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Concentrations of Radionuclides (^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs) in Chernozems of Volgograd Oblast Sampled in Different Years

B. F. Aparin[a, b, *], **E. V. Mingareeva**[a, c], **N. I. Sanzharova**[c], **and E. Yu. Sukhacheva**[a, b]

[a] *Dokuchaev Central Museum of Soil, Birzhevoi pr. 6, St. Petersburg, 199034 Russia*
[b] *Institute of Earth Sciences, St. Petersburg State University, Universitetskaya nab. 7/9, St. Petersburg, 199034 Russia*
[c] *All-Russia Research Institute of Radiology and Agroecology,*
Kievskoe shosse 109 km, Obninsk, Kaluzhskaya oblast, 249032 Russia
**e-mail: soilmuseum@bk.ru*

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Abstract—Data on the concentrations of natural (^{226}Ra, ^{232}Th and ^{40}K) and artificial (^{137}Cs) radionuclides and on the physicochemical properties of chernozems sampled in different years are presented. In 1952, upon the creation of the Penza–Kamensk state shelterbelt, three deep (up to 3 m) soil pits were examined within the former arable field under two-year-old plantations of ash and maple along the transect crossing the territory of the Beloprudskaya Experimental Station of the USSR Academy of Sciences in Volgograd oblast. The samples from these pits were included into the collection of dated soil samples of the Dokuchaev Central Soil Science Museum. Five pits were examined along the same transect in 2009: three pits under shelterbelts (analogues of the pits studied in 1952) and two pits on arable fields between the shelterbelts. In the past 57 years, certain changes took place in the soil structure, bulk density, and the content and composition of humus. The salt profile of soils changed significantly under the forests. The comparison of distribution patterns of natural soil radionuclides in 1952 and 2009 demonstrated their higher contents at the depth of 10–20 cm in 2009 (except for the western shelterbelt). Background concentrations of natural radionuclides in parent materials and relationships between their distributions and the salt profiles of soils have been determined; they are most clearly observed is the soils under shelterbelts. Insignificant contamination with ^{137}Cs (up to 34 Bq/kg) has been found in the samples of 2009 from the upper (0–20 cm) horizon. The activity of ^{137}Cs regularly decreases from the east to the west; the highest concentrations of this radionuclide are found in the topmost 10 cm. This allows us to suppose that ^{137}Cs was brought with aerial dust by eastern winds, and the shelterbelts served as barriers to the wind flow.

Keywords: monitoring, spatial and temporal variability, mantle clays, parent rocks, soil collection

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INTRODUCTION

Nuclear tests, development of nuclear power engineering industry, and application of radiation sources in industry, medicine, and agriculture resulted in the change of radiation situation and appearance of territories with increased concentrations of natural radionuclides (NRN) and artificial radionuclides in the second half of XX century. Accident situations and incidents determined the formation of radioactive trails with high levels of contamination (emergencies at Scientific-Production Association Mayak, Chernobyl NPS, etc.). The main radionuclides of global fallout are two long-lived radionuclides ^{90}Sr ($T_{1/2}$ = 29.12 years) and ^{137}Cs ($T_{1/2}$ = 30.17 years), which were distributed on the Earth's surface after nuclear tests. The contamination caused by disruptions of the technological processes in the work of enterprises of nuclear fuel cycle, transportation, and storage of radioactive wastes, and accident situations produced by the fallout of various radionuclides [2, 21, 22]. Soil is the main environmental "depot" of radionuclides. Soil on one hand takes NRN from parent rocks, which determine natural radiation background of soil, and, on the other hand, it is the "depot" of artificial radionuclides [15, 24, 30]. Unique soil properties open possibilities for revealing pollution scenarios and observing the dynamics of the radiation situation development in different territories. Significant factual material has been accumulated by now on characteristics of soils contaminated by technogenic (artificial) radionuclides [2, 11, 18, 23, 25, 28, 31, 32]. It is known that the behavior of artificial radionuclides in soils depends on many factors: genetic features of soil, regimes (moisture, acidity, and redox), organic matter content, and particle-size, chemical, and mineralogical compositions [5–7, 15, 22, 24, 29, 30].

The following interrelated tasks should be solved in order to determine the danger level of soil contamination by radionuclides: (1) to assess recent concentrations of radionuclides in soils and parent rocks; (2) to study regularities of their behavior; and (3) to arrange monitoring of radionuclides concentrations in soils of natural zones and develop the prognostic models of radionuclides migrations in the environment.

The solving of these tasks is complicated for Russia by the following circumstances: exclusively great diversity of soils and parent rocks, time and scale of radiation pollution, and limited information on background concentrations of radionuclides (prior to nuclear tests) in soils and parent rocks. There are average data on NRN concentrations in some soils and sedimentary, igneous, and metamorphic rocks [17, http://www.phys.rsu.ru/web/nuclear/radioecologie/fRE6.htm]. The unique possibility to study concentrations of natural and artificial radionuclides in soils and parent rocks and temporal variations of their concentrations is provided by soil monoliths and soil samples from different regions of Russia stored in V.V. Dokuchaev Central Soil Museum [13, 33]. Soil monoliths and samples taken in different natural zones and land use types, on different parent rocks, and in different periods (beginning from 1902) allow characterizing natural radiation background in the soils, its dynamics, and input of technogenic radionuclides, i.e. carrying out soil-ecological monitoring.

The task of this work is the integrated study of variations in radionuclides (^{226}Ra, ^{232}Th, ^{40}K, ^{137}Cs) concentrations and of soil properties in the samples of chernozem under forest and cropland, and in parent rocks; samples were taken in different time.

OBJECTS AND METHODS

Studied objects were presented by soil samples taken from the pits described in 1952 and 2009 in the plots, soil cover of which was presented by carbonate-textural (southern) chernozem on mantle clays. The research was performed in the territory of former Beloprudskaya Station, Danilovskii district, Volgograd oblast, first by the Complex expedition of the USSR Academy of Sciences solving the problems of field-protective afforestation. The Station was established on gently sloping watershed divide of the Buzuluk and Tersa rivers. Parent rocks are presented by mantle silty clays of several tens of meters deep. Mean annual temperature is 5.4°C, annual precipitation is 390 mm. Precipitation to evaporation ratio in summer ranges from 0.32 to 0.23. Maximal water penetration into the soil in spring is about 180 cm [4]. E.A. Afanas'eva made three pits (pit 210, pit 209, and pit 211) to the depth of 3 m along the transect crossing an even interstream area from the west to the east on a cropland under the two-years-old plantations of ash and maple in 1952, the State Forest Protective Belt Penza—Kamensk was under construction. [4]. Forest belt was composed of three shelterbelts 60 m in width each and two inter-shelterbelt spaces 300 m in width occupied by arable plots. Hence, total width of the whole forest belt was 780 m [4]. Soil samples weighing about 1.5 kg, were taken from every 10-cm layer in three pits. Air-dry samples were stored in closed glass vessels with friction-fitted stoppers under room temperature in the collection of V.V. Dokuchaev Central Soil Museum.

