Migration of ^{90}Sr and ^{137}Cs in the Soil After Radiation Accidents

N. N. Kazachonok and I. Y. Popova

Abstract Studying the principles of vertical migration and radionuclides distribution in soil is essential for a prediction of their transport dynamics into the root uptake area as well as for the planning of measures for decreasing contamination of agricultural and forest goods. A study of ^{90}Sr and ^{137}Cs distribution in a soil profile, as a result of radioactive contamination due to operation of radiochemical plant, Production Association (PA) "Mayak" was carried out on the territory of northeastern part of Chelyabinsk region in 2008–2012. In forest and meadow soil, the bottom part of forest litter and the upper soil layer (0–5 cm) are the most contaminated with ^{90}Sr and ^{137}Cs. The radionuclides contamination at a depth >20 cm is nearly undistinguishable for different soil types. In alluvial swamp soil, ^{90}Sr is distributed through the whole soil profile, whereas ^{137}Cs is concentrated in the depth of 20–40 cm. Annual plant fallout contributes to a lower extent into contamination of the litter and upper soil layer. In the experiment with a decay of plant fallout, a ^{90}Sr distribution and contamination level in the profile of a model soil was similar to a natural soil, whereas a ^{137}Cs contamination was 10 times lower for the model soil. In the forest-steppe area of Chelyabinsk region, a change of an upward and downward water flow in soil takes place several times in a season, during certain years a fallout and evaporation vary a lot. That is the reason for constant and unpredictable change of vertical radionuclides migration. A computer program for imitation modeling of radionuclides mobility in soil has been developed in our group. The radionuclides behavior in soil is determined by a probability of decay during certain period of time and by a probability of transport up and down within profile for a different distance independent of displacement mechanism. In a certain period of time, a position of a radioactive particle is defined by the sum of opposite directed vectors, each of which in a natural stochastic system is randomly determined, considering probabilities characteristic for given natural system.

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Contents

1 Introduction

The radioactive contamination of the soil is caused in the most cases by the aerosol fallout from the atmosphere. At first are contaminated the soil surface and the aerial parts of plants. The radionuclides migrate gradually and divide in the whole thickness of the soil layer. The study of the regularities of vertical migration and distribution of radionuclides is necessary for forecasting the dynamics of their transfer to the root absorption zone and for planning the measures aimed at the lowering of agricultural and forest products contamination.

The multiyear activity of Production Association "Mayak" has resulted in radioactive contamination of soils in Transurals. The discharges in the air, waste tank explosion in 1957 East-Ural radioactive trace (EURT), carrying away of Karachay lake silt by the wind in 1967 (Karachay Trace) led to the fallout of radioactive aerosols on the soil surface. In the years 1949–1956, the liquid radioactive waste has been disposed in the River Techa and it resulted in the floodplain soil contamination.

Therefore, there are unique conditions in the Transurals for researching the regularities of radionuclides migration in a long-term period by different circumstances of radioactive contamination.

2 Materials and Methods

The researches have been conducted in 2008–2012 in the territory of northeastern part of Chelyabinsk region that suffered from radioactive contamination caused by the activity of radiochemical industrial complex of the Production Association "Mayak." For the analysis of radionuclides distribution dynamics in the soil profile, we used the materials from the database of Urals Research Center for Radiation Medicine.

The specific activity of ^{137}Cs was measured by means of scintillation gamma-ray spectrometer with the software "Progress." From 2011, the spectrometer facility MKC-01A "MULTIRAD" was used for this purpose. In the low-activity samples of ^{137}Cs, the radiochemical iodine-antimony method after concentrating it on the nickel ferrocyanide was used for the measurements. The specific activity of ^{90}Sr in the samples was measured by means of the extractive method based on daughter ^{90}Y using monoisooctyl methylphosphonate. The ß-activity of the selected radionuclides was measured using low-background radiometric facilities УМФ-1500 and УМФ-2000 with flame-photometric control of strontium carrier yield. The method for measuring ^{239}Pu is based on concentrating and cleaning of plutonium isotopes on the anion exchange resin and the following electrochemical deposition on the steel disks. The α-activity was measured using the α-spectrometer facility based on pulsed ionization chamber. The specific activity of plutonium isotopes in the samples was identified and measured on the basis of radiation energy and intensity, using marking (^{236}Pu or ^{242}Pu) with the known activity that was previously brought into the samples. All applied methods and measuring means have State calibration certificates.

3 Results and Discussion

3.1 The Study of Vertical Migration of ^{90}Sr and ^{137}Cs in Automorphic Soils of Terrestrial Ecosystems

In 2008–2011 were taken the samples of soil and ground litter in 130 points of terrestrial ecosystems from the areas of 52 currently existing and 2 evacuated settlements. Virtually in all points where samples were taken the upper soil layer and the forest litter (in the meadow soil–sod cover or steppe mat) were most contaminated by ^{137}Cs and ^{90}Sr. The ratio of the specific activity of radionuclides in the 0- to 10-cm layer to the activity in the 10- to 20-cm layer did not depend on the total contamination density. The value of this ratio is influenced by the type of the ecosystem to the maximum extent: in the forest ecosystems, this ratio for ^{137}Cs is on the average 20.4 ± 4.4, in the meadow ecosystems 2.9 ± 1.6 (although there are exceptions). For ^{90}Sr, the differences are insignificant: 3.4 ± 0.9 in forest, 2.8 ± 1.5 in meadow.

The ratio of the specific activity of ^{137}Cs in the litter to the activity in the 0- to 10-cm soil layer was on the average 0.5 ± 0.1, ^{90}Sr 1.5 ± 0.2. Although the specific activity of ^{137}Cs and ^{90}Sr in the litter is pretty high, it has little effect on the total contamination density, as the litter bulk density is by 2 orders lower than the soil bulk density. Consequently, the importance of the litter in the radionuclide accumulation is significantly lower for the Transurals soils than for forest soils of Polessky radiation reservation that was contaminated as a result of the Chernobyl accident. In the soils of Polessky radiation reservation, the activity of ^{90}Sr and ^{137}Cs in the litter is about 10 times higher than in the upper soil layer. Accordingly, in the litter is deposited 39 % of the total content of ^{90}Sr in the profile up to a depth of 105 cm. The content of ^{137}Cs is 55 % (Mashkov and Malenok 2008).

