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Environmental Migration of Radionuclides (^{90}Sr, ^{137}Cs, ^{239}Pu) in Accidentally Contaminated Areas of the Southern Urals

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1. Introduction

In the late 1940s, the facility Mayak Production Association (Mayak PA) for weapon grade plutonium production was put into operation in the vicinity of the town of Kyshtym. The technology used in plutonium production involved generation of high-level waste. A number of accidents that occurred at the plant were associated with inadequate radioactive waste storage techniques. In 1949-1956, radioactive waste with total activity of about 1.8 10^{17} Bq (4.9 MCi) was discharged into the Techa River which resulted in contamination of all river system components. Currently, at late time after the beginning of contamination, ^{90}Sr and ^{137}Cs still remain essential dose-forming radionuclides on the Techa River. In 1957, the East-Urals Radioactive Trace (EURT), and in 1967 the Karachai Radioactive Trace (KRT), were formed. A distinguishing feature of the radionuclide composition of the releases on the EURT at late time is the prevalence of ^{90}Sr and a minimum content of ^{137}Cs. The composition of radioactive dust on the 1967-Trace is represented primarily by Cs and Sr isotopes in less accessible biological forms compared to those observed on the EURT (Fig. 1). In contaminated areas, measurements of soil contamination levels, analysis of the patterns of radionuclide migration, changes in their biological accessibility, transfer of radionuclides from soil to vegetation, milk and vegetable produce have been conducted on regular basis. Specific activity of ^{90}Sr measured in cross sections of the river at most of the riverside villages has decreased to permissible values since the start of observations in 1960. In flooded areas of the bank line, the processes of deepening of radionuclides into soil and a more uniform distribution of radionuclide contents over the soil layer at a depth of 1.5 m were observed. Mean content of ^{90}Sr in milk produced in the riverside villages has declined to permissible values. On EURT and KRT, of the total radionuclides contained in the soils, 80% remain deposited in the upper 20-cm layer. Biologically accessible and insoluble forms of ^{90}Sr and insoluble forms of ^{137}Cs are prevalent. Reduction in radionuclide content in milk has taken place over the first 1-2 years due to deepening of radionuclides into soil and a decrease in their biological accessibility. The main factor that caused cleansing of radionuclides from food chains was radioactive decay and reduced biological accessibility of radionuclides in soils.

1.1 Natural-climatic characteristics of the affected territory

As a result of the accidents at the Mayak AP, a number of rivers, water basins and lands of the southern and middle zone of the Trans-Urals region were contaminated. The EURT occupies over 3/4 of the forest and forest-steppe part of the Trans-Urals region where there are numerous lakes, swamps, all kinds of depressions and pits, wood lands and forest outliers which account for non-uniformity of radioactive fallouts. The most common are chernozemic-meadowy and meadowy-chrnozemic soils. The Trans-Urals region has a typical continental climate which is formed by the air masses coming from the Atlantic Ocean. The wind conditions of the region are characterized by prevalence of westerly winds. Whirlwinds are not an infrequent phenomenon. Species of wood prevailing in the forest zone include pines and the main hardwood species – birches and aspens. The floodplain vegetation includes grassy and woody-shrubby species. Birches and willows are encountered in the floodplain. Miscellaneous herbs are characteristic of the Techa floodplain in the middle and lower reaches of the river.

