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SOIL POLLUTION AND REMEDIATION

Distribution of artificial radioisotopes in granulometric and organic fractions of alluvial soils downstream from the Krasnoyarsk Mining and Chemical Combine (KMCC), Russia

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Abstract
Purpose The purpose of this study was to compare the distribution of the most significant medium and long-lived radioisotopes, i.e., ^{60}Co, ^{137}Cs, and ^{152}Eu, in granulometric and organic fractions of alluvial soils downstream from the Krasnoyarsk Mining and Chemical Combine (KMCC), Russia, to reveal natural patterns of their behavior and accumulation.
Materials and methods Soil samples collected at different elevations and depths in a floodplain of the Yenisey River downstream from the KMCC (20–250 km) were subjected to granulometric analysis by dry screening and a modified Petelin method. Fractions <0.05 mm were collected by a pipette method. Radionuclide activity in the different soil layers and in their fractions was determined using a spectrometer equipped with an HPGe detector. Concentration of total C (Ctot) and C of carbonates (Ccarb) was determined using a CHN analyzer before and after elimination of carbonates, organic C (Corg) being calculated as the difference between the obtained values. Organic fractions were separated by saturation of the air-dry sample with 0.1 M NaOH and further precipitation of humic acid from filtrate by 1 M HCl at pH 1. The separation resulted in three fractions of the fulvic acids, humic acids, and the residue containing the denuded mineral phase and the refractory organic residue. The selected bulk samples and fractions were analyzed for radionuclide activity.
Results and discussion Based on earlier results, the distribution of the pelite (<0.01 mm) and aleurite (0.01–0.1 mm) fractions in alluvium and soil samples have been analyzed to evaluate the grain-size contribution to radionuclide fixation. A positive correlation between radionuclide activity and the portion of pelite fraction was established for ^{60}Co and ^{152}Eu, while ^{137}Cs accumulation was not related with this fraction. In organic matter (OM) extracts, more than 90 % of ^{137}Cs, at proportions similar to those attributable to ^{238}Th and ^{40}K, were associated with the residue fraction, while 72 % of ^{152}Eu and 46 % of ^{60}Co were found in the mobile fraction of the low molecular fulvic acids. In successive layers of the soil vertical profile, approximately 94 % of the ^{152}Eu variation may be explained by a linear model with Corg and Ccarb values as independent variables.
Conclusions Different associations of ^{137}Cs, ^{60}Co, and ^{152} Eu with particulate and organic fractions in river sediments and floodplain soils could be explained by the dominating discharge form (water soluble or particulate), affinity to organic substances of different mobility, sorption by minerals and their aggregates, and chemisorption.

Keywords Alluvial soils · Organic fractions · Particle distribution · Radionuclides · Yenisey floodplain

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1 Introduction

Operation of the Krasnoyarsk Mining and Chemical Combine (KMCC), located approximately 40 km downstream from the city of Krasnoyarsk (Russia), near the Yenisey River, started in August 1958 and continued until 1992. One of its functions

was the production of weapon-grade Pu (Sukhorukov et al. 2004). Discharges of radioactive wastes from the KMCC to the river channel have led to artificial radionuclide contamination of river water and sediments. However, no data on parameters related to the actual discharges were available for the early years of operation.

The most complete dataset on KMCC discharges covering the period from 1987 to 2000 was published by Vakulovsky et al. 2006. According to these data, radionuclides released to the river can be arranged in order of descending activity as ^{60}Co—671 GBq>^{137}Cs—316 GBq>^{152}Eu—262 GBq. The mean annual discharge of the aforementioned radioisotopes was 111.8, 52.7, and 43.7 GBq, respectively (Table 1).

In 1992, after the shutdown of two reactors, the radioisotope inflow to the river channel decreased by an order of magnitude and the total activity of ^{60}Co, ^{137}Cs, and ^{152}Eu released to the Yenisey River in the period from 1993 to 2000 was about 41.8, 29.1, and 5.4 GBq, respectively. There was a significantly greater decrease in ^{60}Co, and especially ^{152}Eu, contributions to the total radionuclide contamination in the late period (from 21 to 7 % of the total activity for ^{152}Eu).

Traces of various medium-lived radionuclides, such as ^{137}Cs, ^{60}Co, ^{152} Eu, and ^{154}Eu with half-lives of 30.2, 5.3, 13.3, and 8.6 years, respectively, have been registered in alluvial soils even at the beginning of this century (2000 onwards). These radionuclides have been measured at distances of up to 2000 km from the discharge point in the lower reaches of the Yenisey flowing to the Arctic Ocean (Linnik et al. 2013).

Detailed radioecological studies of the Yenisey focusing on the contamination of water and sediments started only in the 1990s and were most frequent in the last decade (Sukhorukov et al. 2000; Bondareva and Bolsunovskii 2008; Bondareva and Artamonova 2011; Sukhorukov et al. 2004, 2006; Linnik et al. 2005, 2006; Vakulovsky et al. 2006, 2014; Bolsunovsky 2011; Kropatcheva et al. 2012; Korobova et al. 2014). The main practical goal of these studies was to evaluate the level and extent of contamination and its radioecological significance. Three radionuclides, i.e., ^{137}Cs, ^{60}Co, and ^{152} Eu, are of the utmost interest nowadays due to their radioecological importance.

