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Radium Distribution in Anthropogenic Soils as a Function of Soil Physicochemical and Mineralogical Parameters

L. M. Noskova and I. I. Shuktomova

Institute of Biology, Komi Scientific Center, Ural Branch, Russian Academy of Sciences,
ul. Kommunisticheskaya 28, Syktyvkar, 167982 Russia
e-mail: lmn06@mail.ru

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Abstract—We investigated the effects of physicochemical and mineralogical compositions of radioactively contaminated podzolic and soddy–meadow soils on the ^{226}Ra distribution in the soil profile. It was shown that the physicochemical compositions of soils depend on the type of contamination and have different effects on the mobility of the radionuclide. The presence of phosphorus and organic matter was shown to reduce ^{226}Ra mobility. The highest Ra content was observed in the fraction <10 μm. The role of the composition of the fine fraction of the anthropogenic soil was evaluated. Relations between ^{226}Ra and organic matter contents in different size fractions are satisfactorily described by the logarithmic function $C_{\text{Ra}} = a + b\ln(C_{\text{org}})$ ($R^2 = 0.9, p < 0.05$). The concentrations of ^{226}Ra and organic matter show a positive correlation with relative percentages of illite- and smectite-group minerals and a negative correlation with chlorites.

Keywords: radium, smectite, chlorite, illite, kaolinite

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INTRODUCTION

In recent years, the levels of radionuclides in the environment have increased considerably due to anthropogenic activity. Under equilibrium conditions, 90.5% of all pollutants released into the environment will finally accumulate in soils (Blum, 2001). Soil is an essential part of the landscape controlling the behavior of chemical elements and their migration into other media.

The concentrations of radionuclides in soils depend on various migration mechanisms. The mobility of radionuclides is governed by the solubility of the compounds containing these elements, on the one hand, and the strength of their sorption by the soil, on the other hand. The sorption processes in the soil could limit the transport of radionuclides in the environment. The sorption strongly depends on the mechanical and mineralogical compositions and physicochemical characteristics of soils, such as the organic matter content, the presence of complexing agents, the acidity of the soil solution, radionuclide concentrations, and a number of other factors (Aleksakhin et al., 1990).

There are ample data in the literature regarding the positive relationship between radionuclide concentrations in soil and contents of clay and silt (Rubtsov and Pravdina, 1971; Yastrebov, 1973; Ishchenko and Butnik, 1990; Shuktomova and Noskova, 2008; Landa, 1982; Taboada et al., 2006), the finest fraction, which is primarily composed of clay minerals, such as illite-group minerals, montmorillonite, kaolinite, and non-clay sesquioxides. Experimental data confirmed the sorption of radium onto soils and soil components, such as quartz, clay minerals, colloidal silicic acid, Fe and Mg hydroxides (Rachkova and Shuktomova, 2006; Benes et al., 1984, 1985; Murray et al., 1992), and organic matter (Ovchenkov, 1974; Vandenhove and Van Hees, 2007).

The study of mechanisms involved in radionuclide fixation in soils can enhance our understanding and facilitate long-term predictions of radionuclide migration and redistribution. This is of particular importance for the nuclear legacy sites that have been operated for a long time in the absence of information needed for safety and environmental performance and without construction of repositories for disposal of radioactive waste.

The goal of this study was to investigate the influence of physicochemical characteristics and mineralogical compositions of radioactively contaminated soils on the distribution of ^{226}Ra, the main dose contributor in the soil cover of a former radium production site (Komi Republic).

MATERIALS AND METHODS

The study was conducted in the Ukhta region (Komi Republic) in areas with elevated natural background radiation due to past activities (1931–1957) involving the recovery of radium from oil field produced water and, in part, waste uranium ore delivered from other regions of the country.