While analyzing the pattern of ^{90}Sr distribution in the soils of EURT at different times after the accident, it was found that in forest soils in the 50 years that have passed since the accident the contamination maximum practically was not transferred from the 0- to 5-cm layer (Fig. 1a, b). There was a more balanced distribution of ^{90}Sr in the soil profile of the black earth, what is obviously connected with the replowing (Fig. 1c).

The distribution of ^{90}Sr in the profile of a 30-cm layer is well described by the exponential function $y = ae^{-bx}$, where y is the content of radionuclide in the layer (% from the total contamination density a 30-cm layer), x is the ordinal number of a 5-cm layer. The determination coefficients of R^2 approximating curves for gray forest soil, and in the initial period after the accident for soddy-podzolic soil and black earth too, are pretty high (Kostyuchenko et al. 2012a, b). In the remote period after the accident the elective accumulation of ^{90}Sr in particular layers of soddy-podzolic soil increases, this is apparently connected with the inhomogeneity of physicochemical properties. The ^{90}Sr distribution inhomogeneity in the profile of black earth is explained by the mechanical replacement of a part of the radionuclide while processing. Of the greatest interest is the coefficient b using which can be calculated the depth where the contamination density decreases up to the specified multiplicity. ^{90}Sr settled on the soil surface migrates slowly into the deeper layers and the coefficient b decreases. Especially, good this ratio can be seen at the gray forest soil: the coefficient b correlates with the number of years after the accident ($r = -0.949$, $p = 0.02$). ^{90}Sr migration in the soddy-podzolic soil goes more intensive. Such soil is formed at higher moisture; coniferous plant litter contains a lot of organic acids that promote leaching of related elements. The ^{90}Sr distribution pattern in the black earth profile was mostly influenced by the replowing in 1960–1990. The values of b for these soils decrease faster, but the dependence of b on the time is less expressed: for the soddy-podzolic soil $r = -0.68$, $p = 0.20$; for the black earth $r = -0.79$, $p = 0.11$.

The distribution of ^{137}Cs and ^{90}Sr in the soil profile was studied more detailed on the EURT axis (20, 30 and 55 km from the industrial site) and to the south of the Production Association "Mayak" (7, 10 and 20 km from the industrial site). The distribution of ^{90}Sr and ^{137}Cs in the soil profile was examined for 3 types of soil: gray forest soil, soddy-podzolic soil and black earth. The results can be found in Table 1. As Table 1 shows that, although the types of soils, the sources of

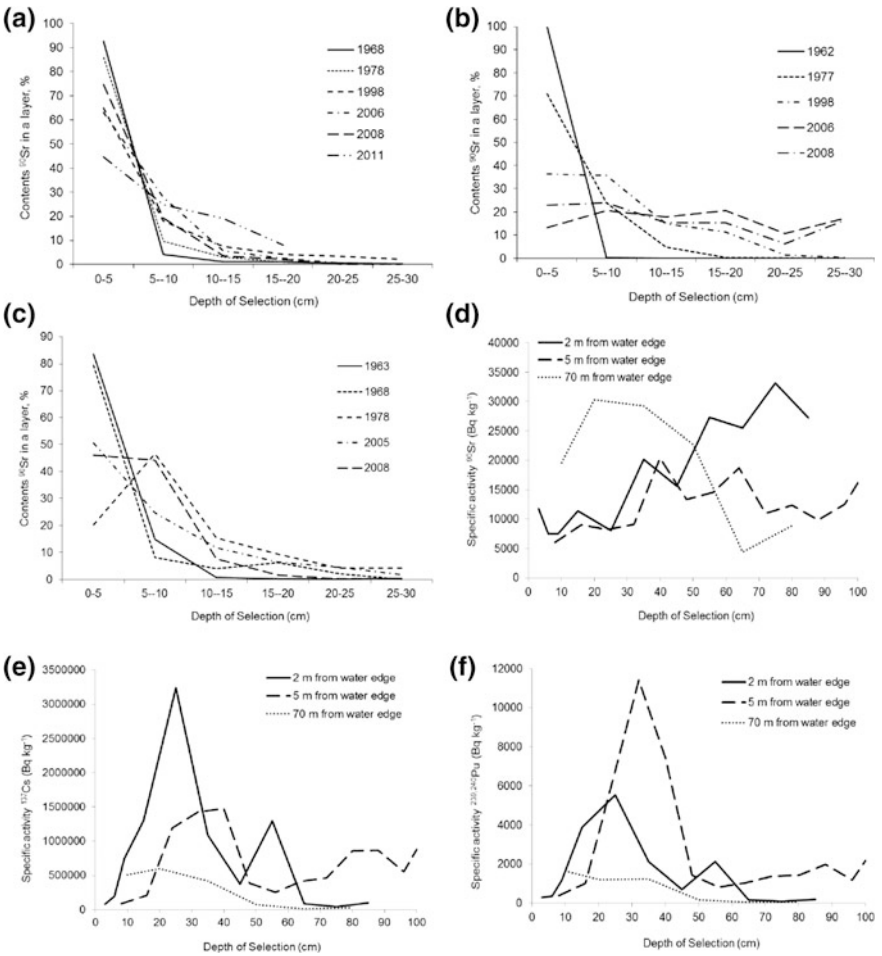

Fig. 1 **a** Dynamics of distribution of ^{90}Sr on a profile of the gray forest soil. **b** Black earth soil. **c** Soddy-podzolic soil. **d** Marsh soil. **e** Distribution of ^{137}Cs on a profile of the marsh soil. **f** Distribution of $^{239,240}Pu$ on a profile of the marsh soil

contamination and the contamination density are various, the specific activity at the depth over 20 cm the specific activity of ^{137}Cs and ^{90}Sr is virtually identical.