Five pits were dug in the same place along the same transect to the depth 2.5 m in 2009. Three pits were adjusted to the same sampling plots as in 1952 (pit BP-1 analogous to pit 210, BP-2 analogous to pit 209, and BP-4 analogous to pit 211) under western, central, and eastern shelterbelts, respectively. The tree stand was composed of red ash (*Fraxinus pennsylvanica*) in the eastern shelterbelt and ash-like-leave maple (*Acer negundo)* in the central and western shelterbelts. Crown density was 0.4—0.5, tree height 12 m, tree age 57 years. Two pits were made in arable plots between shelterbelts (pit BP-5 between western and central shelterbelts and pit BP-3 between central and eastern shelterbelts). Density was determined in pits BP-1, BP-4, and BP-5 to the depth of 90 cm in every 5-cm layer. Soil samples of 2 kg were taken in the layers every 5—10 cm.

Soil sampling, preparing of averaged specimens from the samples taken in 1952 and 2009, and subsequent analysis were carried out according to the commonly adopted methods [8, 19, 26]. Organic carbon (humus) by I.V. Tyurin method, fractional-group composition of humus by I.V. Tyurin in modification of V.V. Ponomareva and T.A. Plotnikova, pH of water suspension, water-soluble substances (water extract), and particle-size composition by pipette method were determined.

Specific activities of radionuclides (^{226}Ra, ^{232}Th, ^{40}K, ^{137}Cs) were measured in the samples from the layers, cm: 0–5, 5–10 (0–10), 10–20 and 90–100 by gamma-spectrometry method in All-Russian Research Institute of Radiology and Agroecology according the method [1].

RESULTS AND DISCUSSION

Comparative characterization of soil properties in the samples taken in different years. Soils of all studied pits were formed on heavy-textured parent rocks (Table 1). Average content of physical clay (<0.01 mm) in all samples was 69.8% (standard deviation (σ) = 7.9%). Clay content at the depth of 90—100 cm varied in a narrow range: soils of 1952 – 37—39%, and those of 2009 – 35—42%.

Physical clay and clay (<0.001 mm) contents in the upper part of soil profiles (0—10 and 10—20 cm) were practically everywhere less than in parent rocks both in the samples of 1952 and 2009. A distinct decrease of physical clay (<0.01 mm) content at the same depths from the west to the east was observed in soils of 1952 (pits 209, 210, and 211), whereas an opposite trend was recorded for soils of 2009 year of sampling. The latter increase was especially well pronounced in pit BP-2 (analogous to pit 209) and pit BP-4 (analogous to pit 211). In this result, we observed leveling of physical clay content in all recent pits along the transect. The difference between soils of 1952 and 2009 in the layers 0–10 and 10–20 cm decreased from the east to the west. This could be connected with the input of fine particles by eastern winds, and capturing of these particles by protective forest belts. Similar trends were observed for the clay fraction.

Well-expressed humus-clay coatings (Munsell color 10YR 3.1) along the cracks and root holes were observed approximately in the depth interval 40 to 200 cm in soils of 2009. Coatings thickness ranged from 1 to 3 mm. Their number and size changed with the depth. The presence of coatings could be indicative of illuviation process of humus substances and clay particles in the soil profile. The presence of numerous tongs at the lower boundary of humus horizon in recent soil is an indirect evidence of this process.

The thickness of humus horizons in soils did not significantly change over a 57-years period. Pronounced adverse changes occurred in soil structure of upper horizons in arable soils (pits BP-3 and BP-5) [9]: subangular blocky and crumb structure was transformed into cloddy and even lumpy one. It was most pronounced in the lower part of plow horizon, where the ped faces were most clearly expressed.

According to the data of Afanas'eva [4], soils of 1952 were characterized by a uniform increase of density from the surface to the depth of 90 cm from 1.0 to 1.6 g/cm^3. Significant change of bulk density occurred in the layer 0–5 cm over the studied period. Density decreased in the soil under forest plantations to 0.8 g/cm^3, and, on the contrary, it increased under arable field to 1.3 g/cm^3. In the whole, bulk density in recent soils ranged within 0.8–1.4 g/cm^3 under forest and within 1.3–1.5 g/cm^3 under cropland.

Content of carbon of organic compounds (C_{org}) in recent arable soils (pits BP-3 and BP-5) did not practically change over the studied period. At the same time, in soils under forest (pits BP-1, BP-2, BP-4), C_{org} content in the layer 0–10 cm increased to 4.4%, and this was 1.4–1.6% higher as compared with the soil 1952. Minimal difference was observed in the pair of pits 211 and BP-4 (eastern shelterbelt), where initially (pit 211) C_{org} content was higher. Similar phenomenon was observed in the layer 10–20 cm. Significant changes occurred in the group composition of humus: the proportions of fulvic acids and humins (nonhydrolyzable residue) increased. Humate type of humus was replaced by fulvate-humate type according to the parameters of the humus state of soil [14].

Soil pH was close to neutral in the upper part of the soil profile (0–55 cm) in soils of both dates of sampling. Minimal value (pH 6.8) was observed at the depth of 10–20 cm (Table 1). Soil pH sharply increased below 55 cm in all soils and exceeded 8.1. The same values were observed in soils of 1952, although by 30 cm lower than in recent soils.

