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Regional specificity of radionuclide fixation in river basins due to soil petrology and mineralogy

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Abstract

Purpose The main goal of this research was to compare and reveal the difference in the character of Cs-137 distribution and fixation in fine fractions of alluvial soils formed by the rivers draining areas of different geochemical origin with the corresponding mineral composition of clay fraction and to discuss the results in light of possible ecological perspectives.

Materials and methods Alluvial soil samples were collected in the Yenisey (Central Siberia) and Iput (the East European Plain) river basin areas contaminated by ^{137}Cs. Granulometric fine fractions (>0.01, 0.01–0.005, 0.005–0.001, <0,001 mm) were obtained by pipette technique; clay minerals were determined by a universal Carl-Zeiss Jena X-ray diffractometer (Germany); and ^{137}Cs activity was measured by a CANBERRA gamma spectrometer with HPGe detector (USA).

Results and discussion Clay mineralogy demonstrated regional specificity of its composition due to different origins of the soil-forming rocks. Clay fraction of alluvial soil samples collected in the Yenisey river floodplain consists mainly of chlorite-vermiculite and chlorite-smectite interstratifications while in the Iput river basin it is mainly composed of hydromicas and mica-smectite interstratifications. We infer that this may result in a higher potential ability of clay fraction for the radionuclide exchange and release in the former region.

Conclusions Specificity of local clay minerals as radionuclide sorbents should be of particular ecological concern in relation to long-term exchange processes and involvement of the radioisotope in local biogeochemical cycles including its transfer to food chains. The phenomenon is considered in light of the developed scientific field inaugurated as petro-mineralogical ecology and needs further investigation.

Keywords Chlorite · Ecological rating of clay minerals · Mica · Petro-mineralogical ecology · Radiocesium · Smectites · Vermiculite

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1 Introduction

As long ago as 1990, B.P. Gradusov, A.G. Chernyakhovsky, and N.P. Chizhikova inaugurated a new field in clay mineralogy as "ecological petrography and mineralogy of soils" (Gradusov et al. 1990a). The main idea was based on investigation of petrological composition and mineralogy of soils in combination with the weathered soil-forming rocks and sediments, both modern and ancient, as well as evaluation of this composition from an ecological perspective, namely, the ability of the soil mineral phase to fix ambient technogenic chemical elements and complexes.

The authors paid attention to the role of mineral composition of the native soils in regional and local plant nutrition, physical soil parameters depending upon crystal chemistry of minerals, and their resistance to weathering. Soils were rated according to their petrological and mineralogical composition with separation of the particular

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Table 1 Petrographic and mineralogical ecological categories (Gradusov et al. 1990b)

No	Edaphic categories	Soil-forming rock petrography	Soil-forming clay component
1	Ash-allophane	Fresh volcanic rocks: basalt, andesite, pyroclast, intermediate, and mafic and the associated consolidated and unconsolidated polymictic igneous-sedimentary deposits	Absent
2	Chlorite-smectite	Intermediate and mafic old magmatic rocks: gabbro, diabase, porphyry, their associated consolidated and unconsolidated polymictic sedimentary derivatives	Chlorite, vermiculite, chlorite-vermiculite, chlorite-smectite, trioctahedral smectite
3	Chlorite-hydromica	Acidic and intermediate magmatic and metamorphic rocks: granite, gneiss, crystalline schist, and their consolidated and unconsolidated polymictic-oligomictic sedimentary derivatives	Di- and trioctahedral hydromica, chlorites
4	Hydromica- smectite	Oligomictic carbonate and non-carbonate sandy-loamy-clay deposits including covering loams and loess	Dioctahedral hydromica and mica-smectite, chlorites, kaolinite
5	Smectite	Polymictic carbonate and non-carbonate clay and sandy clay deposits	Dioctahedral smectite and mica-smectite
6	Kaolinite-goethite-hydrargillite	Eluvial and chemogenic sedimentary iron-rich laterite and bauxite and their products	Goethite, hydrargillite, kaolinite
7	Kaolinite-palygorskite	Monomictic carbonate and non-carbonate clay sands and sandy clays	Kaolinite, palygorskite, dioctahedral hydromica, mica-smectite, chlorite
8	Kaolinite	Monomictic clays and sandy clays	Kaolinite, dioctahedral hydromica, chlorites, goethite, gibbsite

ecological soil categories (grades) corresponding to dominating initial and secondary clay minerals.