The same specific activity of ^{137}Cs and ^{90}Sr—in the range of 9–11 Bq kg^{-1} is described for various subtypes of black earth in Orenburg region in the 20- to 50-cm layers (Efremov 2008). Apparently, the differences in activity of ^{137}Cs and ^{90}Sr that depend on the contamination nature, the contamination density and the type of soil, are observed in automorphic soils of extremely continental climate zone only to a depth of about 20 cm. Then, to a depth of 2–3 m the content of these radionuclides is the same. In the forest soils of Polessky radiation reservation, ^{137}Cs and ^{90}Sr are distributed more equally (Mashkov and Malenok 2008). It should be noted that the

Table 1 Specific activity ^{137}Cs and ^{90}Sr on a soil profile in 2008–2011

Layers in cm	Soddy-podzolic		Gray forest soil						Black earth			
	EURT (55 km)		Southern direction (7 km)		Southern direction (10 km)		EURT (20 km)		EURT (30 km)		Southern direction (20 km)	
	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr
0–5	206	1,910	480	104.3	299	95.8	479	7,809	143	30.8	25.8	33.1
5–10	12.1	1,771	27.3	48.9	192	85.5	23.2	2,029	101.5	32.2	20.1	22.0
10–15	15.2	295	16.4	23.2	26.7	33.5	8.1	363	87	20.8	19.9	17.9
15–20	10.3	57.9	15.2	33.8	5.9	11.6	13.3	223	41	20.7	13.7	20.5
20–25	8.9	10.5	11.3	17.7	12.2	6.6	6.5	12.3	13.2	8.4	12.2	3.5
25–30	6.7	6.9	9.5	2.0	3.3	6.1	6.1	16.7	10.6	22.1	8.2	8.4
30–35	17.6	17.9	3.9	1.6	8.8	2.8	4.4	14.7	12.3	5.1	5.2	14.1
35–40			5.5	0.95	5.8	5.0	9.2	12.1			4.5	2.9
40–45	12.5	14.8	10.9	7.4	6.3	7.8	22.6	15.6	15	8.7	5.8	1.4
45–50			11.3	8.9	7.3	13.4	9.7	14.0			8.2	4.6
50–55	2.4	6.6	11.6	6.1	4.1	4.6			8.7	5.0	3.9	3.9
55–60			–	–	–	–	12.7	34.6			4.5	7.1
60–65	2.4	4.1	–	–	–	–			6.6	5.3	4.1	2.0
65–70			–	–	–	–	4.2	11.1			3.9	6.6
70–75	2.2	4.3	–	–	–	–			9.9	5.0	2.7	2.9
75–80			–	–	–	–	3.5	4.7			8.2	5.3
80–85			–	–	–	–			8.8	9.7	4.7	3.1
85–90	8.6	7.9	–	–	–	–					5.6	2.7
90–95			–	–	–	–	5.5	10.6	41.3	23.2	4.6	0
95–100			–	–	–	–					5.9	10.2
100–105	6.2	8.2	–	–	–	–			10.7	7.4	4.5	3.4
105–110			–	–	–	–	9.0	6.8			7.4	6.1
110–115			–	–	–	–			–	–	3.8	0
115–120	7.6	7.3	–	–	–	–			–	–	7.6	8.6
120–125			–	–	–	–			–	–	–	–
125–150	–	–	–	–	–	–	7.8	2.8	–	–	–	–
150–175	–	–	–	–	–	–	7.4	2.9	–	–	6.5	19.9
180–185	–	–	8.8	4	–	–	–	–	–	–	–	–
295–300	–	–	10	8.5	–	–	–	–	–	–	–	–

area of Polesye has been recognized as a biogeochemical province with high mobility of ^{137}Cs long before the Chernobyl accident (Alexachin and Tichomirov 1976).

Significant influences on the radionuclides distribution pattern in the soil profile have agro technical measures. In 2007, in the vegetable rotation fields of «Sovkhoz Beregovoy» LLC in 47 % cases on the irrigated lands and in 19 % cases on the bogharic lands the 20- to 40-cm subsurface layer contained more radionuclides than the 0- to 20-cm top soil layer (Table 2).

Table 2 The relation of density of pollution of arable and sub-arable layers (0–20 cm/20–40 cm) on fields of a vegetable crop rotation

Cultivation conditions	^{90}Sr	^{137}Cs
Watering	1.4 ± 0.6	1.3 ± 0.9
Not watering	1.8 ± 0.3	2.2 ± 0.9

3.2 The Study of Vertical Migration of ^{90}Sr and ^{137}Cs in Hydromorphic Soils of Floodplain

The floodplain of Techa River is contaminated with radionuclides due to the discharges of liquid radioactive waste in 1950–1956. The soil in the river headwaters is polluted to the maximum extent. Figure 1d–f shows the radionuclides distribution in the profile of the bog soil from the selection place in the area of Asanovsky bogs, 7.5 km away from the dam of B-11 technical water reservoir. Figure 2a–c shows the radionuclides distribution in the profile of the meadow Soddy soil, 24 km away from the dam of B-11 water reservoir. It can be seen that in the bog soil and in the bank Soddy soil that are constantly wet, ^{90}Sr migrates to a greater depth and is accumulated in different layers. ^{137}Cs and $^{239,240}Pu$ migrated in the bog soil up to the depth of 100 cm, but the largest number of them is contained in the 20- to 40-cm layer. In the Soddy soil, ^{137}Cs and $^{239,240}Pu$ remained mainly in the surface layer. These differences have to be taken into account while assessing the territory contamination density and calculating the total supply of radionuclides.