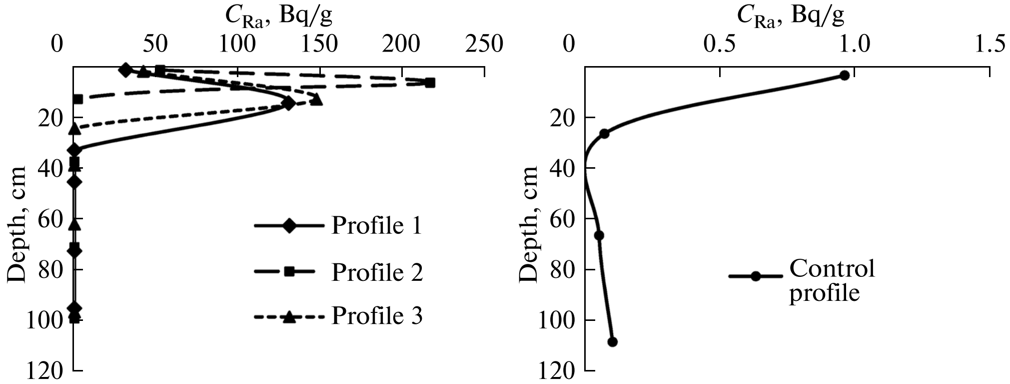

Fig. 1. Vertical distribution of ^{226}Ra in the anthropogenic podzolic soil.

Four soil profiles were taken at each of two sites with different soil types, which were identified using the technical classification of Gerasimova et al. (2003).

Site 1 has an anthropogenic podzolic soil covered with a 0–4 cm thick A_0 horizon, which is underlain by a 7–23 cm thick layer of black radioactive tailings (leached sintered tailings). Below the tailings layer, the soil profile has several subunits, which were identified as A_2 (to 16–65 cm), B (to 51–80 cm), B_1 (to 73–110 cm), and BC (below 73–110 cm) horizons. The upper horizons (A_0 + tailings) have near-neutral pH values (5.84–7.56), and the soil acidity increases down the profile to pH = 4.65–6.80. The upper horizons are enriched in P, K (210 and 207 mg/100 g, respectively) and exchangeable Ca and Mg (109 and 7.12 mmol/100 g, respectively). The concentrations of these elements in contaminated soils are well above those found in an uncontaminated (control) plot at this site. The organic carbon content reaches 43%.

At Site 2, there is a chemically contaminated soddy–meadow soil. The contamination resulted from the discharge of waste water after ^{226}Ra recovery. In 1962, as a remediation measure to reduce background radiation, the site was partially covered by a sand–gravel mix layer, which presently ranges in thickness from 10 to 30 cm, and an 8-cm-thick sod cover (A_s) was eventually established on the filling layer. Compared to Site 1, the soils at Site 2 are much lower in P (up to 23.3 mg/100 g), K (up to 44 mg/100 g), exchangeable Ca and Mg (up to 47.3 and 8.42 mmol/100 g, respectively), and organic carbon (up to 11.7%). Soil pH values vary from 5.6 to 7.7 (average 6.7).

Size fractions (<1, 1–5, 5–10, 10–100, 100–250, and >250 μm) were separated by decantation (Gorbunov, 1972). The mineralogy of the clay fraction was determined by X-ray diffraction analysis of oriented samples after standard diagnostic treatment (Shimadzu XRD-6000 diffractometer, Cu$K\alpha$ radiation, Ni filter at 30 kV and 20 mA). Diffractograms were obtained for three states of each sample: (a) air-dried, (b) glycerol solvated, and (c) heated at 300 and 500°C. All samples were scanned from 2°–32°, 2°–26°, and 2°–20°2θ.

The content of ^{226}Ra in soil samples was determined by the emanation method using an Alfa-1 apparatus. The sensitivity and error were 2.0×10^{-12} g/g and 15%, respectively (Starik, 1969). The physicochemical characteristics of soils were determined using methods described in textbooks (Arinushksina, 1962; *Agrochemical Methods* ..., 1975). The data were interpreted using the analysis of variance (Lakin, 1990) in the Microsoft Excel 2007 and Statistica 6.1 (StatSoft Inc.) programs.