Cs-137 pathways and radioecological effects have been studied intensively worldwide reflecting the radioisotope's wide distribution and its comparative ease of determination compared to other radionuclides. Its migration in the basins of the Yenisey River and those of the Dnieper and its tributaries, in particular, was investigated in detail after the Chernobyl accident (Vakulovsky et al. 1994; Sansone et al. 1996).

Table 1 Evaluation of annual and total discharges of ^{60}Co, ^{137}Cs, and ^{152}Eu by the KMCC in 1987–1992 (GBq, Vakulovsky et al. 2006)

Radionuclide	Mean annual (1987–1992)	Total (1987–1992)	Mean annual (1993–2000)	Total (1993–2000)
^{60}Co	111.8	671	5.2	41.8
^{137}Cs	52.7	316	3.64	29.1
^{152}Eu	43.7	262	0.68	5.4

Cobalt-60, a radioisotope which is almost always present in the releases of the enterprises producing weapon-grade Pu and other radiologically hazardous materials, may significantly contribute to doses to human populations and biota, over considerable time periods. For Pu, this may reflect its practically negligible vertical migration in mineral sediments ($(1–4)\times 10^{-12}$ m^2 s^{-1}) in the Yenisey coastal floodplain deposits (Nosov 1997).

Cobalt forms highly stable complexes with organic ligands, especially humic and fulvic acids (Glaus et al. 2000). Therefore, soils, or horizons that are rich in organic matter (OM), are able to bind Co more strongly and for longer periods. However, the portion associated with mobile fulvic acids is known to enhance its mobility.

Compared to ^{137}Cs and ^{60}Co, the distribution of ^{152,154,155}Eu has been investigated to a narrower extent and in less detail. The Yenisey river valley downstream from KMCC seems to remain the main site in Russia where Eu migration has been studied because of the considerable levels of contamination associated with this radioisotope in this area. In 1993–2000, the specific activity of ^{152}Eu in Yenisey bottom sediments in the near impact zone attained levels of 2000 Bq kg^{-1} and the contamination density of alluvium soils due to this radionuclide attained levels, on several islands, of up to 2.6×10^6 Bq m^{-2}. Both ^{152}Eu and ^{137}Cs isotopes were measured 1000 km downstream from their discharge point (Nossov et al. 2002). Mechanisms of ^{152,154,155}Eu migration in the contaminated alluvium are still poorly studied. Vakulovsky et al. (2006) found that in October 5th, 2001, when only 1 out of the 3 reactors initially constructed to produce weapon-grade Pu was still operating, the major portion (61 %) of the total ^{152}Eu discharge (230 Bq m^{-3}) was water soluble and about 39 % was associated with a suspended load.

It has been previously shown that studies of technogenic radionuclide distributions in different granulometric fractions of alluvial soils are important in enabling the reliable reconstruction of the radiation situation for particular landscape-geochemical conditions following serious radiation accidents, such as Chernobyl (Korobova et al. 2008). This is also true for cases of technogenic contamination arising from more routine releases, such as those that occurred in the Yenisey basin (Linnik et al. 2014).

The purpose of this study was to analyze the distribution of ^{60}Co, ^{137}Cs, and ^{152}Eu in alluvial soils downstream from the KMCC and to compare these radionuclides in terms of their ability to associate with different granulometric and organic fractions. The study is based on the data (published and

unpublished) of the international project STREAM (Brown et al. 2002; Linnik et al. 2005, 2006).

2 Study area and methods

The study was carried out over several islands and the coastal plain of the Yenisey River, 20 to 250 km downstream from the KMCC (Fig. 1). Regarding radionuclide decay, data concerning radioactive contamination that occurred mainly from the 1960s to the 1980s can now be obtained only by investigation of soil samples and fractions containing measureable quantities of radionuclides. In this publication, we analyze the data obtained from nine soil profiles located at different distances from the KMCC (Fig. 1). The sampling sites were selected at different elevations on the floodplain after a preliminary field radiometric survey indicating areas of maximum radionuclide contamination (Linnik et al. 2005, 2006). Samples were collected to a depth of 40–50 cm and divided into different increments according to the soil master horizons and profile lithology. The samples were air dried and stored under cool conditions.

Radionuclide activity in different soil horizons and in their fractions was determined using a Canberra spectrometer equipped with an HPGe detector. The minimum measured activity for ^{137}Cs was less than 0.1 Bq per sample within a counting period of 3600 s. Granulometric analysis was performed for 13 samples with maximum specific activity for ^{137}Cs, ^{60}Co, and ^{152}Eu. In the zone nearer to the KMCC (the Beryozovy Island and B. Balchug), the set of samples included the following profiles and depth horizons (cm): B2-14 (0–5), BP4-1 (8–12), and BP4-45 (5–8). Tolsty Island is represented by samples TOP1-44 (5–10; 35–40); TOP1-48 (4–8; 28–33); the Kazachinskoye site—by KP1-11 (0–5), KP1-28 (5–10); and Ust-Tungusky Island—by UTP-8 (2–12; 19–23), UTP-10 (0–2; 15–20). The resulting numbers of sites with an analytically detectable amount of ^{137}Cs, ^{60}Co, and ^{152}Eu in the different fractions were 13, 9, and 8, respectively.