While comparing the specific activities of radionuclides in the samples, it was found that the horizontal and vertical distribution of ^{137}Cs and $^{239,240}Pu$ in the soil is similar. For 352 soil samples taken at different sites along the bed, at different distances from the river and at different depths, the correlation coefficient between the activities of these radionuclides equaled to 0.869 (Fig. 2d). The dependence of activities of ^{137}Cs and $^{239,240}Pu$ in the bottom silt was lower, the correlation coefficient—0.688 for 176 samples (Fig. 2e). These relations enable us to assess approximately the $^{239,240}Pu$ contamination level based on the results of γ-spectrometric measurement of ^{137}Cs. The equation for the soils: $A_{Pu} = 0.044\ (A_{Cs}^{0.73})$; A_{Pu} and A_{Cs}—specific activities of $^{239,240}Pu$ and ^{137}Cs ($R^2 = 0.816$); for the bottom silt—$A_{Pu} = 0.033\ (A_{Cs}^{0,78})$, ($R^2 = 0.847$).

The correlation coefficients between the activities of ^{137}Cs and ^{90}Sr, ^{90}Sr and $^{239,240}Pu$ in all taken samples are also statistically significant, but low (from 0.30 to 0.52). This is explained by the fact that the behavior of ^{90}Sr in the soils differs from ^{137}Cs and $^{239,240}Pu$. During the time that has passed since the massive discharges of radionuclides the correlation between the radionuclides has changed.

Therefore, only the samples taken at the earliest in 2007 were chosen for the calculation of the equation for calculating the activity of ^{90}Sr using the dates on the activity of ^{137}Cs. Figure 2f shows the relation of specific activities of ^{90}Sr and

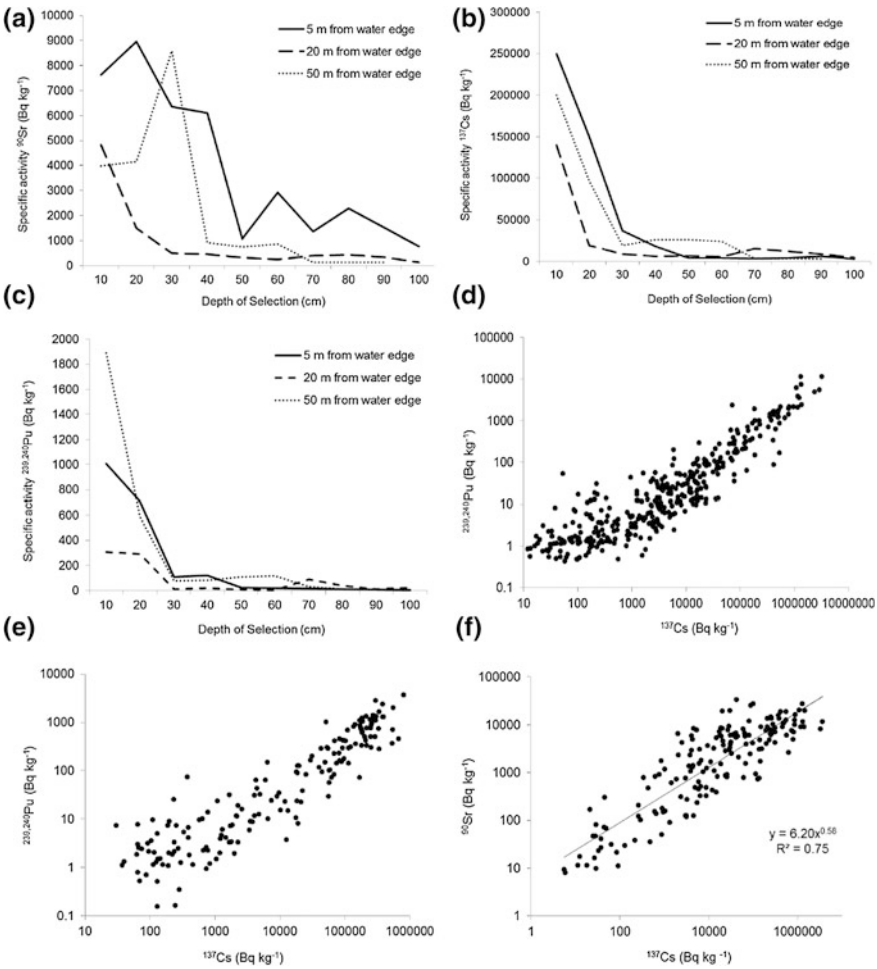

Fig. 2 **a** Distribution of ^{90}Sr on a profile of the cespitose meadow soil. **b** Distribution of ^{137}Cs on a profile of the cespitose meadow soil. **c** Distribution of $^{239,240}Pu$ on a profile of the cespitose meadow soil. **d** Correlation of specific activity of ^{137}Cs and $^{239,240}Pu$ in samples of inundated soils. **e** Correlation of specific activity of ^{137}Cs and $^{239,240}Pu$ in samples of ground deposits. **f** Correlation of specific activity of ^{137}Cs and ^{90}Sr for all samples

^{137}Cs in soil samples taken from different depths. Figure 3a shows information about the samples taken from the upper layer. The approximating curves equations and determination coefficients can be seen on the same figures.