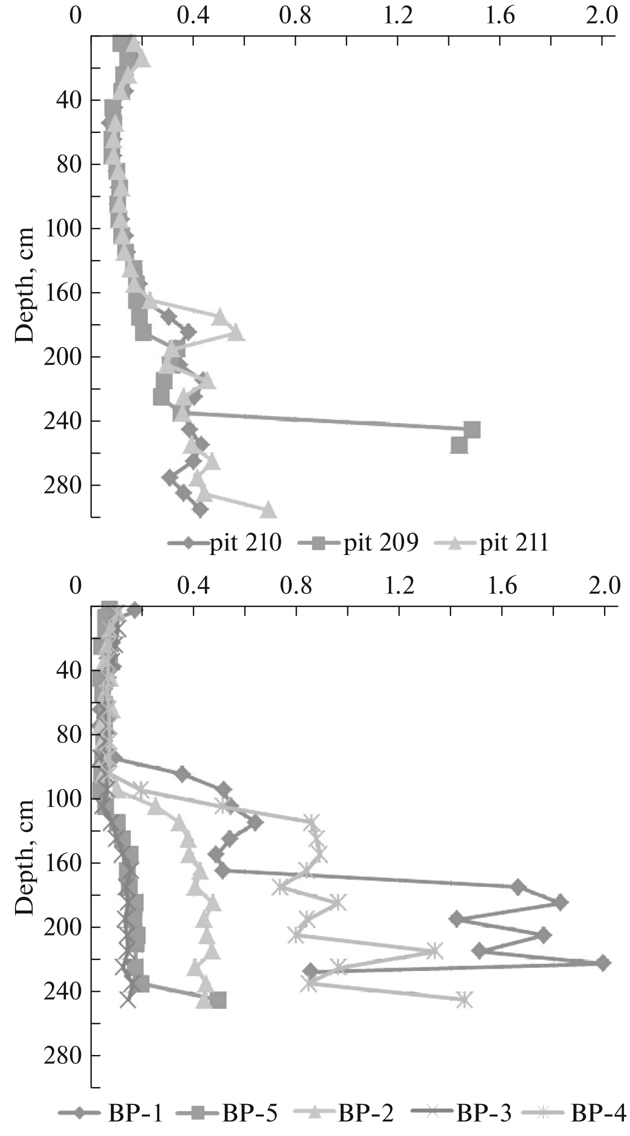

Fig. 1. Dry residue in soils of different time of sampling.

Significant changes occurred in salt profiles of soils over 57 years. The upper boundary of salt profile (dry residue 0.2% was taken as its initial point) raised from 175–200 cm (1952) to 100–140 cm (2009) (Fig. 1). Maximal value of dry residue increased from 0.4% (1952) to 1.0–1.4% in soil of eastern shelterbelt and to 2.0% in soil of western shelterbelt (2009). Concentrations of easily soluble salts insignificantly increased in the soil of central shelterbelt. The dry residue in recent arable soils changed slightly all over the soil profiles (to 240 cm) and was about 0.1% in the upper one-meter layer and did not exceed 0.2% in the lower layers.

Table 1. Chemical and physicochemical properties of studied chernozems

Horizon	Depth, cm	pH_{water}	C_{org}, %	Particles <0.01 mm	Particles <0.001 mm
			%		
Pit 210, 1952, western shelterbelt					
PU	0–10	6.9	2.8	64	32
PU	10–20	6.8	2.6	64	30
AU_{ca}	20–30	7.1	2.4	39	32
AU_{ca}	30–40	7.8	2.0	67	30
CAT	40–50	8.0	1.5	70	32
CAT	50–60	7.7	1.2	70	32
CAT	60–70	7.8	0.9	75	35
C_{ca}	70–80	7.9	0.7	72	36
C_{ca}	80–90	8.1	0.7	72	39
C_{ca}	90–100	8.2	0.4	73	39
Pit BP-1, 2009, western shelterbelt					
AU	0–5	7.0	4.4	70	32
AU	5–10	6.9	3.2	76	37
AU	10–20	6.8	2.8	75	32
AU_{ca}	20–30	7.7	2.6	74	33
AU_{ca}	30–40	8.0	2.1	71	32
CAT	40–50	8.2	1.1	79	37
CAT	50–60	8.2	1.2	78	36
CAT	60–70	8.4	0.7	77	36
C_{ca}	70–80	8.4	0.6	77	38
C_{ca}	80–90	8.4	0.5	81	39
C_{ca}	90–100	8.3	0.4	81	38
Pit BP-5, 2009, arable field between shelterbelts					
PU	0–5	7.0	2.7	73	33
PU	5–10	7.0	2.6	73	34
PU	10–20	7.0	2.4	74	35
AU_{ca}	20–30	7.2	2.1	72	34
CAT	30–40	7.8	1.8	74	34
CAT	40–50	8.2	1.7	69	39
CAT	50–60	8.3	1.2	73	35
CAT	60–70	8.4	1.0	74	36
CAT	70–80	8.4	1.0	76	41
C_{ca}	80–90	8.4	0.5	71	46
C_{ca}	90–100	8.4	0.5	73	42
Pit 209, 1952, central shelterbelt					
PU	0–10	7.5	2.9	58	16
PU	10–20	7.7	2.8	59	20
AU	20–30	7.8	2.6	65	25
AU	30–40	8.2	2.2	68	29
AU	40–50	8.2	2.1	73	37

Table 1. (Contd.)

Horizon	Depth, cm	pH$_{water}$	C$_{org}$, %	Particles	
				<0.01 mm	<0.001 mm
			%		
CAT	50–60	7.9	1.2	70	36
CAT	60–70	8.1	1.0	71	35
CAT	70–80	8.3	0.7	72	37
CAT	80–90	8.4	0.6	72	39
C$_{ca}$	90–100	8.4	0.4	73	37
Pit BP-2, 2009, central shelterbelt					
AU	0–10	7.4	4.0	67	33
AU	10–20	7.0	3.1	69	36
AU	20–30	6.8	2.6	71	36
AU	30–40	6.8	2.4	69	35
CAT	40–50	7.6	1.7	70	37
CAT	50–60	8.1	1.7	70	35
CAT	60–70	8.3	1.1	70	36
CAT	70–80	8.5	0.7	74	38
CAT	80–90	8.4	0.7	71	35
C$_{ca}$	90–100	8.5	0.5	70	35
Pit BP-3, 2009, arable field between shelterbelts					
PU	0–10	7.9	2.7	73	33
PU	10–20	7.9	2.5	74	35
AU$_{ca}$	20–30	7.9	2.5	Not det.	
CAT	30–40	8.0	1.9	″	
CAT	40–50	8.3	1.4	″	
CAT	50–60	8.4	0.9	″	
C$_{ca}$	60–70	8.4	0.6	″	
C$_{ca}$	70–80	8.5	0.5	″	
C$_{ca}$	80–90	8.5	0.3	″	
C$_{ca}$	90–100	8.6	0.4	73	42
Pit 211, 1952, eastern shelterbelt					
PU	0–10	7.0	3.3	50	21
PU	10–20	6.9	3.3	51	22
AU$_{ca}$	20–30	7.2	2.3	68	33
AU$_{ca}$	30–40	7.3	1.9	64	32
CAT	40–50		1.2	67	32
CAT	50–60	7.8	1.2	68	32
CAT	60–70	8.0	0.7	71	32
CAT	70–80	8.0	0.5	72	41
C$_{ca}$	80–90	8.1	0.4	72	38
C$_{ca}$	90–100	8.1	0.3	71	40