Granulometric techniques included dry screening and a modified Petelin method (Sval'nov and Alekseeva 2005) with a 1-min ultrasonic pre-treatment by Fritsh Laborette (Germany). Fractions <0.05 mm were collected utilizing a pipette method. The initial weights of the samples were approximately 30 g. Fractionation allowed separation of the following particle size ranges (mm): 1.0–0.5, 0.5–0.25, 0.25–

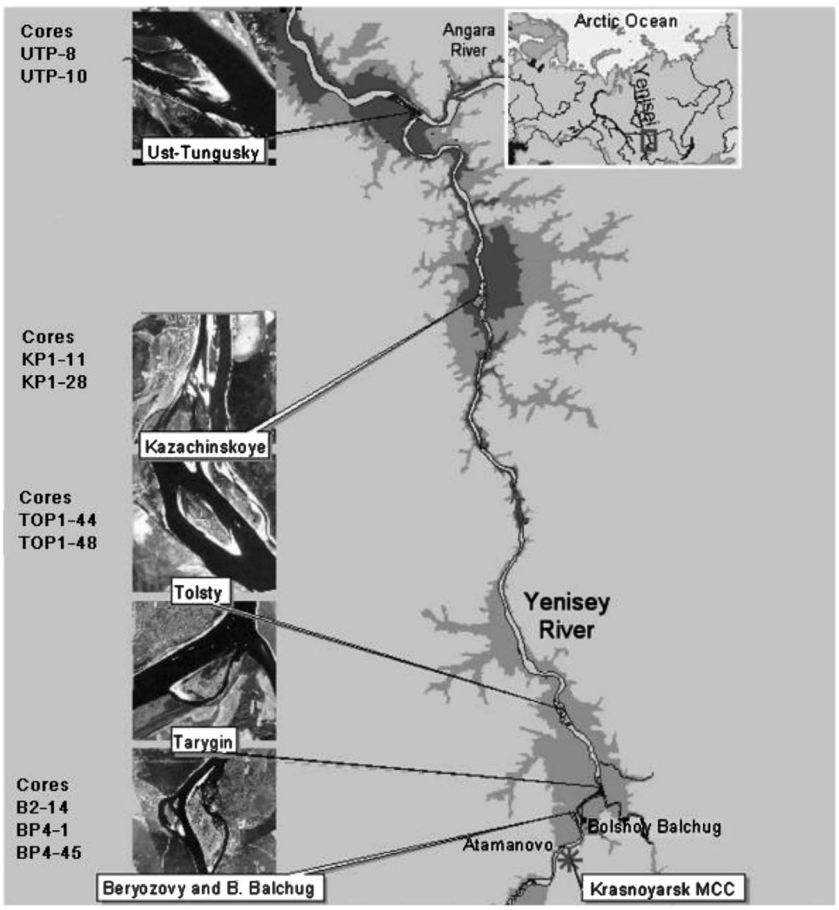

Fig. 1 Study sites and samples' location (a modified copy from Standring et al. 2009)

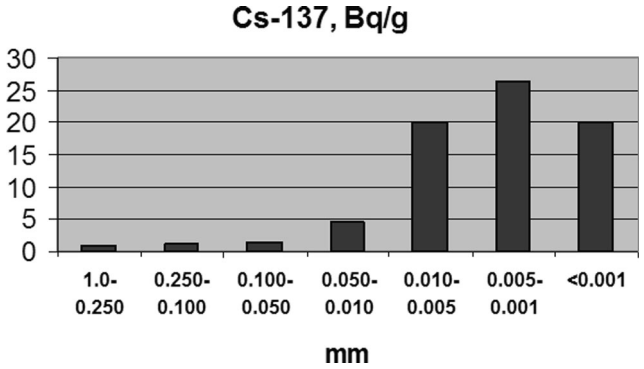

Fig. 2 Specific activity of ^{137}Cs in alluvial soil sampled at the low-level floodplain of the Beriozovy Island (core BP2-14, layer 15–20 cm deep)

0.125, 0.125–0.1, 0.1–0.063, 0.063–0.05, 0.05–0.01, 0.01–0.005, 0.005–0.001, and <0.001. In this paper, we discuss pelite (<0.01 mm) and aleurite (0.01–0.1 mm according to Puffengoldz 1978) fractions as the most significant for radionuclide fixation (Korobova et al. 2008, 2014).

To study radionuclide association with the total (Ctot) and organic fraction of C (Corg), we analyzed correlations between ^{60}Co, ^{137}Cs, and ^{152}Eu activity and the content of Ctot and Corg in 20 bulk samples collected from different horizons to a depth of 40–50 cm at soil profiles BP4-1, BP4-45, KP1-11, KP1-28, and TOP1-48 (Fig. 1). Concentrations of C in the soil horizons were determined using a Hewlett-Packard CHN analyzer Model 185. Carbon concentrations were measured before and after elimination of carbonates to separate organic carbon and carbonates. Organic C (Corg) was C determined after removal of carbonates, while Ccarb was calculated as the difference between the total C (Ctot) determined before carbonate removal and Corg.