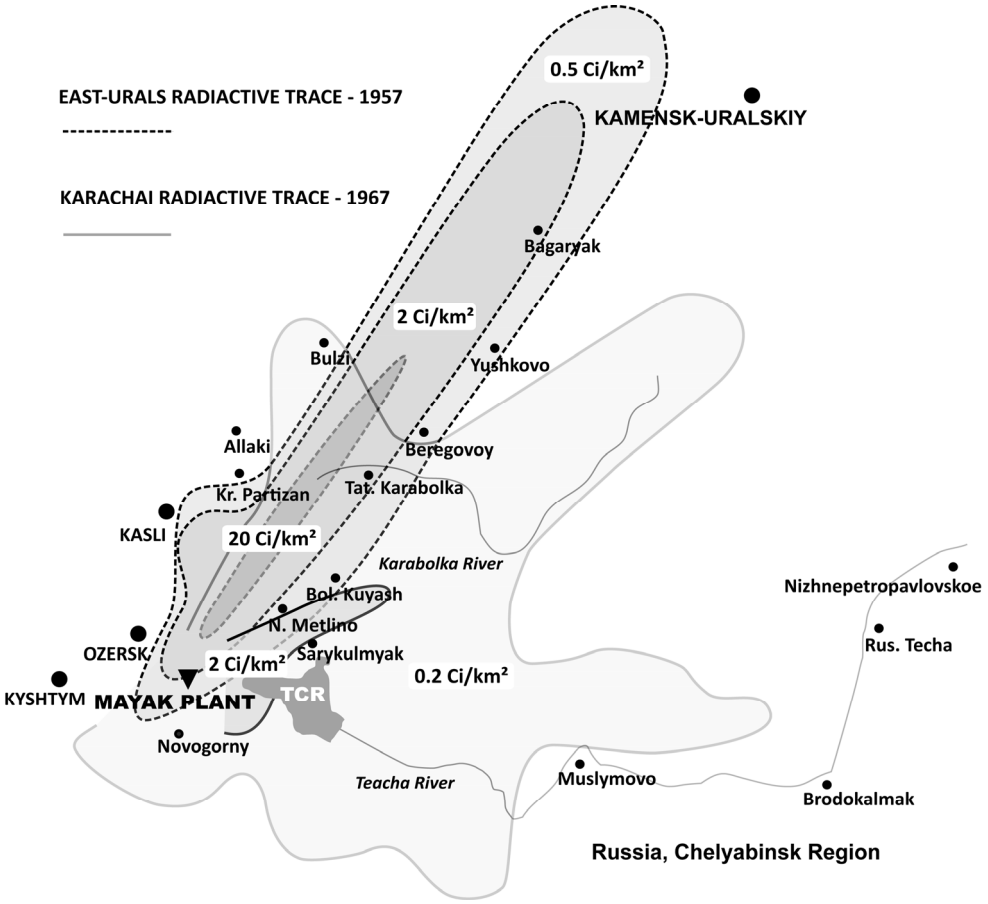

Fig. 1. Schematic map of radiation accidents in the Southern Urals

1.2 Methods of the study

The key contaminants of the environment at late phase of the accident are represented by a small number of radionuclides: ^{90}Sr, ^{137}Cs, ^{239}Pu and a few other. Samples of water, bottom sediments, aquatic vegetation, soil and grass were collected at 8 control cross sections on the Techa River in accordance with conventional standard methods of sample drawing. (Basic Requirements…, 1999, in Russian [Основные требования…, 1999]). Samples of bottom sediments were collected at a depth of 60 cm using metallic tubes, the volume of the samples ranged from 20 to 100 cm^3. To allow assessment of the distribution and deposits of radionuclides in floodplain soil, samples were drawn at a depth of up to 150 cm. Samples of grass were taken from the area of 0.25 m^2. Water was sampled at a depth of 10-50 cm using a sampling device. Measurements of radioactive contamination in the riverside villages and the adjacent areas, contents of radionuclides in food products, migration of activity from soil to food products, horizontal and vertical migration of radionuclides in soils were conducted In the EURT zone. Processing and preparation of food products and other samples to be used in a corresponding assay were conducted using the generally accepted methodology. Measurements of ^{137}Cs and other gamma-emitting radionuclides were performed using the gamma-spectrometric method. ^{90}Sr concentration in samples was measured by the radiochemical separation of daughter ^{90}Y using monoisoochthyl-methyl ether of phosphonic acid (1966) and a subsequent measurement of its activity in a small-background β-metric installation analogous to UMF-2000 based on a flame photometric control of strontium carrier yield. ^{90}Sr measurement error accounts for 20% at activities of <0.7 Bq/g, and for 10% at higher activity levels. The range of measured values was $0.02\text{-}1 \cdot 10^5$ Bq/dm^3.

The method for measuring ^{239}Pu involves increasing concentration of plutonium ions, and cleaning of isotopes using anion-exchange tar followed up by electro-chemical precipitation on steel disks. Measurements of α-activity were performed in an α-spectrometric installation. Identification and measurements of specific activity of plutonium isotopes was conducted using an indicator mark (^{236}Pu or ^{242}Pu) with a known activity ranging from 1.5 to $1 \cdot 10^5$ Bq/kg, dm^3 preliminarily introduced into the sample.