Ecological categories are characterized by a number of mineralogical and crystal-chemical parameters (Chizhikova 1992) that on the global scale have definite zonal features of weathering of the parent rocks (Chernyakhovsky et al. 1998, Table 1).

Ecological significance of the distinguished categories involves their different fixation and therefore control of exchange and migration of trace technogenic elements entering the environment.

The approach seems reasonable enough to be tested. In our previous publication, it was shown that ^{137}Cs fixation in the contaminated alluvial soil horizons in the Iput river basin contaminated after the Chernobyl accident depended mainly on soil enrichment in clay fraction (Korobova et al. 2007). However, the specificity of radioisotope fixation in fine fractions in different regions was not discussed.

The main goal of this research was to compare and reveal the difference in the character of ^{137}Cs distribution and fixation in fine fractions of the alluvial soils of two rivers draining areas of different geochemical origin with the corresponding mineral composition of clay fraction.

2 Study area and methods

There are several river basins in Russia subjected to local and regional technogenic radionuclide contamination (Bulatov 1996). We studied alluvial soils in the Yenisey basin contaminated by waste discharges of the Krasnoyarsk Chemical and Mining Combine directly to the stream from 1960 to 1992, and floodplain soils of the Iput river contaminated by aerial fallout from the Chernobyl accident (April 1986).

The Yenisey river drains the areas of intermediate and mafic old magmatic rocks presented by gabbro, diabase, and porphyry and their associated consolidated and unconsolidated polymictic sedimentary derivatives, while the Iput river floodplain is composed of the reworked non-carbonate and carbonate sandy-loamy sand and clay loamy fluvioglacial and moraine deposits. Therefore, a different character of radiocesium fixation by clay minerals was suggested.

We should mention that despite the difference in sources and forms of ^{137}Cs contamination (aerial fallout of fission products and waste discharge to the river), the contaminated areas have much in common in respect to radiocesium phases in the initial release and their involvement in natural processes that have taken place in river basins. First of all, there is some