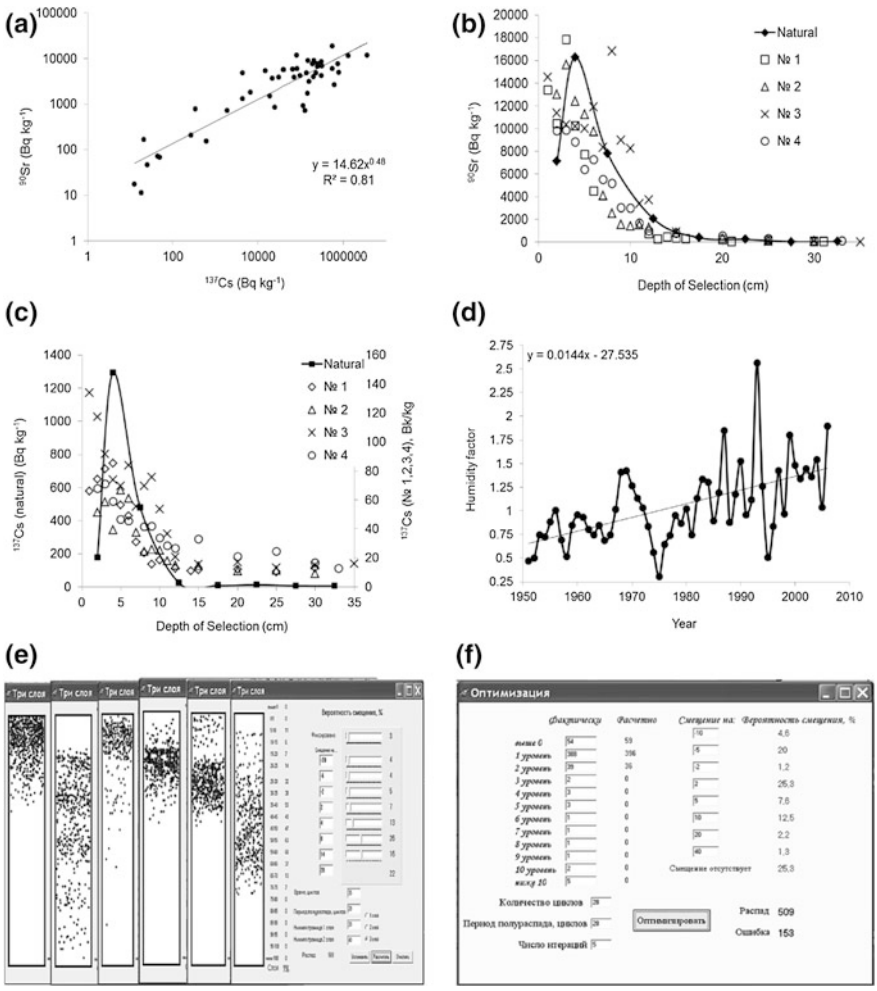

Fig. 3 **a** Correlation of specific activity of ^{137}Cs and ^{90}Sr in soil samples from a layer of 0–10 cm. **b** Specific activity of ^{90}Sr in the soil after decomposition of the remains of plants. **c** Specific activity of ^{137}Cs in the soil after decomposition of the remains of plants. **d** Dynamics of humidity factor. **e** Examples of the distributions received at modeling of a local circulation of radionuclides. **f** Result of optimization of values of probabilities of shift of radionuclide

3.3 Regularities of Local Circulation of Radionuclides

The most researchers, who studied the distribution of radionuclides in the soil, note that in the first years after atmospheric fallouts the greatest specific activity of ^{90}Sr and ^{137}Cs is observed in the sod cover or in the forest litter. However, it is known that the plant litter formed by the grass decomposes during one season and the leaf forest litter–during 3–4 years. A little longer decomposes the coniferous forest

litter, but in 50 years that have passed after the formation of EURT the forest litter that was contaminated by atmospheric fallouts had to decompose a long time ago and the steppe mat of the black earth began to form at the earliest in 1991. Thus, the high specific activity of the litter observed at the moment in the territory of EURT is determined not by primary fallouts of 1957 and even of 1967, but ether by a high speed of local circulation of radionuclides, or by the ongoing atmospheric fallouts of production waste from the Production Association "Mayak."

The supply of radionuclides in the litter and in the upper soil layer is constantly refilled with the plant litter. This litter is in its turn contaminated by atmospheric fallouts and due to the transfer of the radionuclides to the top by the root system. By virtue of the fact that the over ground biomass of the grass transforms to the litter at the end of the year almost completely, the annual transfer of ^{90}Sr to the surface is determined by annual productivity of the ecosystem.

The litter formed by the woody plant formation makes only one part of annual gain, apart from the leaves and the grass the litter consists of slowly decomposing wood, needles and cones. That is why the radionuclides are deposited not only in the soil, but also in the wood and in the litter. When the dead biomass is mineralized, the relative content of radionuclides in the litter increases.

In the territory of EURT, the ratio of the specific activity of ^{90}Sr in the grass to the activity in the 0- to 10-cm soil layer 20 km away from the Production Association "Mayak" was 0.7 ± 0.3, 30 km away 2.1 ± 1.0. For ^{137}Cs is this ratio accordingly 0.08 ± 0.04 and 0.4 ± 0.4. Since the grass is relatively more contaminated on a greater distance, we can assume that now prevail the entry of ^{90}Sr and ^{137}Cs to the biomass through the roots.

In 2008, on the axis of EURT 20 km away from the Production Association "Mayak" the plant litter from the current year was collected in the birch forest. The specific activity of ^{90}Sr in the litter amounted to 5,904 Bq kg^{-1} and ^{137}Cs 54 Bq kg^{-1}. With the litter of over ground phytomass, 2.3 kBq m^{-2} ^{90}Sr and 0.28 kBq m^{-2} ^{137}Cs were transferred to the surface of the soil (litter). This amounted to 2.1 % of ^{90}Sr and 3.4 % of ^{137}Cs of the contamination density of the whole forest litter layer and to 0.36 % of ^{90}Sr and 0.8 % of ^{137}Cs of the total contamination density of the litter and the 0- to 20-cm forest layer. Consequently, the plant litter of the aboveground organs provides a small proportion of contamination density of the litter and the upper soil layer.

Table 3 shows the values of the specific activity of radionuclides in the litter and the upper soil layer. Activity of ^{90}Sr in the upper part of the litter coincides virtually with the activity in the upper soil layer; the activity in the lower part is considerably higher. The activity of ^{137}Cs in the upper and lower layers of the litter differs to an even greater degree, than the activity of ^{90}Sr.

The specific activity of ^{90}Sr in the litter was higher than in the grass at a distance of 20 km from PA"Mayak" 2.2 ± 0.7-fold, 30 km 6.4 ± 3.6-fold; of ^{137}Cs- 14 ± 8-fold and 12 ± 8-fold accordingly. Question arises by which mechanisms is maintained a high specific activity in the lower layer of the soil litter.