The relationships between radionuclide and OM were studied on a sample from a layer at 5–10 cm depth from the soil profile BT2-14 (a duplicate of profile B2-14) located in the upper part of the Beriozovy Island (20 km from KMCC). The sample was subjected to extraction of humic and fulvic acid fractions and subsequent determination of radionuclides in the separated phases and the residue component. Organic fractions were separated by saturation of the air-dry sample with 0.1 M NaOH, with further precipitation of humic acid from an extract brought to pH 1 with 1 M HCl (Varshal 1994; Varshal et al. 1996). The separation resulted in three fractions of fulvic acids, humic acids, and residue containing the denuded mineral phase and the refractory organic residue.

In general, the data were useful in the process of characterizing the sites but were not amenable to statistical analyses. However, we still consider it worthwhile to present these data in the form of a case study. The experimental data may be used for supporting hypotheses pertaining to patterns of artificial radionuclide migration and sedimentation for particular sites (each with their own characteristic granulometry, content of organic and mineral C in alluvium).

3 Results and discussion

3.1 Parameters of KMCC radionuclide discharge behavior

Radionuclides may be discharged in water-soluble (ions, complexes), molecular, colloidal, and suspended particulate forms (Varshal 1994; Novikov 2010).

Once released, these forms could interact with natural mineral and organic suspended particles and colloids. During high water periods, with concomitant elevated suspended sediment loads, contaminated river waters come into contact with soils. The locations where this occurs can provide sedimentation areas for radionuclides. This has been simulated and experimentally proven by Nosov et al. (2010). Apart from physical and chemical sorption, a characteristic deposition of the suspended load, controlled by the prevailing hydrodynamic conditions, contributes to sedimentation of the suspended fraction of radionuclides (Linnik et al. 2006, 2014). It is worth noting that chemical properties of radioisotopes are similar to their stable analogues, but their mass amounts are not comparable and, in general, the behavior of trace elements depends upon the distribution of the major elements defining geochemical conditions (the so-called typomorphic elements, Perel'man 1975).

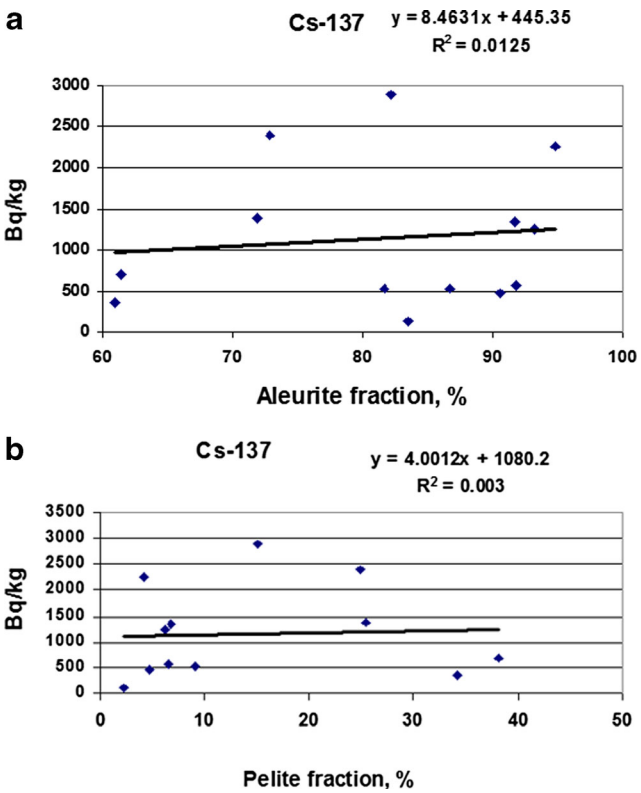

Fig. 3 Cs-137 and percent of aleurite and pelite fractions in the alluvial soil samples collected 20–250 km downstream from the KMCC