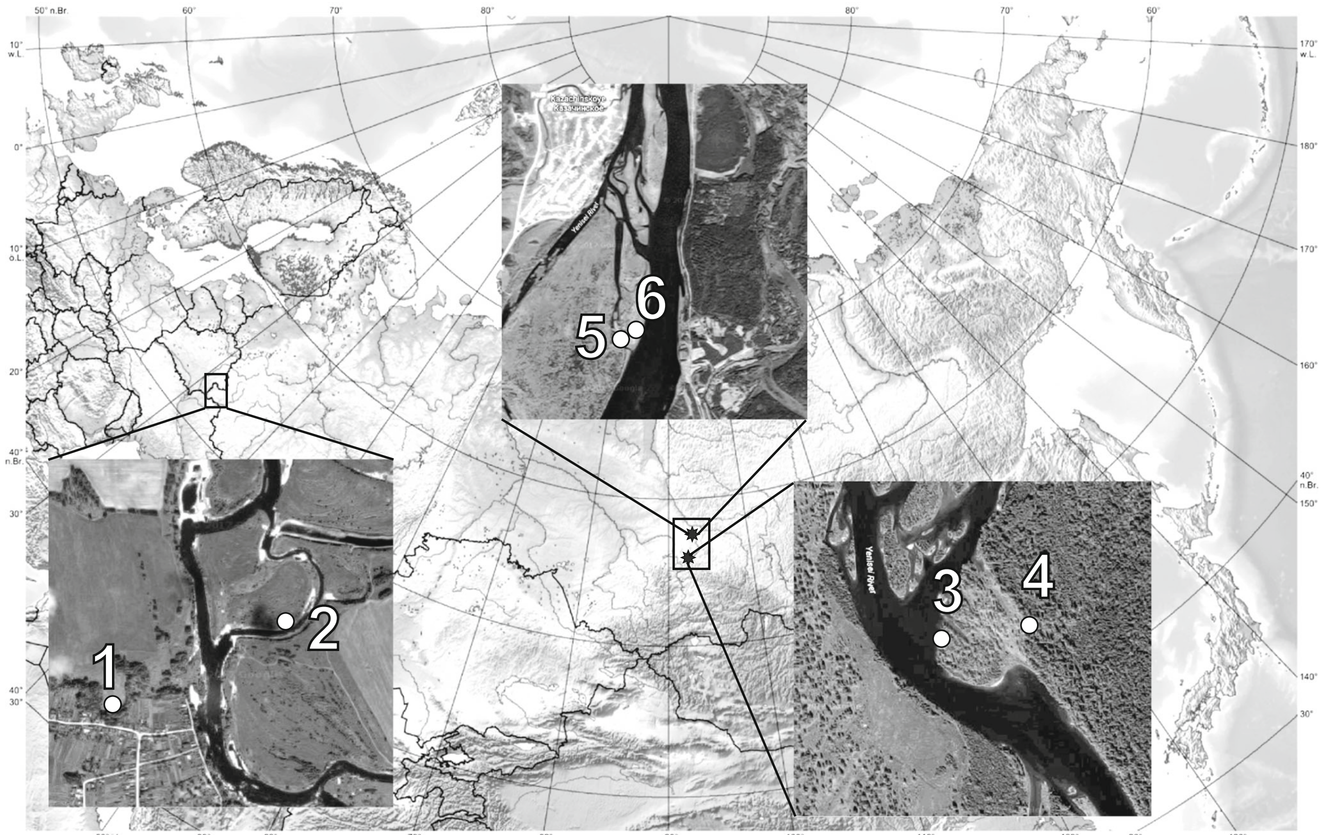


Fig. 1 Sampling locations and their co-ordinates (Table 2) on the Iput and Yenisey rivers

similarity in the initial phases of ^{137}Cs contamination: in both cases, a considerable part of the radioisotope was water-soluble. Water-soluble fraction of ^{137}Cs in the condensation type of the initial fallout characteristic for the Iput river basin could reach 50% (Konoplev et al. 1993; Shcheglov 2000). The mentioned proportion of liquid and solid phases of radiocesium in the initial Chernobyl deposition was later used to explain the effect of almost twofold decrease of ^{137}Cs inventory in Iput flood plain areas (Linnik 2008). Portions of water-soluble and suspended forms of ^{137}Cs in its discharge to and transfer by the Yenisey river were also almost equal (Nosov et al. 1993; Vakulovsky 2003). In addition to similar proportions of liquid and solid phases, both river basins were subjected to radionuclide contamination during the high water periods that prove similarity in the initial radiocesium contamination of the floodplains. Further behavior of radiocesium in both areas was defined by soil sorption of its water-soluble part and deposition of the contaminated suspension controlled by hydrological processes (Linnik et al. 1999).

Soil samples were collected from soil profiles examined in the summer period of 1999 (Iput basin) and 2000 (Yenisey basin, Fig. 1): (1) in the islands (Kazachij, the Yenisey river, profiles KP1–11 and KP1–28, and the Iput island formed at confluence with its tributary Buldynka river outlet, profile OEP-1) and on the riversides (profiles MBP-1 and 2, Bulchug site, Yenisey river and Buldynka river, profile B-2, Korobova et al. 2008, 2014; Korobova and Chizhikova 2007). Soil sampling sites were selected after the preliminary field gamma spectrometry data obtained using a CORAD field gamma spectrometer (Chesnokov et al. 1997), to identify areas of highest contamination. Soil profiles characterized different floodplain levels (high, medium, or low) with a corresponding moisture content of soils, i.e., wet soddy-gley according to Russian classification or Fluvisol Histic in WRB in low-level position and a comparatively dry soddy soil (Fluvisol Eutric and Dystric in WRB) in medium- and high-level locations (Table 2). Cs-137 was measured in the collected samples by a laboratory gamma spectrometer (CANBERRA, USA) with a HPGe detector; the efficiency of the detector at 661.6 keV was ca. 1.2%, with an energy resolution for the 122 keV line (^{57}Co) of 0.909 keV, and that of the 1332 keV line (^{60}Co) of 1.87 keV. Most contaminated soil layers were later analyzed for their clay composition. Clay fraction was extracted by a Gorbunov pipette technique that included preliminary wetting of the soil and its manual grinding with a rubber pestle (Gorbunov 1971). Clay mineral composition was determined by a universal Carl-Zeiss Jena XZG-4A X-ray diffractometer, voltage 30 kV, current 30 mA (Germany). Oriented specimens saturated with magnesium were investigated (1) in air-dry form, (2) after glycol