Table 3 Activity of radionuclides in the soil and a lying in EURT territory (kBq kg^{-1})

Distance from Mayak PA (km)	Soil	Top part of a laying		Lower part of a laying		Layer of earth (0–5 cm)	
		^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
20	Gray forest soil	7.1	0.18	16.3	1.3	7.8	0.48
	Black earth	0.6	0.02	1.2	0.2	1.1	0.3
30	Black earth	0.03	0.005	0.09	0.02	0.03	0.14
55	Soddy-podzolic	2.8	0.06	10.0	1.2	1.9	0.2

In 2010–2011, the influence of water regime and burrowing detritophagous (*Lumbricus* sp.) on the migration of ^{137}Cs and ^{90}Sr to the soil from the contaminated plant litter was studied under laboratory conditions. The litter was collected in October 2010 in the birch forest in the area of the evacuated village Alabuga. The initial activity of ^{137}Cs in the plant litter was 180 Bq kg^{-1}, of ^{90}Sr 6,100 Bq kg^{-1}. The soil was taken from the horizon A_1 of gray forest soil that was radioactively polluted only as a result of global fallouts. The initial activity of ^{137}Cs in the soil 11.0 ± 7.0 Bq kg^{-1}, the initial activity of ^{90}Sr in the soil 7.6 ± 7.1 Bq kg^{-1}. A 35-cm soil layer was placed into the plastic cylinders with a diameter of 10 cm and with a length of 45 cm. The 5-cm high plant litter layer was placed on the soil surface. Once a week during the whole experiment, we added fresh litter in order that the litter layer equaled to 5 cm.

The experiment was carried out during 12 months in 4 variations:

No. 1.
Sprinkling of litter surface with 10 ml distilled water 3 times a week.
No. 2.
Before the litter placement 10 grown-up earthworms *Lumbricus* sp. were placed on the soil surface. Sprinkling of litter surface with 10 ml distilled water 3 times a week.
No. 3.
Sprinkling of litter surface with 50 ml distilled water 3 times a week.
No. 4.
Before the litter placement 10 grown-up earthworms *Lumbricus* sp. were placed on the soil surface. Sprinkling of litter surface with 50 ml distilled water 3 times a week.

We stopped to add the litter after 12 months. The experiment was terminated after the complete disappearance of the litter from the soil surface, the soil was divided into 1-cm-thick layers, dried and the content of ^{90}Sr and ^{137}Cs was measured in these layers. It was found after ending the experiment that there is the rest of semi decomposed litter (parts of branches) in the upper soil layers. Apparently, the detritophagous kept in the litter and soil contributed to the mixing of the branch rests with the soil.

The thickness of mixed soil layer was:

Variation No. 1–6 cm
Variation No. 2–7 cm
Variation No. 3–10 cm
Variation No. 4–10 cm

In the variations No. 3 and No. 4 have formed a washing water regime. The washing water was collected and analyzed. It was found that during the whole experiment in the variation No. 3 2.03 Bq of ^{90}Sr and 0.14 Bq of ^{137}Cs were washed away with the washing water. It were washed away in the variation No. 4 5.88 Bq of ^{90}Sr and 0.35 Bq of ^{137}Cs. Consequently, the transfer of radionuclides with the soil water was insignificant.

Figure 3b and c shows the values of specific activity of ^{90}Sr and ^{137}Cs that were transferred into the humus horizon of gray forest soil during the decomposition of plant litter collected in the birch forest on the axis of EURT. For comparison, the values of the specific activity in the natural gray forest soil in the place where the plant litter was collected are written on the same figure.

The pattern of ^{90}Sr distribution from the contaminated plant litter to the humus horizon of the gray forest soil during the 12-month-long experiment corresponds generally with the contemporary ^{90}Sr distribution in the gray forest soil of EURT. The highest specific activity was noted in the rest of the litter and in the upper soil layer. During the experiment in different moisture regimes, the average specific activity of ^{90}Sr in the 0–5-cm layer was from 0.8×10^5 to 1.2×10^5 Bq kg^{-1}. In the natural soil where the litter was collected the activity of ^{90}Sr in the lower litter part was 1.6×10^5 Bq kg^{-1}, in the 0–5-cm layer 0.8×10^5 Bq kg^{-1}.

The distribution of ^{137}Cs in experiment differed significantly from the distribution in the natural soil of EURT. In the upper litter layer of the natural soil, the activity of ^{137}Cs was 178 Bq kg^{-1}, in the litter collected for the experiment the activity of ^{137}Cs was 180 Bq kg^{-1}, in the lower litter layer of the natural soil the activity of ^{137}Cs was 1,293 Bq kg^{-1}, in the 0- to 5-cm layer of the natural soil 480 Bq kg^{-1}. And in experiment in the upper soil layers the activity of ^{137}Cs was not higher than 100 Bq kg^{-1} in most cases. But in the natural soil in the 10- to 15-cm layers and lower the activity of ^{137}Cs was 2- to 3-fold lower than in similar soil layers in experiment.

It was found while comparing the results of different variations of experiment that the moisture regime had the greatest influence on the migration of ^{90}Sr and ^{137}Cs in the soil. Many authors have noted that the most important factor in determining the mobility of radionuclides is soil moisture (Perevolotsky 2006). It should be expected that in natural conditions in the washing water regime the radionuclides migration downward the profile will increase and in the not washing regime, it will decline. In the forest-steppe zone of Chelyabinsk region, the most of precipitation falls in July in the form of showers. In May and June, the amount of precipitation is small. Thus, several times per season occurs the change of upward and downward water flows in the soil. In addition, in particular years, the amount of precipitation and evaporation capacity can vary greatly. Figure 3d shows the values of humidity factor (HF) calculated by us on the grounds of the dates from

Ozersk meteorological station (Mayak 2007). It is to see that in the initial years after the start of PA "Mayak" the water regime of automorphic soils was mostly not washing (HF < 1) and the fluctuations of HF were relatively smooth. Since 1982 there are sharp changes of HF values, in most cases HF > 1 (it corresponds to the washing water regime for automorphic soils). Therefore, the vector of radionuclides vertical migration is constantly and unpredictably changing. This uncertainty creates problems for the mathematical modeling of the vertical migration of radionuclides.