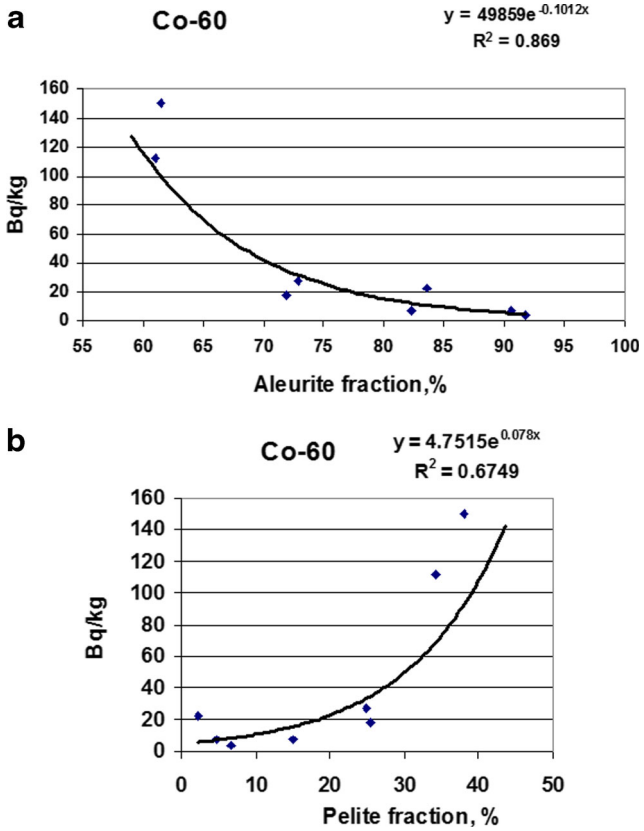

Fig. 4 Co-60 and percent of aleurite and pelite fraction in the alluvial soil samples collected 20–250 km downstream from the KMCC

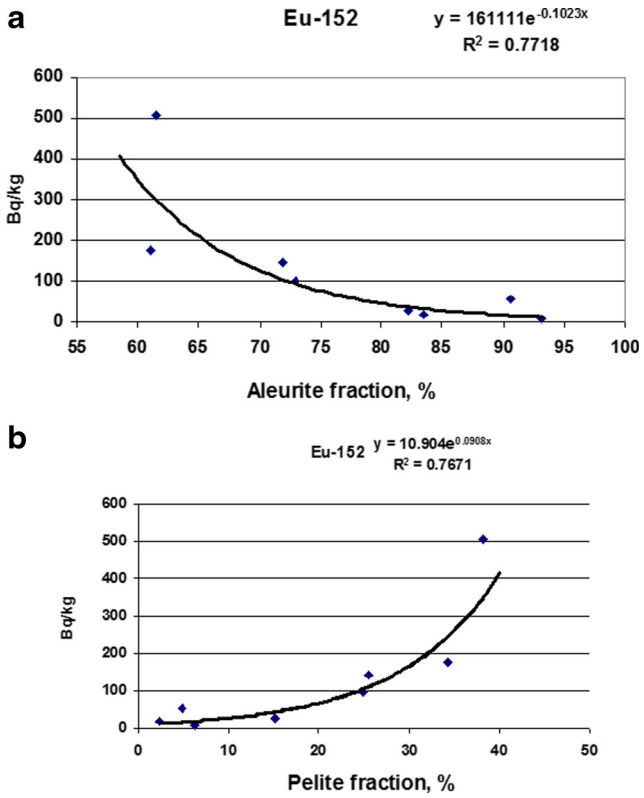

Fig. 5 Eu-152 and percent of aleurite and pelite fractions in the alluvial soil samples collected 20–250 km downstream from the KMCC

3.2 Distribution of ^{60}Co, ^{137}Cs, ^{152}Eu, and pelite and aleurite fractions in soil samples

Alluvium composition of the coastal and central parts of the floodplain is often variable in terms of texture. This is believed to reflect differences in the hydrological regime for sedimentation under high water discharge periods.

The distribution of ^{137}Cs-specific activity in different size fractions for a soil layer at a depth of 15–20 cm (sampled in the low-level floodplain of the Beriozovy Island in a dry shallow floodplain lake) is shown in Fig. 2. Here, we have the case of classical radionuclide contamination of floodplain soils due to sedimentation of fines. A considerable increase in specific ^{137}Cs activity was characteristic in three particle size classes