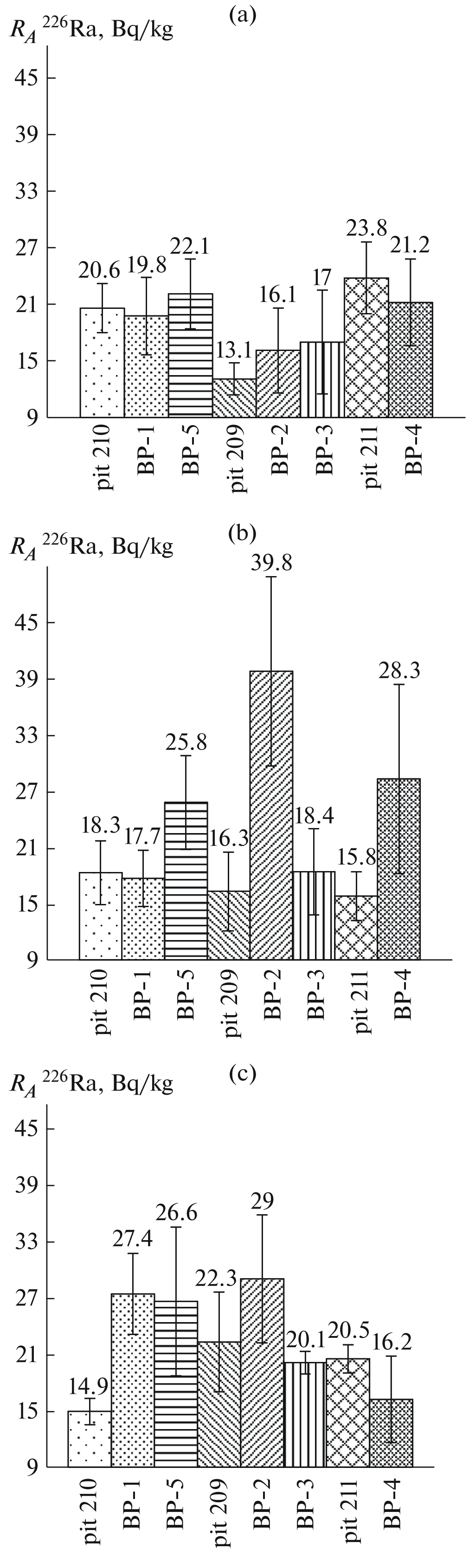

Fig. 2. Specific activity of ^{226}Ra in the layers of (a) 0–10, (b) 10–20, and (c) 90–100 cm. Range of values in bar charts characterizes the errors of radiometric measurements.

The composition of salts also changed over this period. Sodium sulfate Na_2SO_4 was dominating salts in the soil in 1952 [4]. Concentrations of calcium and magnesium sulfates and to a lesser degree of sodium chloride increased in recent soils. Layer with maximum concentration of chlorides in the soil profile was by 40 cm higher than that of sulfates.

Afanas'eva [4], performed in Beloprudskaya Station a complex study of soil properties, water regime, and salt profiles in 1950–1953; she concluded that the character of soil water regime could be inferred by the distribution of gypsum and easily soluble salts in soil-rock layer. Trees can cause the upward flows of moisture with dissolved salts due to strong and deep root systems [4].

Characteristic features of salt profiles in soils 2009 could be explained by the rise of easily soluble salts in soils under forest and on the contrary by their removal from two-meter layer of arable soils. This could be connected with the influence of protective forest belts on soil moisture regimes. Drying of the middle part of the soil profile occurred probably in marginal shelterbelts. It was accompanied by ascending moving of moisture from deep layers with associated moving of easily soluble salts. Maximal accumulation of these salts was at the depth of 170–230 cm, and their upper boundary rose to 100 cm.

Easily soluble salts were removed below 240 cm in arable soils between shelterbelts due to the effect of forest on moisture circulation in the studied territory.

Concentrations of radionuclides in the soil samples taken in different years. When analyzing the activity of NRN, we adhered to the following schedule: concentrations of each NRN were compared by layers for the totality of samples, and then separately background and recent samples were compared with each other. Obtained data were analyzed in the upward order—from the parent rock to the humus horizon. Then, land use types were also compared.

Radium. The range of specific activity (R_A) ^{226}Ra values in parent rock (90–100 cm) in all samples was relatively narrow, 14.9–29.0 Bq/kg (mean (M) = 22.1 Bq/kg) (Fig. 2), and this range was narrower in 1952 (19.2–23.9 Bq/kg) in comparison with the samples 2009, whereas mean values were relatively close. Variation coefficients (V_σ) in the samples of two dates of sampling were 22.8 and 28.7%, respectively. It was taken, basing on literature data [10, 16] and statistical analysis of obtained material that V_σ exceeding the 30% threshold was the index of heterogeneity of radionuclide activity.

The range of R_A ^{226}Ra in the layer 10–20 cm was wider (15.8–39.8 Bq/kg) than in parent rock and was determined by radionuclide content in the samples 2009. The difference between ^{226}Ra activity at the depth 10–20 cm and in parent rock did not exceed 11 Bq/kg in all pits. This was also true for soil of central shelterbelt (pit BP-2), where specific activity of ^{226}Ra was maximal (depth 10–20 cm) and comprised 39.8 Bq/kg. Activity of ^{226}Ra at this depth in samples 1952 was practically the same in all pits. Variation coefficients calculated for the samples 1952 was 19%, whereas it was 44% for the samples 2009. Variation coefficients V_σ for arable soils 2009 were less than 27%, whereas it equaled 42% under the forest (at the same depth). It can be assumed that this difference may be attributed to the spatial heterogeneity.

The range of radium activity was the narrowest in the layer 0–10 cm (13.1–23.8 Bq/kg, M = 18.0 Bq/kg) in comparison with the lower layers. There was not practically no difference between the samples of two dates (within the instrument accuracy). The character of variation of radionuclide activity along the transect under forest actually repeated the variation pattern in soils 1952. Insignificant deviations were observed in arable soils.

In the whole, the samples of humus horizons (0–20 cm) were characterized in 2009 by the widest range of R_A (13.1–39.8 Bq/kg, M = 21.1 Bq/kg). Coefficient of variation for all measurements of R_A in the samples of humus horizons equaled 36.2%; it was less than 30% in the samples 1952.