3.4 Modeling of Local Circulation of Radionuclides

Development of models was conducted till present in the following areas.

3.4.1 Analytical Models

Analytical or logical models of radionuclides migration in the soil represent functions in which the variables are the parameters of the processes and factors that influence, in the opinion of the authors of the models, the value characterizing the result of migration (speed of activity front or activity maximum replacement, a.s.o.). Initially, we considered 2 factors influencing the vertical migration: diffusive transfer, convective transfer and their combined action. The indices of the variables were then calculated empirically. Baturin (1997) has obtained a semiempirical equation for the soils of EURT using the parameters of convective transfer speed and diffusive penetration speed that he has found by exhaustion. Based on this equation, he calculated that the maximum concentration of radionuclides moves at a speed of 0.3–0.4 cm per year. At the same time in 25 years, the rate of diffusion of ^{137}Cs decreases by 0.01 cm $year^{-1}$ and of ^{90}Sr increases by 0.02 cm $year^{-1}$ (Baturin 1997). A fundamental study of the factors influencing these factors has performed Prochorov (1981) showed that according to the influence on the diffusion, the soil properties are arranged in a row: humidity > amount of exchange calcium and magnesium = exchange capacity $\Longrightarrow$ humus pH > temperature. He also evaluated the role of adsorption in the migration of ^{90}Sr (Prochorov 1981).

These three models are used nowadays too. But apparently, they are suitable for a homogeneous unstructured and constantly moist soil which has been exposed to a onefold surface contamination. For most types of soil, an effective diffusion is only possible within the structural aggregate at a time when the soil is sufficiently moistened. While analyzing the convectional transfer, it is necessary to distinguish the transfer of radionuclides with gravity water and the movement in capillary pores, which can be multidirectional.

Istomin et al. (2005) have developed a model of one-dimensional vertical migration of radionuclides in a multiphase system of topsoil. The model takes into account many parameters, including soil pore tortuosity, energy release capacity

during evaporation/condensation and crystallization/melting of water, as well as radioactive decay (Istomin et al. 2005). It is clear that the measurement of all these parameters in a specific soil-plant system is a very labor-intensive process. In addition, it is first necessary to define empirically in a multifactorial experiment and in natural conditions the values of the indices of equations describing the dependences of input and output parameters. Considering that an increase in the number of variables leads to an avalanche accumulation of error such models may be mainly of a theoretical interest.

3.4.2 Empirical Models

In empirical models, the available data on the specific activity or the content of the radionuclides in particular soil layers are used for calculating the spatiotemporal characteristics of migration process (speed of maximum replacement, half-residence time in the layer a.s.o.) or the predicted values of the activity or the content of radionuclides in the respective layers (Trapeznikov et al. 2007).

Several researchers calculate simply the regression equations for the actual values of radionuclide activity in soil layers. Most researchers use for this purpose an exponential function. So, Efremov (2008) has calculated the parameters of the regression equation of exponential type for ^{137}Cs and ^{90}Sr in the profile of several soil types and subtypes of the Orenburg region (Efremov 2008). Arastovich (2004) has calculated the formulas of exponential dependence of the concentration of ^{137}Cs and ^{90}Sr on the time in the soil layer of sod-podzolic swampy uncultivated soil in the Gomel and Brest regions (Arastovich 2004).

We have calculated the regression equations for the distribution of ^{90}Sr in the profile of three types of soil that are common in the territory of East-Ural Radioactive Trace and the 30-km zone of PA "Mayak" based on 5 selections of each type made at different times after the accident (between 1962 and 2008). The distribution of ^{90}Sr in the profile of a 30-cm layer is well described by an exponential function: $y = ae^{-bx}$, where y—the radionuclide content in the layer as % of total contamination density of the 30-cm layer, x—the number of the 5-cm layer. ^{90}Sr fallen on the surface of the soil migrated slowly into the deeper layers and the coefficient b decreased gradually during the years passed since the accident.

For gray forest soil: the value of the coefficient b correlates with the best way with the number of years that have passed since the accident ($r = -0.94$, $p = 0.02$). b values for other soils eventually decrease over time faster, but the dependence of b on the time is less expressed: for the sod-podzolic soil $r = 0.68$, $p = 0.20$; for the black earth $r = 0.79$, $p = 0.11$. In the remote period after the accident, the selective accumulation of ^{90}Sr in particular layers of sod-podzolic soil increases, and it is apparently connected with the heterogeneity of the physicochemical properties. Heterogeneous distribution of ^{90}Sr in the profile of black earth, its accumulation above the plow pan level is explained by the mechanical transfer of a part of radionuclide during the processing (Kostyuchenko et al. 2012a, b).

3.4.3 Structural Dynamical Models

The models are based on the presentation of the ecosystem in a form of flows and levels. The information about levels is contained in databases; the flows are calculated on the basis of analytical model equations of transfer functions. In the model are used the parameters of equations that are derived empirically and are also contained in the database. Detailed models based on structural dynamical principles are developed by Mamikhin (2003). Such a model can very detailed describe the condition and behavior of an ecosystem, but requires a huge amount of actual data during the development stage. In case of lack of actual data, it is suggested to use numerical experiments with different options of functions and to assess the unknown parameters through the method of successive approximations (Mamikhin 2003).