RESULTS AND DISCUSSION

Figure 1 shows the distribution of ^{226}Ra in the anthropogenic podzolic soil. Despite the long period of time after the contamination, the highest radionuclide concentrations were detected in the upper soil horizons (A_0 + tailings), which account for 98% of the total radionuclide content (Noskova and Shuktomova, 2009). The radionuclide migration down the soil profile is insignificant. The high concentrations in the upper soil layers in the control plot of the site not occupied by tailings are indicative of secondary contamination owing to the lateral migration of contaminants.

Figure 2 shows the distribution of ^{226}Ra in the chemically contaminated soddy–meadow soil. The highest radium concentrations were also detected in the upper soil layers, but the depth distribution of this radionuclide indicates illuviation to the lower soil layers. The distribution of the radionuclide in profile 3 was influenced by the presence of a sand–gravel filling layer in top soil, where ^{226}Ra concentrations are lower than in the upper layers of the other profiles.

Stepwise multiple regression analysis was used to examine the relationship of soil composition and properties with ^{226}Ra content (Statistica 6.1). For example, the relation between the ^{226}Ra concentration and physicochemical characteristics of the anthropogenic podzolic soil, including soil pH, contents of organic carbon (C_{org}), phosphorus (P_2O_5), potassium (K),

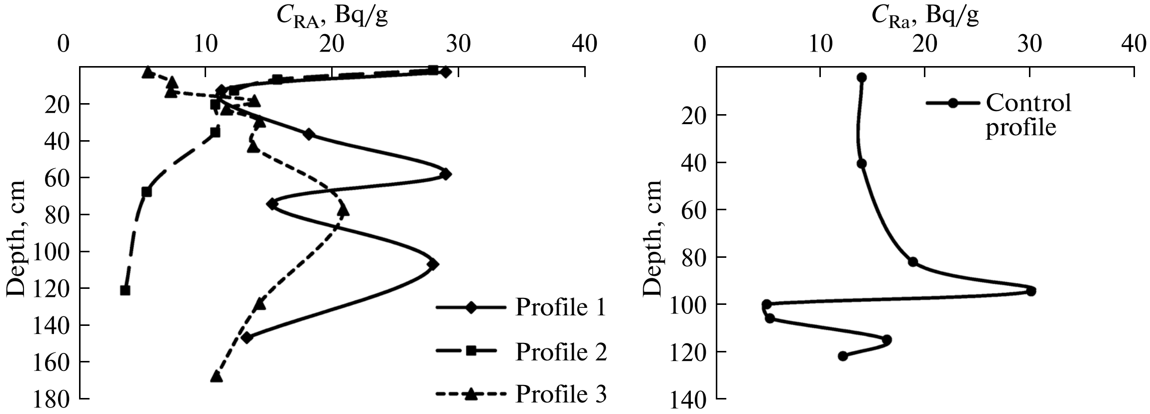

Fig. 2. Vertical distribution of ^{226}Ra in the chemically contaminated soddy–meadow soil.

absorbed calcium (Ca^{2+}), and magnesium (Mg^{2+}) can be described by the equation

$$C_{Ra}\ (\text{Bq/g}) = 0.839C_{P_2O_5}\ (\text{mg/100g})$$
$$(R^2 = 0.885, p = 0.002).$$

Thus, the P_2O_5 content of soil is a key factor controlling the depth distribution of the radionuclide. Weaker correlations were found between the specific activity of ^{226}Ra, pH ($r = 0.54, p \leq 0.05$), and C_{org} (0.40).

For the chemically contaminated soddy–meadow soil, a significant correlation was found between the depth distribution of the radionuclide activity and organic carbon content. The regression equation can be written as

$$C_{Ra}\ (\text{Bq/g}) = 0.715C_{org}\ (\%)\ (R^2 = 0.870, p = 0.001).$$

A strong ($p < 0.001$) correlation was also found between the concentrations of ^{226}Ra and absorbed Ca^{2+} and Mg^{2+} (0.67 and 0.72, respectively).