Soils under shelterbelts (pits BP-1, BP-2, and BP-4) had wider ranges of radionuclide concentrations (13.2–39.8 Bq/kg, M = 23.5 Bq/kg) in comparison with the soils of arable fields (13.1–26.6 Bq/kg, M = 19.9 Bq/kg). Coefficient of variation exceeded the 30% threshold in soils under forest, and the maximal value was observed at the depth 10–20 cm (42%).

Thorium. Specific activity of ^{232}Th in all samples of parent rock (90–100 cm) ranged in 2009 within 32.4–47.5 Bq/kg (M = 38.5 Bq/kg) (Fig. 3). Activity R_A in the samples 1952 was practically similar, it varied within the measurement accuracy (35.1–38.1 Bq/kg, M = 36.5 Bq/kg). Coefficient of variation did not exceed 21% for the samples of both dates of sampling as well as for the totality of parent rock samples.

The character of changes of ^{232}Th activity in the samples of parent rock 2009 along the transect in comparison with the values of 1952 was similar to that of ^{226}Ra. High concentration of ^{232}Th in recent soil of western shelterbelt (pit BP-1) and low one in soil of eastern shelterbelt (pit BP-4) were recorded.

The R_A value of thorium in the layer 10–20 cm of the samples 2009 was greater than in 1952 with narrow range (37.3–44.5 Bq/kg). The difference between ^{232}Th activities at the depth of 10–20 cm and in parent rock did not exceed 12 Bq/kg in all pits.

Specific activity of ^{232}Th in surface layer (0–10 cm) was lower in the samples 2009 than in 1952: M = 38.9 and 45.9 Bq/kg, respectively. The samples of central shelterbelt (pit 209 and pit BP-2), in which the R_A values were similar, were exclusions.

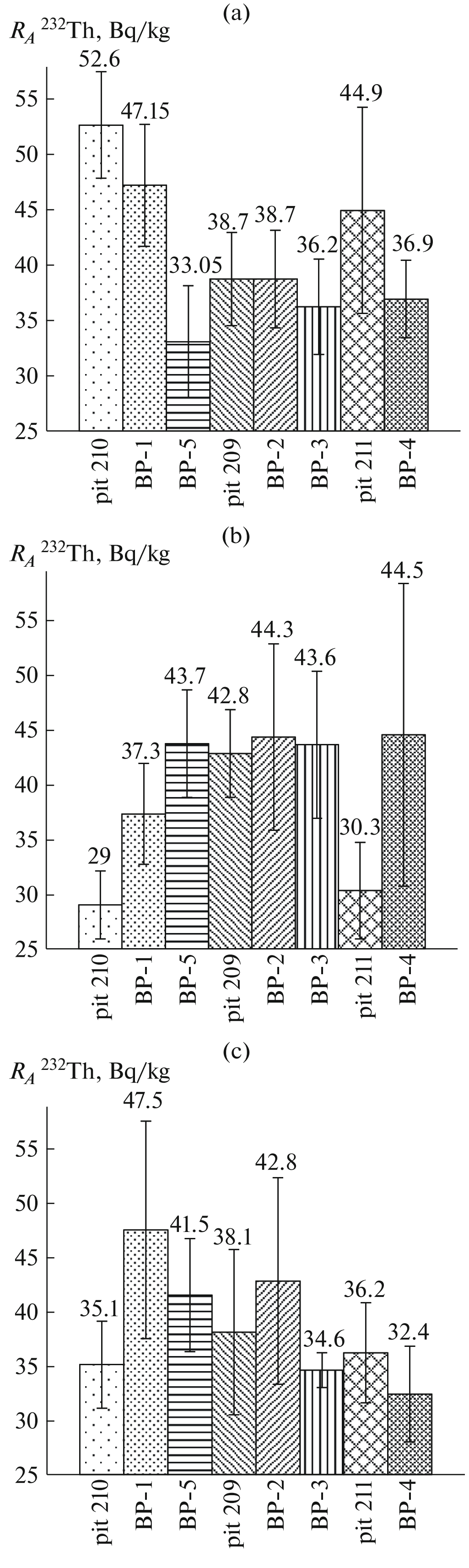

Fig. 3. Specific activity of ^{232}Th in the layers of (a) 0–10, (b) 10–20, and (c) 90–100 cm.

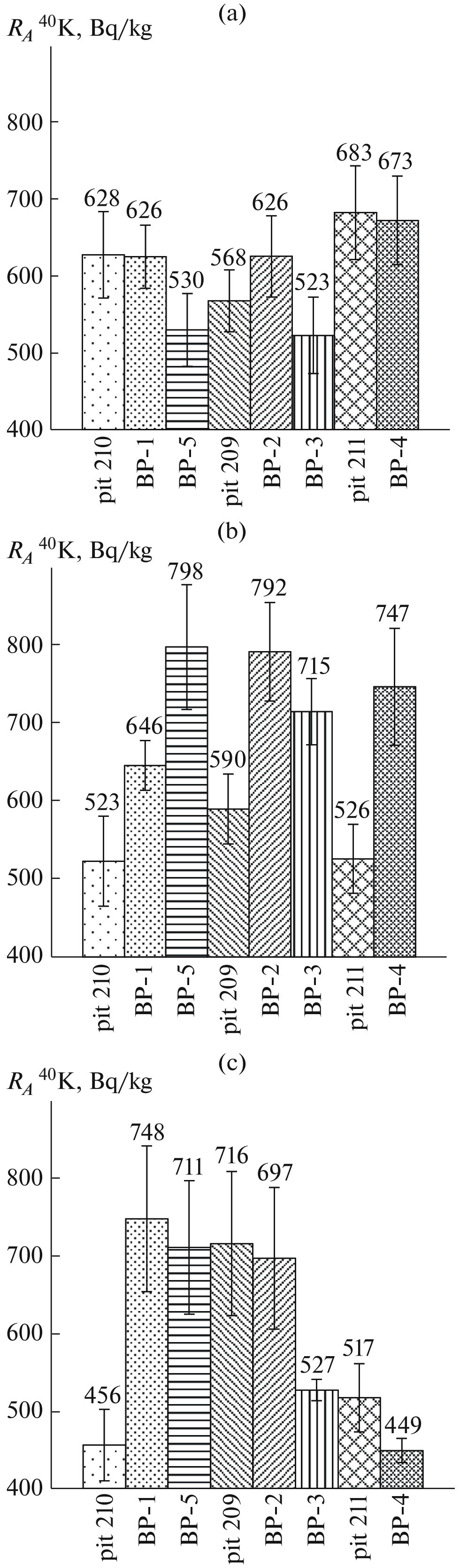

Fig. 4. Specific activity of ^{40}K in the layers of (a) 0–10, (b) 10–20, and (c) 90–100 cm.