Table 2 Cs-137 activity of bulk sample, its clay fraction, and mineralogical composition of the latter in soil specimens collected in the Yenisey and Iput river basins

Location of the soil cut, soil type in Russian classification and WRB, and the soil profile index	Soil layer depth, cm	Cs-137 activity (Bq/g)		Cs-137 activity in clay fraction (%)	Mineral composition of clay fraction (%)		
		Bulk sample	Clay fraction		Kaolonite, chlorite	Hydro-mica	Smectites[1,2]
Yenisey young riverside floodplain near set. Bolshoy Balchug, soddy-gley sandy-loam soil (Fluvisol Histic, MBP-1, Fig.1, plot 3, N 56°28’05.13”, E 93°41’35.12”)	5–8 8-10	1.0 0.9	5.1 5.6	45.7 53.0	18 17	33 48	50[1] 35[1]
The same site, middle-level floodplain, soddy silty-sand soil (Fluvisol Umbric, MBP-2, Fig. 1, plot 4, N 56°28’06.94”, E 93°41’55.82”)	6–8 30-35	1.7 0.4	3.5 0.6	31.5 14.2	17 23	40 54	43[1] 23[1]
High floodplain of the Kazachy Island, Yenisey basin, soddy sandy soil (Fluvisol Dystric, KP1–11, Fig. 1, plot 6, N 57°39’36.97”, E 93°17’31.91”)	5–10	0.4	4.6	16.4	33	49	18[1]
The same island, middle-level floodplain soddy sandy soil (Fluvisol Dystric, KP1–28, Fig.1, plot 5, N 57°39’35.25”, E 93°17’20.32”)	5–10	2.0	9.7	47.5	19	55	26[1]
Iput river basin, island floodplain, soddy gley-laminated soil (Fluvisol Histic, OEP-1, Fig. 1, plot 2, N 52°35’47.70”, E 31°44’56.89”)	0–3 3–6 6–10 10–14	2.1 3.5 31.2 9.5	18.3 34.1 222 97.4	25.1 35.5 53.8 42.1	7 17 11 24	53 68 54 49	40[2] 15[2] 35[2] 27[2]
Buldynka river floodplain, soddy-gley-laminated soil (Fluvisol Histic, B-2, Fig. 1, plot 1, N 52°35’38.91”, E 31°44’26.41”)	2–4	4.5	37.4	47.2	28	60	12[2]

[a] Chlorite-vermiculite, chlorite-smectite

[b] Mica-smectite interstratifications

treatment, and (3) after calcination at 550 °C for 2 h. Diagnosis of minerals has been carried out using a standard technique (Brown 1965).

3 Results and discussion

Radiocesium fixation in soils is known to depend mainly on sorption processes that are controlled by granulometric and mineral fractions, and the associated crystal chemistry of clays (Comans et al. 1991; Cornell 1993; Konoplev and Konopleva 1999; Bostick et al. 2002). Smectites and illitic clays are known to be extremely important in ^{137}Cs binding and fixation, and there are many publications considering peculiarities of ^{137}Cs sorption (e.g., Comans and Hocley 1991; Hird et al. 1996; Poinsott et al. 1999; de Koning et al. 2000; de Koning and Comans 2004) and the relevance of mineralogy in assessing soil vulnerability (Facchinelli et al. 2001). The major part of the studies were performed in laboratory-modeled conditions. Investigation of the soils contaminated after the Chernobyl accident proved that during long-term soil-water contact, the surface-sorbed cations diffuse inside minerals