Thus, the physicochemical characteristics of contaminated soils have different effects on the radionuclide mobility. In the plot contaminated by solid radioactive tailings, a significant correlation was established between the contents of Ra and P_2O_5, which is present at elevated levels in the tailings. One of the stable forms of Ra absorption in the tailings material is insoluble radium phosphate compounds. In the soddy–meadow soil contaminated with produced water, ^{226}Ra interacts strongly with soil organic matter, which can result in a relatively low mobility in the soil profile.

The particle size distribution is one of the major factors controlling the concentration of an element in the soil profile. However, the soil is composed of materials of different origin. On the one hand, up to 95% of the soil mass consists of various minerals derived from rocks. On the other hand, soils contain organic compounds dominated by humus.

To study the effects of mineral and organic compounds on the mobility of ^{226}Ra, we determined the radionuclide concentrations in different fractions of anthropogenic soils, as well as the mineralogical composition and organic matter content of each fraction.

The results show that the sand-size fraction dominates the bulk of the anthropogenic podzolic soil (without considering the tailings layer). This fraction accounts for 80.3–86.2% of the bulk soil mass, most of which (66–73%) are particles 10–250 μm in size. The clay fraction shows the following particle size distribution: clay > fine silt > medium silt. All profiles show a slight (2–4%) increase in the proportion of clay in the lower layers.

The sand-size fraction accounts for much (58–78.3%) of the chemically contaminated soddy–meadow soil. The uppermost soil layers are mostly medium–heavy loam, and the lower layers are composed of loose sand. The particle size distribution in profile 3 has been changed considerably because of the presence of the filling layer, which resulted in the occurrence of loose sand in the upper layer.

Therefore, the particle size compositions of the studied soil profiles display similar characteristics (Fig. 3). The major portion of the soil mass is composed of >10 μm particles. The clay and sand fractions account for 16.5 and 83.5% in the anthropogenic podzolic soil and 29.9 and 70.1% in the contaminated soddy–meadow soil, respectively. In both types of soils, the contents of size fractions decrease in the following order: 10–100, 100–250, >250, <1, 1–5, and 5–10 μm.

The results show that the highest ^{226}Ra levels in the anthropogenic podzolic soil are associated with <1 μm particles of the clay fraction (Fig. 4a). In general, the percentage of radionuclide activity associated with the clay and sand fractions are 80.8% and 19.2%, respectively. The analysis of the distribution of Ra concentrations by particle size in the soil profile revealed the following trends (Fig. 4b). The ^{226}Ra activity is mostly distributed among 10–100 μm particles (32%), <1 μm particles (23.4%), and 100–250 μm particles (16.5%). The contribution of the <1 μm particles is related to their high Ra concentration, whereas that of the 100–

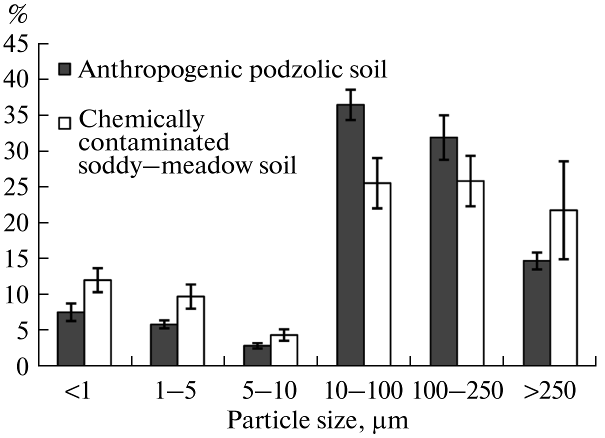

Fig. 3. Particle size composition of radioactively contaminated soils.