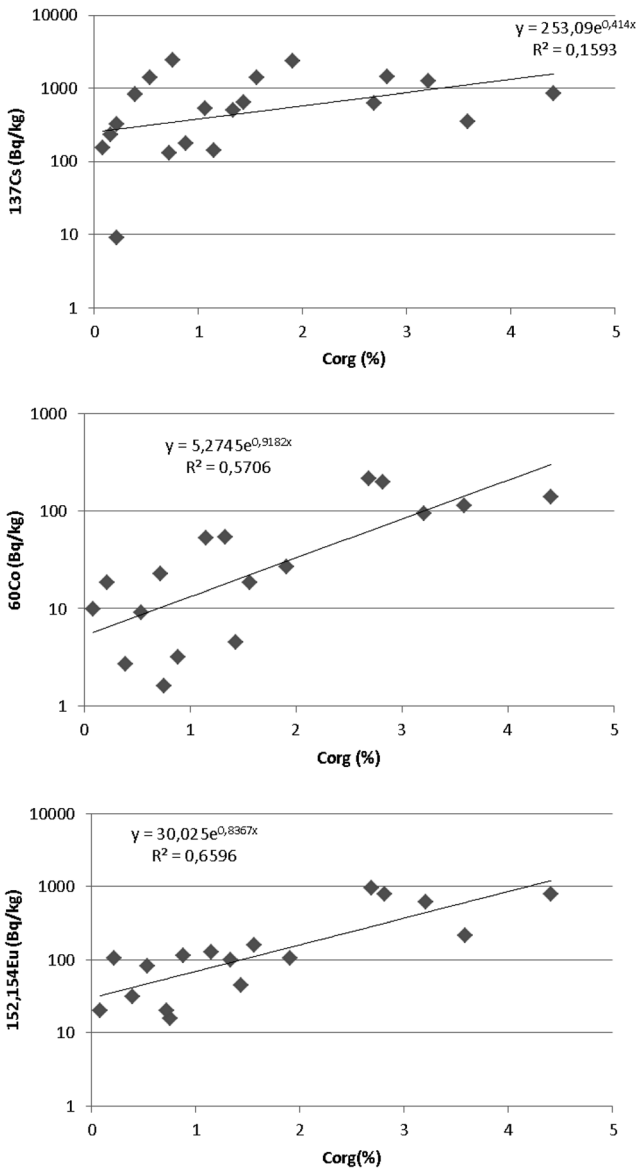

Fig. 6 Correlation between ^{137}Cs, ^{60}Co, ^{152,154} Eu activity, and organic carbon (Corg) concentration in layers of floodplain soil samples collected 20–250 km downstream from KMCC

belonging to the pelite fraction (<0.01 mm), with no significant difference among them: 20.0, 26.4, and 20.2 Bq g^{-1}, respectively (Fig. 2). A statistically significant higher activity of particles in the 0.01–0.05 mm range (coarse silt or fine sand depending on different classifications) as compared to 0.05–0.100 mm grains (very fine sand, U.S.D.A) was found in the aleurite fraction: 4.6 versus 1.3 Bq g^{-1}.

The relationship between ^{137}Cs activity and percent of aleurite and pelite fractions in the bulk alluvial soil samples (down to a depth of 40 cm) collected at study sites at a distance of 250 km downstream from the KMCC (Fig. 1) is shown in Fig. 3.

No correlation was found between ^{137}Cs-specific activity and the content of pelite and aleurite fraction of the bulk samples. In contrast to ^{137}Cs, both ^{60}Co and ^{152}Eu exhibited a clear positive exponential correlation with the pelite content in the bulk samples (R^2=0.67 and 0.77) and a negative relationship with the aleurite content (R^2=0.87 and 0.77, Figs. 4 and 5). Several reasons may be considered to explain this behavior.

The lack of correlation for ^{137}Cs may be explained by the river transport of this radionuclide in water-soluble and suspended forms in almost equal proportion. Moreover, the aggregation and adhesion of the most contaminated clay particles, as shown by Korobova et al. (2014), and a predominance of the low radioactive coarse fractions in the bulk samples, leading to the reduction in the contribution of the accumulating fraction to the total activity of the sample, may account for this observation. According to Sukhorukov et al. (2006), from 3 to 40 % (18 % on the average) of ^{152,154}Eu contained in floodplain and bottom sediments occurred in water-soluble and easily exchangeable forms. The percent of ^{152}Eu in the acid-soluble extract ranged from 79 to 99.2 %, while for ^{154}Eu, this fraction varied from 69 to 96.7 %. The authors suggested that ^{152,154}Eu radioisotopes exist in the Yenisey alluvial soils in the forms of acid-soluble microparticles, organic complexes, and some (unspecified) sorbed state. Taking into account the consideration that the time elapsed following major discharges was sufficient to enable a considerable degree of fixation, it can be inferred that the initially discharged fraction of ^{60}Co and ^{152}Eu was mainly water soluble, but both radionuclides formed mobile organic complexes that were readily adsorbed by the finest particles. As Eu is also a rare-earth element, the possibility of its chemisorption (in valence state II) by carbonates that are present in water and sediments cannot be excluded a priori.