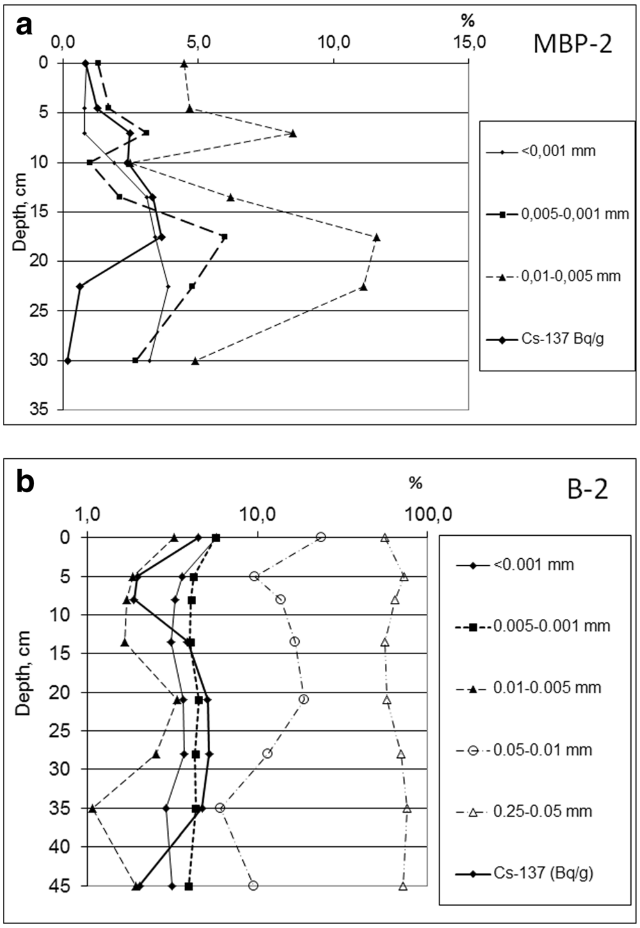


Fig. 2 Vertical profiles of fine fraction content (%) and ^{137}Cs activity (Bq/g) in soil cross sections. **a** Alluvial soddy silty-sandy soil, middle-level flood plain of the Yenisey river (profile MBP-2) **b** Alluvial soddy-gley, low-level floodplain of the Buldynka stream close to its confluence with the Iput river (profile B-2)

leaving the surface (Konoplev et al. 1988; Sobotovich et al. 1991). Recent experiments with vermiculite performed by Dzene et al. (2015) showed that most of cesium sorbed on interlayer sites remains fixed due to the collapse of the structure under aqueous conditions while radioions sorbed on external (edge and basal) sorption sites can be desorbed by ammonium ions. Our case study confirmed a correlation between vertical distribution of fine fractions in soil layers and radiocesium activity of these layers (Korobova et al. 2005, 2008, 2014; Fig. 2).

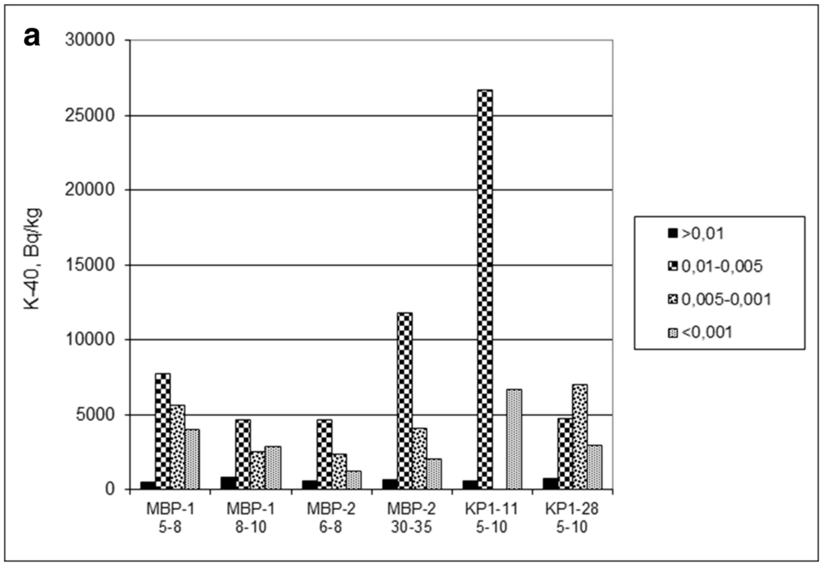
In five specimens out of six ones sampled in the Yenisey floodplain, maximum ^{137}Cs activity was found in clay fraction (<0.001 μm) while ^{40}K was mostly associated with middle-sized and fine silt (0.01–0.005 μm). Taking into account mass contribution of each fine-sized fraction, silty and clay fractions contained the major part of ^{137}Cs (47–73%) except for samples from sandy layers (MBP-2, 30–35 cm deep, and KP1–11, 10–15 cm deep) relatively enriched in ^{40}K (Fig. 3), where ^{137}Cs activity in clay fraction was considerably lower (14 and 16%, correspondingly, Table 2). This may be explained by mineralogical composition of the coarser fraction. For example, the data obtained in the framework of INCO-Copernicus project STREAM showed that the mineralogical composition of the 0.25–0.5 mm fraction from the sample 5–10 cm deep taken from the KP1–11 soil profile was noted for domination of feldspar (up to 65%, including microcline) known for exchange reactions as Cs^+-Me^+ (Brown et al. 2002, pp. 66–67). A significant correlation between ^{137}Cs and ^{40}K activity in the bulk samples taken from the successive KP1–11 soil profile horizons ($r_{0.01} = 0.833$, $n = 13$) is an additional argument in favor of the above explanation.