250 μm particles is due to the highest mass fraction of this fraction in the soil. The contribution of the other fractions to the ^{226}Ra activity is considerably lower.

In the chemically contaminated soddy–meadow soil, elevated ^{226}Ra levels are detected in the clay fraction (67.9%), which are slightly lower than that of the anthropogenic podzolic soil. The highest specific ^{226}Ra activity of 23.7 and 23.3% is characteristic of <1 and 5–10 μm particles, respectively; and that of 1–5 μm particles is slightly lower (20.9%). The specific activity of ^{226}Ra in the sand-size fraction tends to decrease in the order 10–100, >250, and 100–250 μm (Fig. 4a). The highest percentage (26.7%) of the total ^{226}Ra activity in the chemically contaminated soddy–meadow soil is associated with 10–100 μm particles, and the lowest percentage (7%), with 5–10 μm particles. A large proportion of ^{226}Ra is associated with <1 μm particles. The contributions of the other particle-size fractions are almost identical (Fig. 4b).

Thus, the distribution of ^{226}Ra among the particle-size fractions and the contribution of the individual particle-size fractions to the total radionuclide activity are similar in both soil types. The highest specific activity is associated with the <10 μm fraction. In the anthropogenic podzolic and chemically contaminated soddy–meadow soils, the specific radionuclide activity decreases in the order clay > medium silt > fine silt. The ^{226}Ra concentrations in these particles are higher in the anthropogenic podzolic soil than in the chemically contaminated soddy–meadow soil. In both soil types, the greatest contribution to the total ^{226}Ra activity is associated with the 10–250 and <1 μm fractions. At the same time, the high Ra activity of clay-size particles can be attributed to their higher specific activity, and that of coarse silt and fine sand (10–250 μm), to their high mass fraction. In general, about 95% of the radionuclide activity in the anthropogenic podzolic soil is associated with <250 μm particles, while the contribution of coarser particles is negligible. In the soddy–meadow soil, the lowest contribution to the total Ra activity is associated with 5–10 μm particles.

It has long been recognized that the minerals of the clay fractions of soil play a crucial role in the accumulation of trace elements by soil-forming rocks and soils. The mineralogy of clay determines the physicochemical characteristics and chemical composition of soils. The most common clay minerals of soils and parent rocks are kaolinites, micas, hydromicas (illites), vermiculites, montmorillonites (smectites), chlorites, soil chlorites, and mixed-layer minerals. The role of clay minerals in determining the behavior of chemical elements in soils is related to their ability to adsorb elements from the water phase and fix them in exchangeable and nonex-

