeable, Carbonates, Fe–Mn Oxides and Organic Matter) of sequential extraction. Radionuclides differed in their degree of potential environmental availability in the analysed sediments: the highest was recorded for ^{241}Am and ^{152}Eu (up to 85% of the initial activity), followed ^{60}Co (up to 32%), and finally, ^{137}Cs (up to 15%). In a few samples, ^{241}Am was present in the unextractable form (Residuals), which may be accounted for by the presence of microparticles of the reactor fuel.

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