The samples of humus horizons (0–20 cm), as in the case with ^{226}Ra, were characterized by a widest range of R_A of thorium (29.0–52.6 Bq/kg, M = 40.2 Bq/kg). This range was determined by the activity of ^{232}Th in soil samples of 1952 year of sampling. Coefficient of variation for all samples of humus horizons for both dates of sampling did not exceed 30%.

The following features in the character of change of thorium activity in soil profile should be emphasized: (1) minimum content of radionuclide in soils of 1952 (pit 210 and pit 211) was observed at the depth 10–20 cm in comparison with parent rock and the layer 0–10 cm; concentrations of thorium in pit 209 varied at all depths within the measurement accuracy; (2) similar varying of radionuclide activity along the transect was observed in the samples of 2009 at the depth 0–10 and 90–100 cm: the activity of radionuclide decreased from the west to the east. Unlike ^{226}Ra, soils under shelterbelts of forest belt were characterized by narrower ranges of ^{232}Th concentrations (32.4–48.3 Bq/kg, M = 41.9 Bq/kg), than soils under arable fields (29.0–52.6 Bq/kg, M = 36.9 Bq/kg).

Potassium. The ^{40}K radionuclide originated million years ago, and its presence in rocks and soils can be explained by a long period of half-life (1.248 × 10^9 years) [23]. The range R_A ^{40}K for all samples of parent rock (Fig. 4) was wide and equaled 449–748 Bq/kg (M = 603 Bq/kg). There were no significant differences between the ranges of specific activity of potassium in the samples of both dates of sampling, and this was confirmed by calculation of V_σ (separately for the samples of each date of sampling and for the totality), which did not exceed 23%.

Higher concentration of radionuclide in the layer 90–100 cm in soil of western shelterbelt (pit BP-1) relative to the value in the samples of 1952 (pit 210) should be noted. The difference was ~280 Bq/kg. However, a significant difference in ^{40}K activity was not observed in two other pairs of pits (209 and BP-2 and 211 and BP-4).

Activity of ^{40}K at the depth of 10–20 cm in pits BP-2, BP-3, BP-4, and BP-5 of 2009 varied in a narrow range (715–798 Bq/kg, M = 739 Bq/kg). Only pit. BP-1 stood apart with its lower value of activity, 646 Bq/kg. The samples of 1952 also displayed narrow ranges similar to those of 2009 with lower activity (523–590 Bq/kg, M = 546 Bq/kg). In the whole, the difference in radionuclide concentrations between the samples of two dates of sampling increased at this depth from the west to the east (from 120 to 220 Bq/kg).

The range of ^{40}K activity in the layer 0–10 cm in the samples of 1952 and 2009 was wider (523–673 Bq/kg, M = 590 Bq/kg) than in lower layer. It should be noted that the difference in radionuclide activity at this depth between the samples of two dates was practically insignificant. Maximal difference between the pit 209 and its analog – pit BP-2 was 60 Bq/kg. Potassium concentrations in arable soils 2009 (the layer 0–10 cm) were practically similar (523 and 530 Bq/kg) and lower than in the samples 1952 and those under the forest in 2009. The difference in activity did not exceed 50 Bq/kg in soil samples under the forest.

In the whole, humus horizons (0–20 cm) of soil were characterized in 2009 by wider range of R_A ^{40}K (523–798 Bq/kg, M = 652 Bq/kg). The range was clearly narrower in samples of 1952: 523–683 Bq/kg (M = 586 Bq/kg). Coefficient of variation for all samples from humus horizons and for each date separately did not exceed 18.1%.

The increase of ^{40}K activity was observed in the samples of pit 210 in comparison with parent rock by 70 Bq/kg in the layer 10–20 cm and by 170 Bq/kg in the layer 0–10 cm. The difference between parent rock and the layer 10–20 cm was insignificant (9 Bq/kg) in pit 211, higher activity characterized only the layer 0–10 cm. Opposite pattern was observed in pit 209 (central shelterbelt): activity of ^{40}K was significantly lower in upper horizons than in the parent rock. Analysis of ^{40}K activity in soil profiles of shelterbelts of 2009 year of sampling demonstrated that the activity in pit BP-1 (western shelterbelt) in the layers 10–20 and 0–10 cm was significantly lower than in the parent rock (by 100 and 123 Bq/kg, respectively). Activity R_A ^{40}K in the soil of eastern shelterbelt (pit BP-4) was significantly greater in both layers (0–10 and 10–20 cm). Specific activity of radionuclide in central shelterbelt (pit BP-2) was practically similar in the layer 0–10 cm and the parent rock and greater in the layer 10–20 cm than in the parent rock.

Cesium. Artificial radionuclide ^{137}Cs, which input on the soil surface was caused by global fallout after nuclear tests, was found only in soil samples 2009 at the depth of 0–10 and 10–20 cm (Fig. 5). The range of its concentrations at these depths was relatively wide: from 2.8 to 34.1 Bq/kg (M = 13.4 Bq/kg). The range R_A ^{137}Cs at the depth of 10–20 cm was significantly narrower (2.8–13.9 Bq/kg), and the absolute values smaller than at the depth of 0–10 cm in the same pits. Similar character of the changes in soil activity was observed at this depth: decrease of R_A from the east to the west as in the surface layer. Coefficient of variation for data totality was very high (86.6%) and comprised 67.1 and 59.4% for the depths 0–10 and 10–20 cm, respectively.

The highest concentrations of ^{137}Cs at the depth of 0–10 cm was found in soils of central and eastern shelterbelts (R_A = 31.9 and 34.1 Bq/kg respectively). The R_A ^{137}Cs value in soil of western shelterbelt was the same as in soils under arable fields (R_A = 9.0, 10.4 and 10.6 Bq/kg, respectively). Coefficient of variation V_σ for soils under forest equaled 61.6% in each layer, whereas V_σ was significantly lower in soils of arable fields (52.2% in the layer 10–20 cm and 20.1% in the layer 0–10 cm).