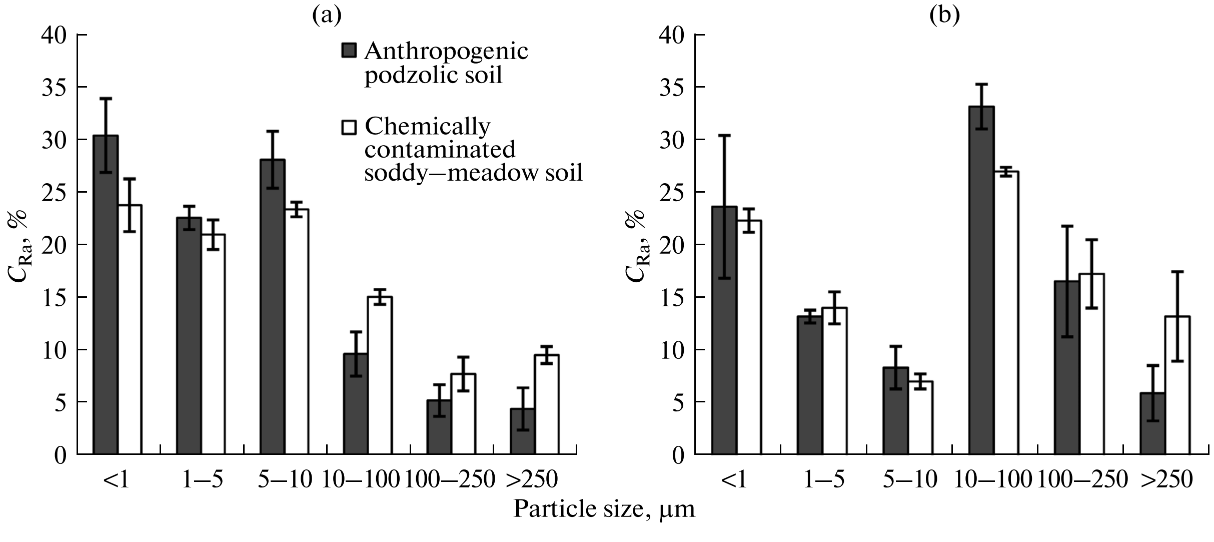

Fig. 4. (a) Relative percentages of ^{226}Ra in particle size fractions and (b) the contribution of each fraction to the total ^{226}Ra activity in different soil types.

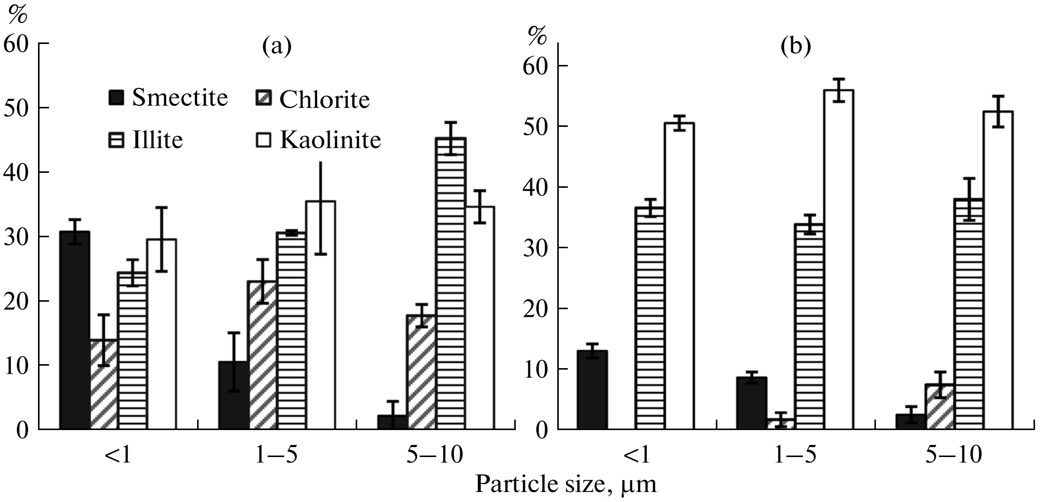

Fig. 5. Contents of minerals in the fine fractions of the (a) anthropogenic podzolic soil and (b) chemically contaminated soddy–meadow soil.

changeable states. Three-layer montmorillonite-group minerals have the greatest adsorption capacity, and two-layer kaolinites, the lowest. Illites take an intermediate position (Sokolova et al., 2005). The greatest sorption capacity was detected for the clay-size and colloid particles attached to organic matter, which forms different chemical bonds with soil mineral constituents. High molecular weight organic compounds (i.e., humates and fulvates) have relatively large sizes and peculiar shapes, which prevents their penetrating into the interlayer space of clay minerals; therefore, they are attached to the surface through adhesive and cohesive forces. Low molecular weight compounds derived by decomposition of organic matter can penetrate the interlayer space of clay minerals owing to intramicellar adsorption (Gorbunov, 1978).

It was shown that the 5–10 μm fraction is dominated by quartz. It becomes less abundant in the 1–5 μm fraction but still dominates over clay minerals. The highest proportion of clay minerals was detected in the <1 μm fraction, in which quartz is almost absent and does not mask other minerals. Despite the large difference in the amounts of clay minerals in different particle fractions, the concentrations of ^{226}Ra are almost identical in all three fractions.