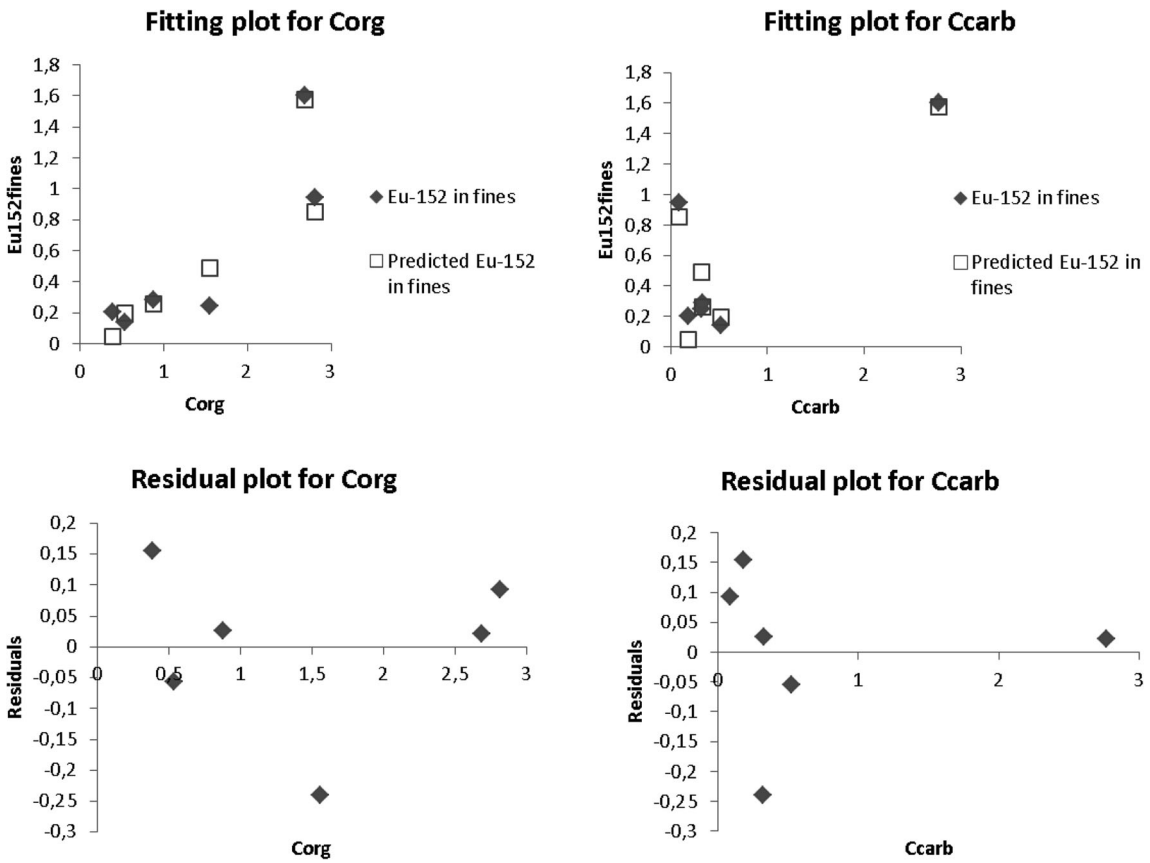

Fig. 7 Plots of regression analysis showing model fitting the real measurement data in B2-14 soil profile layers

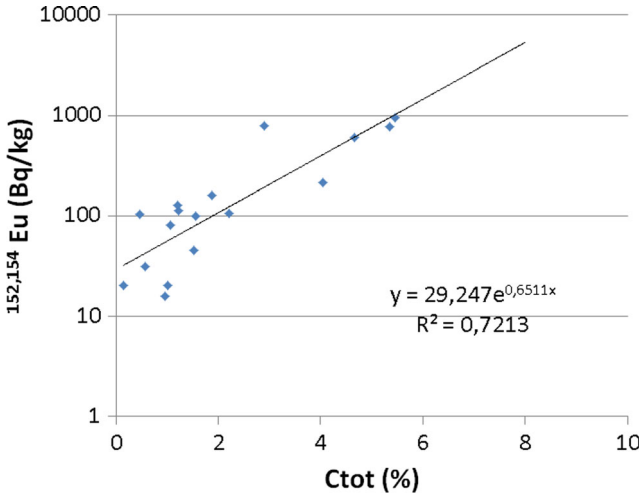

Fig. 8 Correlation between ^{152,154}Eu activity and total carbon content (Ctot) in layers of floodplain soil samples collected 20–250 km downstream from the KMCC

3.3 Distribution of ^{60}Co, ^{137}Cs, ^{152}Eu, and organic carbon in soil samples

Carbon concentration (Corg) in 20 samples from five soil profiles sampled at the Beriozovy, Tolsty, and Kazachinskoye sites varied in a wide range from 0.085 to 4.41 %, with an average value of 1.10 %. Cesium-137 did not show any relation to Corg, while Co and Eu activity appeared higher in samples with larger OM content. In particular, ^{152}Eu demonstrated a distinctly higher affinity to OC than ^{60}Co (Fig. 6). This finding is in agreement with published data on radionuclide behavior (Varshal 1994; Varshal et al. 1996; Novikov 2010). The same order of affinity of these radionuclides to organic substances (^{152}Eu>^{60}Co) was reported by Bondareva and Bolsunovskii (2008) who studied radionuclide speciation in sediments sampled from the near impact zone by sequential extraction.

3.4 Relation of vertical ^{252}Eu distribution in soil profile to mineral and organic forms of C

Recent studies of the Yenisei River bottom sediments, 15 km downstream from the discharge point, showed that 40–60 % of ^{152}Eu is present in the organic fraction and 2–7 % in carbonates (Bondareva and Bolsunovskii 2008). However, the vertical distribution of the radionuclide in soil profiles was not investigated.

Regression analysis performed for six successive layers of BP4-1 soil profile, located in the Beriozovy Island, showed that about 94 % of ^{152}Eu variation in fines of the layers could be explained by a linear model with Corg and Ccarb values as independent variables in the form

$$Eu{-}152(Bq\ g^{-1}) = 0.34 * Corg(\%) + 0.29 * Ccarb(\%) - 0.14$$

(Fig. 7).

This relation supports the hypothesis of a chemisorption of Eu by carbonates that may be critical for its migration.

The relevance of both forms of Eu accumulation on C was also supported by a higher correlation found between ^{152,154}Eu activity and TOC, as compared to ^{152,154}Eu and Corg in 20 samples collected in five soil profiles at three sites (BP, TOP, KP; Fig. 1) located 20, 50, and 180 km downstream from the KMCC (Figs. 8 and 6c).