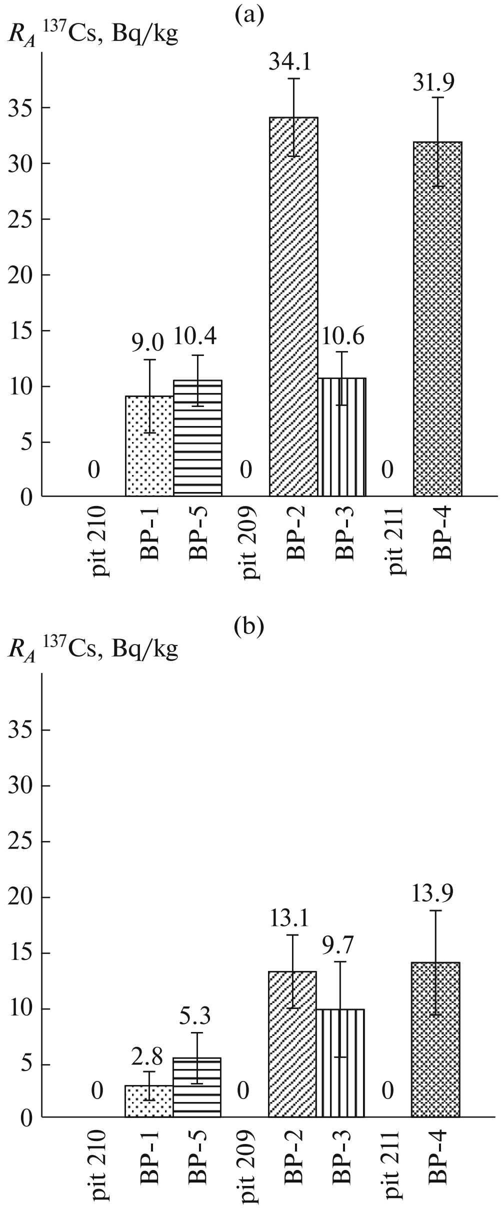

Fig. 5. Specific activity of ^{137}Cs in the layers of (a) 0–10 and (b) 10–20 cm.

CONCLUSIONS

Significant qualitative and quantitative changes occurred in soils of former Beloprudskaya Station, the USSR Academy of Sciences, from 1952, and this could affect the redistribution of NRNs.

The contents of clay and physical clay increased in the upper part (0–20 cm) of the profile of recent soils over the 57-years period. The difference between the contents of these fractions in soils in 1952 and in 2009 decreased along the transect from the east to the west. Humus content decreased, the structure changed for the worse, and density increased in arable soils. The density on the contrary decreased in soil under forest, humus content increased, and humus type changed from humate to fulvate-humate due to increased content of fulvic acids.

Pronounced trends were observed in the distribution of NRNs in soils over the studied period.

The comparison of NRN activities in pair samples of parent rocks 1952 and 2009 (pits 210 and BP-1, 209 and BP-2, and 211 and BP-4) demonstrated that significant changes in the character of activity distribution along the transect happened in all samples. The difference between NRN activities in the samples of 2009 (pit BP-1) and samples of 1952 (pit 210) reached maximal values in parent rocks of western shelterbelt. Activity of NRN was greater in samples 2009. The difference between activities in samples of two dates of sampling (pits 209 and BP-2) decreased in the central shelterbelt. The difference was minimal in the eastern shelterbelt (pits BP-4 and 211).

We determined the background values of NRN concentrations in the parent rocks (mantle loams and clays) and relatively low variability of absolute values of specific activity of radionuclides (R_A, σ) and of mean values (M):

$$^{226}\text{Ra M} = 19.2 \text{ Bq/kg},$$
$$R_A \text{ } 14.9\text{–}22.3 \text{ Bq/kg}, \sigma = 4.4 \text{ Bq/kg};$$

$$^{232}\text{Th M} = 36.5 \text{ Bq/kg},$$
$$R_A \text{ } 35.1\text{–}38.1 \text{ Bq/kg}, \sigma = 5.0 \text{ Bq/kg};$$

$$^{40}\text{K M} = 563 \text{ Bq/kg},$$
$$R_A \text{ } 456\text{–}716 \text{ Bq/kg}, \sigma = 130.5 \text{ Bq/kg}.$$

Activities R_A of ^{232}Th and ^{40}K in the samples of arable soils 2009 at the depth of 0–10 cm were lower than in forest soils, but clear difference between soils with different land use types was not observed for ^{226}Ra.

It was determined in numerous studies [2, 3, 6, 7, 11, 12, 15, 20, 22, 28, 29, 31] that NRNs from technogenic sources were firmly absorbed in the upper soil layers. Another character of distribution of radionuclides observed in studied soils suggested their natural origin. The position of the transect on one landform, relatively close positions of pits and the ratio of particle-size fractions, permit to conclude that the parent rocks belonged to the same genetic type, namely, mantle clays and loams. It can be believed that NRN were inherited from parent rock, and their redistribution was connected with soil processes.

The character of profile change of NRN specific activities demonstrated that relatively high concentrations of NRNs in the layer 10–20 cm were observed in all pits, excluding BP-1 (western shelterbelt). At the same time, maximal increase of R_A of all NRNs in parent rock in comparison with those in 1952 was recorded in this pit. It was in agreement with the character of changes in the soil salt profile. It was the recent soil of western shelterbelt (pit BP-1), where maximal increase of salt content and highest boundary of salt profile were recorded.

The samples from the upper part of the humus horizon could be divided into two groups by the value of ^{137}Cs activity. First group included soils with maximal activities of radionuclides from eastern and central shelterbelts. The second compact group included all other soils. Soil samples from the western shelterbelt had minimal activity at the depth of 0–10 cm. Input of ^{137}Cs on the soil surface was the result of global fallout. However, redistribution of ^{137}Cs within the studied territory was apparently connected with wind transfer, and forest belt served the barrier for winds.

The presence of ^{137}Cs at the depth of 10–20 cm permit to suggest that the redistribution of this radionuclide is the result of vertical migration caused by natural processes, especially in soils of the first group.

Soil collection and materials of soil studies obtained practically at the initial period of nuclear tests along with the data on the changes of soil properties and activities of radionuclides over the period from 1952 to 2009 allow considering former Beloprudskaya Station, the USSR Academy of Sciences, as a unique object for soil-ecological monitoring in the southeastern part of European territory of Russia.