The results of semi-quantitative X-ray diffraction analysis reveal differences in clay mineralogy among different size fractions of the same soil and among the same fractions of different soils. The analysis showed the presence of almost all clay mineral species in the clay-size fraction of the anthropogenic podzolic soil, and chlorite was only absent in some soil horizons of profiles 2 and 3. The highest content of chlorite (23.1%) was detected in the fine silt fraction. The percentage of smectite decreases from 30.8 to 2.2% with increasing particle size. The relative percentage of illite increases from 24.4 to 45.3% with increasing particle size. The proportion of kaolinite in these fractions is approximately the same (30–35%) within the error. The phyllosilicates of the 1–5 and 5–10 μm fractions are dominated by illite and kaolinite, accounting for 52–82% of the total of this mineral group. The clay-size fraction of the chemically contaminated soddy–meadow soil contains smectite, illite, and kaolinite. The proportion of smectite decreases in the bulk soil with increasing particle size. The relative percentage of smectite in these fractions is much lower than in the anthropogenic podzolic soil. Chlorite is present in the coarser fractions (1–5 and 5–10 μm) of some soil horizons (lowermost, in particular). The phyllosilicates of all fractions are illite and kaolinite, which account for 85–92% of the total clay minerals of each fraction. Therefore, the relative percentages of clay minerals are variable in all fractions analyzed and may change with depth. The average percentages of minerals in the clay fractions of soils studied are shown in Fig. 5.

No evident correlation exists between ^{226}Ra concentrations and the percentages of clay minerals in different fractions. Nevertheless, a negative correlation was established between radionuclide and chlorite concentrations in the 1–5 μm fraction of the anthropogenic podzolic soil (–0.67, $p \leq 0.05$) and 5–10 μm fractions of the chemically contaminated soddy–meadow soil (–0.56). No correlation with chlorite was found in other fractions. Chlorites have low cation-exchange capacity and low specific surface area. Because of the presence of an additional octahedral sheet between three-layer packets, the minerals of this group lack the ability to adsorb within the interlayer space because of the absence of an active internal surface. The absence of chlorite in the <1 μm fraction of soddy–meadow soil with the highest specific ^{226}Ra activity suggests that chlorite does not participate in radionuclide adsorption.

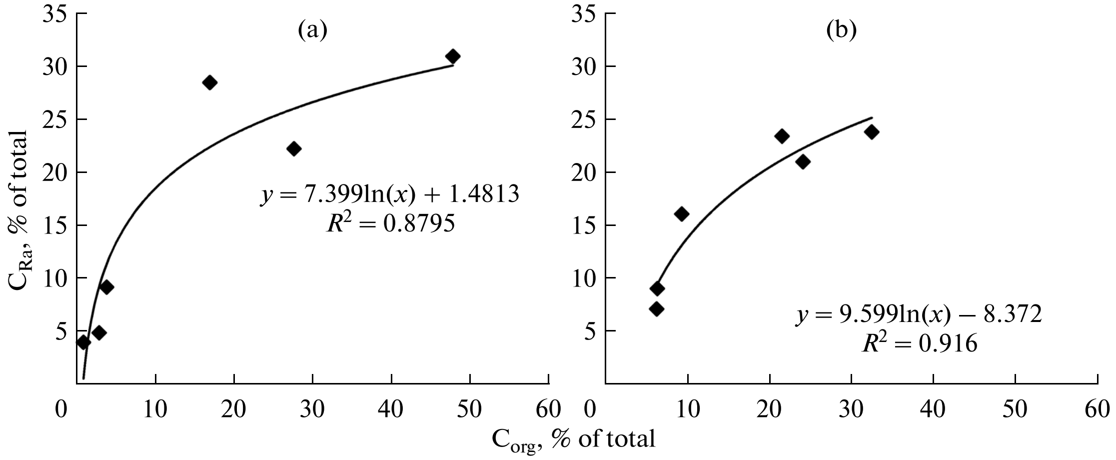

Fig. 6. Relationships between the concentrations of ^{226}Ra and organic matter in different size fractions of the (a) anthropogenic podzolic soil and (b) chemically contaminated soddy–meadow soil ($p \leq 0.05$).