No dependence was found for the corresponding ^{137}Cs values.

3.5 Radionuclide distribution in organic fractions of different mobility

Figures 9 and 10 show distributions of artificial and natural radionuclides in the fractions of fulvic acids, humic acids, and the residue containing the denuded mineral phase and the refractory organic residue. Radionuclides found in the first fraction, characterized by low molecular weight organic compounds, were the most mobile, whereas those in the second fraction were less mobile as they were associated with acid-resistant humus complexes that might be considered to be virtually insoluble in natural waters of the humid zone. The presence of radionuclides in the residue fraction is indicative of a portion strongly fixed onto the mineral matrix.

Radiocesium showed higher affinity to the high molecular mass organic compounds. Eu-152 and ^{60}Co, which are well known to undergo organic complexation, were present in considerable relative amounts in the fraction of the most mobile,

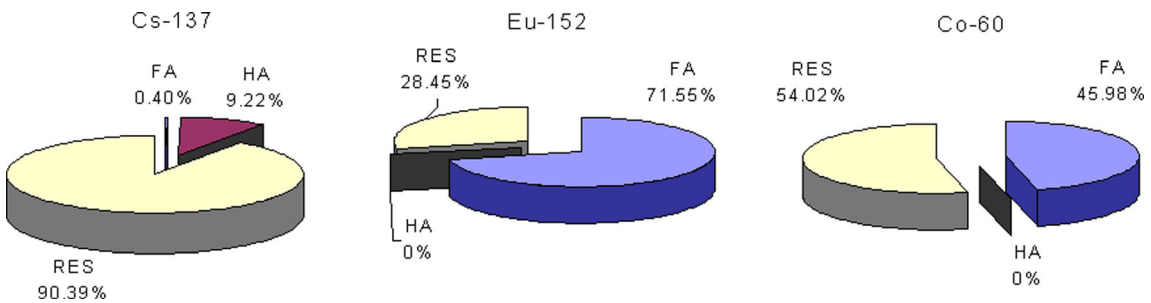

Fig. 9 Distribution of the artificial radionuclides in organic and residue fractions (*FA*, fulvic acid fraction; *HA*, humic acid fraction; *RES*, residue)

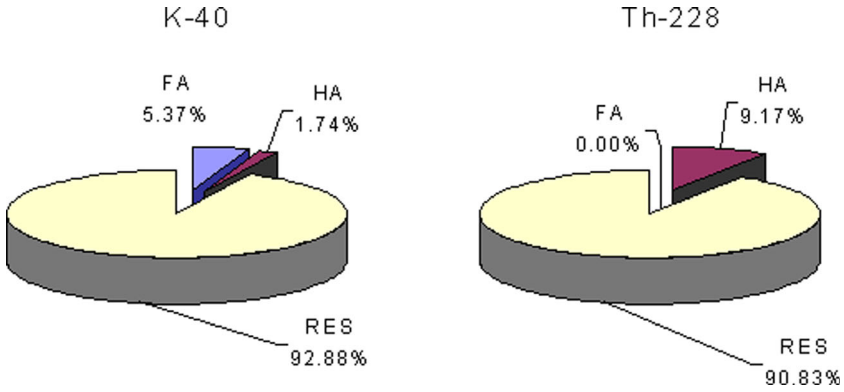

Fig. 10 Distribution of the natural radionuclides in organic and residue fractions (*FA*, fulvic acid fraction; *HA*, humic acid fraction; *RES*, residue)

low molecular mass fulvic acids (Fig. 9). K-40 and ^{228}Th associated with various minerals dominated in the residue fraction. Cs-137, in a similar way to ^{238}Th and ^{40}K, was mainly associated with the residue mineral fraction, presumably due to exchangeable sorption and further fixation in K-minerals (Fig. 10).

In general, our analysis of the affinity of radioisotopes to C forms showed that ^{152}Eu>^{60}Co were likely fixed in soil particles mostly by the organic and carbonate components, while ^{137}Cs association with OM was mainly phenomenological and indicated radiocesium fixation by the top organic soil horizons.

4 Conclusions

Cs-137 discharged from KMCC is mainly associated in floodplain soils and sediments with mineral particles of fine sands, silt, and clay that may form aggregates. Europium radioisotopes are mainly hosted by the pelite fraction.

^{152}Eu>^{60}Co are fixed in soil particles predominately by the organic and carbonate components, while ^{137}Cs association with OM is mainly phenomenological and indicates radionuclide contamination of the top humic horizons.

A different association of ^{137}Cs, ^{60}Co, and ^{152} Eu with particulate and organic fractions of river sediments and floodplain soils could be explained by the dominating discharge form (i.e., water soluble or particulate), affinity to organic substances of different mobility, sorption by minerals and their aggregates, and chemisorption.

This study confirms the contention that a comparative analysis of radionuclide distributions in soil fractions of different sizes and C content in soil layers and sediments subjected to radionuclide contamination can give valuable information for evaluation. This relates not only to the general radioecological situation and contamination levels but also to elucidation in terms of expanding the possibilities for interpretation of contamination events and migration patterns.

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