REFERENCES

1. *Activity of Radionuclides in Countable Samples. Methodology of Measurements on Gamma Spectrometers Using the SpectraLine Software* (Mendeleevo, 2014) [in Russian].
2. R. M. Aleksakhin, N. I. Sanzharova, S. I. Spiridonov, and S. V. Fesenko, "Chernobyl and the environment," Radiats. Biol., Radioekol. **47** (2), 196–203 (2007).
3. N. P. Arkhipov, V. P. Mamedov, L. A. Grishina, and T. A. Fedorova, "Behavior of natural technogenic radionuclides in soils," Ekologiya, No. **1**, 31–38 (1982).
4. E. A. Afanas'eva, *Water-Salt Regime of Ordinary and Southern Chernozems in the Southeast of the European Part of the Soviet Union* (Nauka, Moscow, 1980) [in Russian].
5. A. O. Barsukov and D. V. Yazykeev, "Horizontal and vertical migration of ^{40}K, ^{137}Cs, ^{226}Ra, ^{232}Th, and ^{241}Am in agrolandscapes on cultivated slopes of different steepness in Penza oblast," Izv. Penz. Gos. Pedagog. Univ. im. V.G. Belinskogo, No. **29**, 369–374 (2012).
6. M. K. Bogatova, A. I. Shcheglov, and O. B. Tsvetnova, "Spatial heterogeneity of radioactive pollution of soils in forest phytocenoses of Tula oblast," Aktual. Probl. Lesn. Kompl., No. 10, 78–81 (2005).
7. E. A. Buraeva, V. S. Malyshevskii, T. V. Varduni, E. I. Shimanskii, A. N. Tribolina, A. A. Goncharenko, L. Yu. Goncharova, V. S. Totskaya, and V. S. Nefedov, "Content and distribution of natural radionuclides in different soils types in Rostov oblast," Sovrem. Probl. Nauki Obraz., No. 4, 1–9 (2013).
8. L. A. Vorob'eva, *Theory and Practice of the Chemical Analysis of Soils* (GEOS, Moscow, 2006) [in Russian].
9. P. D. Gurin, B. F. Aparin, and E. Yu. Sukhacheva, "Influence of forest plantations and long-term agricul-

tural use on the properties of southern chernozems," Vestn. S.-Peterb. Gos. Univ. **3** (2), 109–119 (2012).

10. E. A. Dmitriev, *Mathematical Statistics in Soil Science* (Moscow State Univ., Moscow, 1995) [in Russian].
11. A. V. Konoplev, V. N. Golosov, V. I. Yoschenko, K. Nanba, Y. Onda, T. Takase, and Y. Wakiyama, "Vertical distribution of radiocesium in soils of the area affected by the Fukushima Dai-ichi nuclear power plant accident," Eurasian Soil Sci. **49**, 570–580 (2016).
12. S. V. Lukin and A. F. Greber, "Distribution pattern of radionuclides in soil profiles of Belgorod oblast," Dostizh. Nauki Tekh. APK, No. 1, 19–20 (2008).
13. E. V. Mingareeva, "Use of a collection of monoliths to study the natural radiation background of Russian soils, *Proceedings of International Scientific Conference (XVI Dokuchaev Conference of Young Scientists) "The Laws of Soil Science: New Challenges"* (Moscow, 2013), pp. 195–198.
14. D. S. Orlov and L. A. Grishina, *Practical Manual on Humus Chemistry* (Moscow State Univ., Moscow, 1981) [in Russian].
15. F. I. Pavlotskaya, *Migration of Radioactive Products of Global Emissions in Soils* (Atomizdat, Moscow, 1974) [in Russian].
16. A. I. Popov, V. M. Igamberdiev, and Yu. V. Alekseev, *Statistical Analysis of Experimental Data* (St. Petersburg State Univ., St. Petersburg, 2009) [in Russian].
17. *Natural Radiation Background: Origin and Evolution* (Southern Federal Univ., Rostov-on-Don, 2007) [in Russian]
18. *Radiation Situation in Russia and Adjacent Countries in 2013: Yearbook* (Obninsk, 2014) [in Russian].
19. O. G. Rastvorova, *Soil Physics* (Leningrad State Univ., Leningrad, 1983) [in Russian].
20. N. G. Rachkova, I. I. Shuktomova, and A. I. Taskaev, "The state of natural radionuclides of uranium, radium, and thorium in soils," Eurasian Soil Sci. **43**, 651–658 (2010).
21. N. I. Sanzharova, A. A. Sysoeva, N. N. Isamov Jr., R. M. Aleksakhin, V. K. Kuznetsov, and T. L. Zhigareva, "Role of chemistry in rehabilitation of agricultural resources affected by radioactive pollution," Ross. Khim. Zh. **49** (3), 26–34 (2005).
22. A. N. Silant'ev, I. G. Shkuratova, and Ts. I. Bobovnikova, "Vertical migration of Chernobul-derived radionuclides in soils," At. Energiya **66** (3), 194–197 (1989).
23. N. A. Titaeva, *Nuclear Geochemistry* (Moscow State Univ., Moscow, 2000) [in Russian].
24. A. A. Titlyanova, "Cesium sorption by layered soil minerals," Pochvovedenie, No. **12**, 88–94 (1964).
25. A. D. Fokin, A. A. Lur'e, and S. P. Torshin, *Agricultural Radiology* (Lan', St. Petersburg, 2011) [in Russian].
26. *Chemical Analysis of Soils* (St. Petersburg State Univ., St. Petersburg, 1995) [in Russian].
27. O. B. Tsvetnova and A. S. Novikova, "Radioecological situation in the forest and agricultural ecosystems of Tula oblast," Aktual. Probl. Lesn. Kompl., No. 13, 253–257 (2006).
28. "Environmental consequences of the Chernobyl accident and their remediation: twenty years of experience," *Report of the Chernobyl Forum Expert Group "Environment"* (International Atomic Energy Agency, Vienna, 2006), p. 166.
29. S. Forsberg, A. Rosen, V. Fernandez, and H. Juhan, "Migration of ^{137}Cs and ^{90}Sr in undisturbed soil profiles under controlled and close-to-real conditions," J. Environ. Radioact. **50**, 235–252 (2000).
30. M. E. P. Gomes, L. M. O. Martins, L. J. P. F. Neves, and A. J. C. S. Pereira, "Natural radiation and geochemical data for rocks and soils, in the North International Duoro Cliffs (NE Portugal)," J. Geochem. Explor. **130**, 60–64 (2013).
31. J. Koarashi, M. Atarashi-Andoh, T. Matsunaga, and T. Sato, "Factors affecting vertical distribution of Fukushima accident-derived radiocesium in soil under different land-use conditions," J. Sci. Total Environ. **431**, 393–401 (2012).
32. H. Koch-Steindl and G. Prohl, "Considerations on the behavior of long-live radionuclides in the soil," J. Radiat. Environ. Biophys. **40**, 93–104 (2001).
33. E. Mingareeva and M. Lasareva, "Using a collection of soil monoliths for the study of natural radiation of soils in Russia," *Proceedings of the 20th World Congress of Soil Science* (Jeju, 2014), pp. 1–12.

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