For the 5–10 μm fraction of the soddy–meadow soil, a positive correlation (0.66, $p \leq 0.05$) was established between the contents of ^{226}Ra and smectite, which has very large surface area and high cation exchange capacity. Because of the low charges and a weak electrostatic bond between three-layer packets and interlayer cations, the minerals of this group are capable of retaining cations, water molecules, and many organic compounds through interlayer adsorption.

Moreover, similar to the specific ^{226}Ra activity, the organic matter concentration decreases with increasing particle size. A strong correlation was found between the contents of ^{226}Ra and organic matter in the anthropogenic podzolic and chemically contaminated soddy–meadow soil (0.862 and 0.925, $p < 0.01$, respectively). The regression analysis of the data indicates that the relations between the above parameters can be described best by logarithmic functions (Fig. 6). Thus, Ra concentrations in each particle size fraction increase with increasing organic matter content.

The correlation analysis of the contents of organic matter and relative percentages of clay minerals in each size fraction revealed a negative correlation for chlorite in the 1–5 and 5–10 μm fraction of the anthropogenic podzolic soil (–0.75 and –0.78, respectively, $p \leq 0.01$) and in the 5–10 μm fraction of the chemically contaminated soddy–meadow soil (–0.61, $p \leq 0.05$). A positive correlation was found for smectite in the 5–10 μm fraction of the anthropogenic podzolic soil (0.71, $p \leq 0.05$) and in the 5–10 μm fraction of the chemically contaminated soddy–meadow soil (0.69, $p \leq 0.05$), as well as for illite in all three size fractions of chemically contaminated soddy–meadow soil (0.56, 0.46, and 0.51 in the order of increasing particle size).

CONCLUSIONS

The mobility of radionuclides in anthropogenically contaminated soils is largely controlled by the soil physicochemical composition, which, in turn, depends on the type of soil contamination. At a site contaminated by solid radioactive materials, a significant correlation was established between the contents of radium and phosphorus, which is present at elevated levels in the tailings material. One of the most stable forms of the radionuclide absorption from the tailings material can be insoluble radium phosphate compounds. In the soddy–meadow soil, which was contaminated by produced water and has different physicochemical characteristics, complexing of Ra with organic matter reduces the mobility of the radionuclide.

The distribution of ^{226}Ra among particle size fractions in both types of soil (not considering the tailings layer in the anthropogenic podzolic soil) exhibits a similar trend. The greatest specific activity of the radionuclide is associated with clay particles. The concentration of ^{226}Ra in the clay particles is higher in the anthropogenic podzolic soil than in the chemically contaminated soddy–meadow soil.

The composition of soil fractions has a significant effect on the radionuclide mobility. For example, the distribution of ^{226}Ra and organic matter is similar in all size fractions. The high content of smectite and illite group minerals with the high interlayer sorption capacity is accompanied by enrichment in organic matter and ^{226}Ra. In contrast, the presence of chlorite, which has no active internal surface, decreases the concentrations of organic matter and radionuclide in the fine fractions. According to the data of Simon and Ibrahim (1987), organic matter is capable of absorbing approximately 10 times the amount of radium absorbed by clays. Our study shows that correlations between contents of organic matter and ^{226}Ra in the soil and soil fractions may indicate that the radionuclide is incorporated in a soil absorbing complex bound to organic compounds, which are sorbed either onto the surface or within the interlayer spaces of soil clay minerals. This may be the reason for the high specific activity of the radionuclide in the fine fractions of the anthropogenic podzolic soils, in which smectite and organic matter contents are higher than in the chemically contaminated soddy–meadow soil.

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