Diffraction analysis of the obtained clay fractions confirmed that the Yenisey and Iput river soils belong to two different ecological edaphic categories (Table 1): hydromica-smectite (the Iput river site) and chlorite-smectite (the Yenisey river site), corresponding to peculiarities of composition of the soil-forming rocks of the regions.

Indeed, clay fraction of the samples collected in Yenisey is characterized by chlorites, chlorite-vermiculite, and chlorite-smectite interstratifications with low share of smectite layers and hydromicas. In samples from the Iput river floodplain, the dominating position belongs to hydromica of dioctahedral type with admixture of trioctahedral type as well as mica-smectite interstratifications with high share of smectite layers. The amount of chlorite and kaolinite in clay fraction is insignificant (Table 2).

The clay XRD analyses performed in our earlier study (Korobova and Chizhikova 2007) showed that in the samples from the Iput river, maximum ^{137}Cs concentration was found in fractions enriched in hydromicas and mica-smectite interstratifications with high share of smectite layers, while in the Yenisey floodplain soils clays with maximum radiocesium activity were presented by hydromica, chlorite, and chlorite-vermiculite interstratifications. Later studies of mineralogical composition of the alluvial soils were performed by Sukhorukov et al. (2012) near set. Abalakovo downstream the Yenisey confluence with the river of Angara confirmed that fractions most contaminated by Cs-137 were composed of chlorite, vermiculite, smectite, and micas (muscovite).

Radiocesium sorption by chlorite and chlorite-vermiculite interstratifications could be mainly basal or in its frayed edges (FES) while in mica-smectite interstratifications the radioisotope may penetrate the interlayer space where its fixation is considerably stronger (Comans et al. 1991; Chang and Hsu 1993; Facchinelli et al. 2001; de Koning and Comans 2004). Further destruction of crystallites would lead to higher mobilization of radiocesium sorbed by chlorite than that hosted by mica-smectite interstratifications. If this inference is correct, radiocesium mobilization from chlorite-vermiculite interstratifications made of radionuclide hosting chlorites would be higher than for interstratification domains characterized by hydromicas and mica-smectite. This may explain a strong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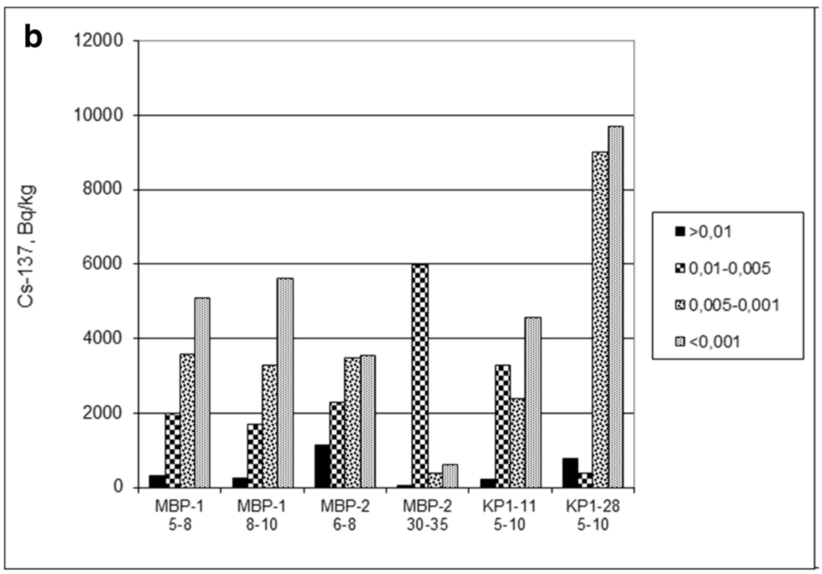
KP1-11 5-10
KP1-28 5-10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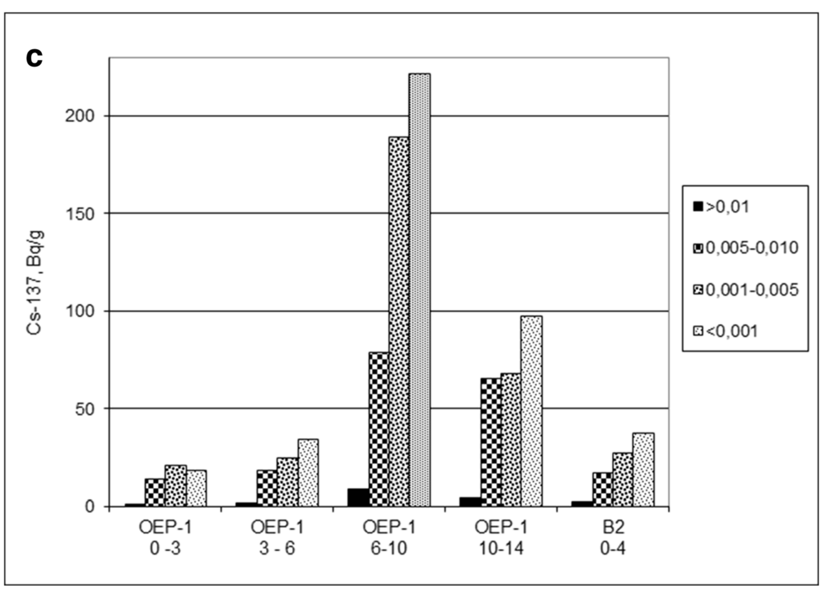
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OEP-1 0 -3
OEP-1 3 - 6
OEP-1 6-10
OEP-1 10-14
B2 0-4