The principles of structural dynamics are similar to the principle of neural networks used for modeling the vertical and horizontal migration of contaminants. For such models is also needed a large amount of initial information that is systematized in the form of the databases. In the models developed under the leadership of Kundas et al. (2011) are used multilayer networks. The entry layer reflects the conditions (the initial substance concentration, time, type of soil, etc.), the auxiliary layers and the auxiliary networks are used to specialize the conditions (physicochemical properties of a particular type of soil, etc.). For signal processing are generated intermediate and hidden layers. An important advantage of neural networks is their ability to self-learning. In conventional structural dynamical models various errors are accumulated when the number of structural elements increases and in the neural networks due to the "back-drive" and self-learning of the system the errors are minimized (Kundas et al. 2011).

3.4.4 Use of Virtual Machines

In our opinion, a promising direction in the modeling of the processes of contaminant transfer in a heterogeneous environment can be virtual machines that simulate the behavior of contaminant particles (ions, molecules, colloidal particles) in a heterogeneous system. Such models can be used for setting up virtual experiments for predicting the behavior of particles under the known parameters of transfer probability and for the assessment of these parameters under the known transfer result.

For example, Zhikharevich and Shumilyak (2012) have showed the congruence of temperature distribution in the sample between the cellular automaton model and the exact analytical solution of the equation of the heat transfer at a point in time (Zhikharevich and Shumilyak 2012) and also examined cellular automaton models of continuous and discrete diffusion and pointed out that the diffusion coefficient should be in the form of a probability function taking values from 0 to 1 with some degree of probability (Zhikharevich and Ostapov 2009).

A radioactive particle that is fallen on the soil surface is displaced after a certain time interval along the soil profile for the vector that is the resultant of all forces acting on the particle during this time. Whereas the soil is heterogeneous, the fate of each individual particle is determined by a number of random factors, so the particles do not move in a united front, but "spread" in the profile. Ions, colloidal micelles, isotopes connected in the crystalline grid can migrate at different speeds and in different directions. In soils without significant incline none of horizontal directions will prevail, the possibility of the horizontal displacement of a particle is equally compensated by the displacement of another particle in the opposite direction. Therefore, only vertical transfer, i.e., the projection of the resultant of all the forces acting on the vertical, should be taken into account while modeling.

The behavior of radionuclides in the soil is determined by the decay probability in a given period of time, and the probability of their displacement at various distances up or down along the profile during this period, regardless of the mechanism of displacement. After a certain number of timeslots the position of a decayed particle is determined by the result of the addition of oppositely directed vectors, each of which is defined randomly in the natural stochastic system in accordance with the probabilities that are typical for this natural system.

In our opinion, it is reasonable to use simulation modeling with cellular automata method for solving some tasks related to the study of dynamics regularities of vertical migration and distribution of radionuclides in the soil profile.

The space is discretized in the form of an array, the time—in the form of a rotation of the cycle. For the array, elements (in this case—for ions or colloidal particles) are given the rules of status change. The basic algorithm of the module on each rotation of the cycle simulates the random selection of the particle displacement vectors in accordance with the given probabilities.

Currently, we have developed, tested and tweaked in Delphi the principal algorithms of the model of radionuclide particles migration in the soil and created functional modules of this model:

1. Module that simulates the migration of particles in a homogeneous soil, with taking the decay into account. The researcher can change the displacement probabilities for different vectors and perform a complex of virtual experiments in order to study the behavior of particles, find the leading vectors and compare them with the natural physicochemical processes.
2. Module that simulates the migration of particles in a 3-layer-soil, with taking the decay into account. While working with this module, the researcher can specify arbitrary or actual values of capacity of 3 soil layers and specify a different set of displacement probabilities for each layer.
3. Module for optimizing the particles migration parameters according to the actual distribution. Using this module, the researcher specifies the actual distribution of the particles in the soil profile. The program specifies the probability values using the Monte Carlo method and gives preference to the options where $\Sigma(x_{i(\mathrm{act})} - x_{i(\mathrm{mod})})^2$ is the smallest. During a number of iterations, the program selects a set of

probabilities in order that the distribution resulting from the virtual experiment corresponds in the best way to the specified natural distribution.

The model studies have showed that, depending on the selected displacement probability, the pattern of particles distribution in the soil profile after a specified period of time can vary significantly. Specifying a different set of probabilities for different soil layers, we can obtain a «layered» distribution that is typical for alluvial floodplain soil or a depth ward monotonic decrease in the content of radionuclides that is typical for arid automorphic soils. It is reasonable to use simulation modeling with cellular automata method for solving some tasks related to the study of local circulation of radionuclides, dynamics regularities of vertical migration and distribution of radionuclides in the soil profile. The examples of distribution are shown in the Fig. 3e. Using this module for optimizing the parameters of particles migration, the researcher can specify the actual distribution of the radionuclides in the soil profile? During a number of iterations the program selects a set of probabilities in order that the distribution resulting from the virtual experiment corresponds in the best way to the specified natural distribution. Figure 3f shows the results of modeling the actual distribution of ^{90}Sr in the gray forest soil using the module for optimizing. In our opinion, the module for optimizing allows us to select the values of probability of displacement for specified vectors for homogeneous soil or a particular homogeneous layer with a high accuracy.

4 Conclusion

1. In natural forest and meadow soils of Transurals, the lower litter part and the upper soil layer (0–5 cm) are most contaminated with ^{90}Sr and ^{137}Cs. The contamination of different soils with radionuclides at the level deeper than 20 cm is slightly differed.
2. In floodplain bog soils, ^{90}Sr is distributed in the whole profile, and ^{137}Cs and $^{239,240}Pu$ are concentrated at a depth of 20–40 cm.
3. In floodplain meadow Soddy soils, ^{90}Sr is concentrated on a depth of 40 cm, and ^{137}Cs and $^{239,240}Pu$ remain mainly in the upper 0- to 10-cm layer.
4. In experiment with the decomposition of plant litter the pollution levels and the distribution of ^{90}Sr in the profile of laboratory soil were similar to the distribution in the natural soil, and the levels of contamination of laboratory soil with ^{37}Cs were approximately tenfold lower.
5. We have developed a computer program to simulate the transfer of radionuclides in the soil.

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