◀ **Fig. 3** ^{40}K (**a**) and ^{137}Cs (**b**, **c**) activity in fine fractions (mm) of some of the alluvial soil layers (for location of the soil cut, soil type in Russian classification and WRB, and the soil profile index, see Table 1)

fixation of ^{137}Cs in the top layer of sandy alluvial soils widespread in areas of the Chernobyl contamination (in our case—the Iput study plots).

Taking into account the fact that both floodplain areas (the Iput and the Yenisey) are used for grazing, we consider worth discussing some differences in long-term contamination of local food chains due to differences in radiocesium fixation, secondary release, and burial in local alluvial soils. In Iput floodplain where soils contain radiocesium in fixed state in the upper soil layers, one may expect milk contamination due to transfer of radiocesium to milk via soil ingestion (for example, see Abrahams and Steigmajer 2003). Therefore, taking into account amounts of grass and soil ingested by dairy cattle, the soil to milk transfer in Iput local food chains may become comparable to soil to grass to milk transfer or even more important than the latter. In contrast, ^{137}Cs higher release from the hosting chlorite soil mineral fraction in the Yenisey floodplain contaminated with this radioisotope may contribute to its faster transfer to freshwater and self-purification of floodplain locations as compared to alluvial soils in the contaminated Iput river basin. However, the hypothesis needs further investigation and more detailed testing in the future.

4 Conclusions

A study of ^{137}Cs distribution in fine fractions of alluvial soil profiles of geochemically different river basins contaminated by ^{137}Cs discharge to the river (Yenisey river basin) and atmospheric fallout (Iput river basin) confirmed that anthropogenic pollution by the particular radioisotope leads to its involvement in natural processes and its sorption by mainly fine fractions of the site-specific clay minerals. Performed studies showed that clay fraction of alluvial soils sampled in the Yenisey river floodplain was presented mainly by chlorite-vermiculite and chlorite-smectite interstratifications while in the Iput river basin it consisted of mainly hydromicas and mica-smectite interstratifications which may have resulted in a higher radionuclide exchange and release from clay fraction in the former region. Specificity of local clay minerals as radionuclide sorbents are believed to be of particular ecological concern in relation to long-term exchange processes and involvement of radioisotopes in secondary mobilization to local biogeochemical cycles including transfer to food chains.

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