2. Techa River

2.1 Hydrographic characteristics of the Techa River area

The Techa River basin catchment area is situated between the mountainous Urals region and the Tobol River valley. The water from the Kasli-Irtyash lake system is flowing through the Techa River. After the construction of the Mayak plant was started, a cascade of industrial water reservoirs (IWB) was built for storing low-level liquid waste. In 1956 reservoir B-10, and in 1963-1964 reservoir B-11 were built. Since 1966, the IWRs have been functioning as stagnant water reservoirs. Since 1965, the Techa River was conditionally assumed to rise from the dam of reservoir B-11 (Fig. 2). The drainage of the Kasli-Irtyash lake system takes place through the bypass canals (BC).

From dam P-11 to the village of Muslyumovo, the river flows through a wide valley with numerous swamps. The floodplain is mainly composed of peaty soils. The river bottom is peaty-silty and uneven. Along the river stretch from the village of Muslyumovo and further on, the river flows through a flat even country. The surface of the floodplain is meadowy, formed by sandy-loam soils. The bottom of the river is sandy, miry and loamy at places. In

the lower reaches of the river, the width of the valley ranges from 240 m to 2 km. The floodplain is meadowy, loamy, usually flooded during high water. The watercourse is moderately winding. The river is mostly supplied with snow water. The swampy floodplain of the upper reaches retain a considerable amount of thaw water. The river's tributaries are water-short. Floods usually occur in April. Low water lasts till mid-October. Water discharge during the low water periods increases along the river length from 0.84 m^3/d near Muslyumovo to 2.62 m^3/d near the village Klyuchevskoye. The coefficient of ground water supply accounts for 10-30% of the total river drainage.

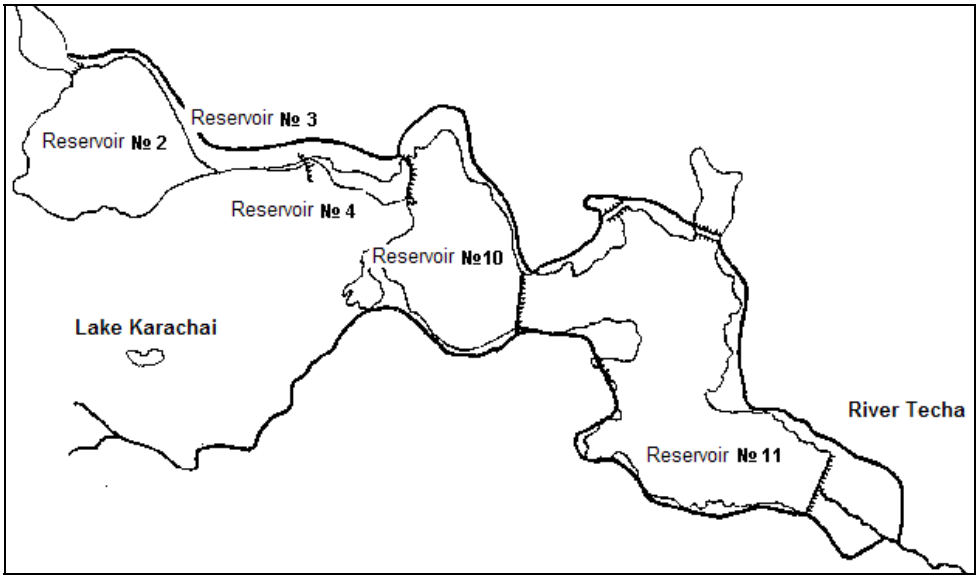

Fig. 2. Schematic map of the Techa cascade of reservoirs

The contamination of the Techa River was caused by the unavailability of reliable technology for reprocessing and storage of liquid radioactive waste (LRW). The approximate total releases into the Techa River over the period 1949-1953 were as follows: $^{89}Sr+^{140}Ba$ 240 kCi, ^{90}Sr -320 kCi, ^{137}Cs -350 kCi, REE 740 kCi, $^{95}Zr+^{95}Nb$ 37 kCi, $^{103}Ru+^{106}Ru$ kCi (Glagolenko Yu.G., 1966).

2.2 Radioactive contamination of water

During the initial period, the studies of radioactive contamination of water were based on measurements of β-emitting nuclide activity. The most well-systematized data were presented in (Marey A.N., 1959). The highest level of β-activity was observed in water in 1951; it was decreasing appreciably with advancing years and increasing distance from the site of releases (Fig. 2). The activity of α-emitters in water was significantly lower. The dependences governing the changes in the concentration of these emitters are similar to those identified for β-emitters. Reduced concentrations of radionuclides in the river water with increasing distance from the release site were accounted for primarily by the dilution processes in the water flow, sedimentation and radionuclide sorption by bottom sediments.

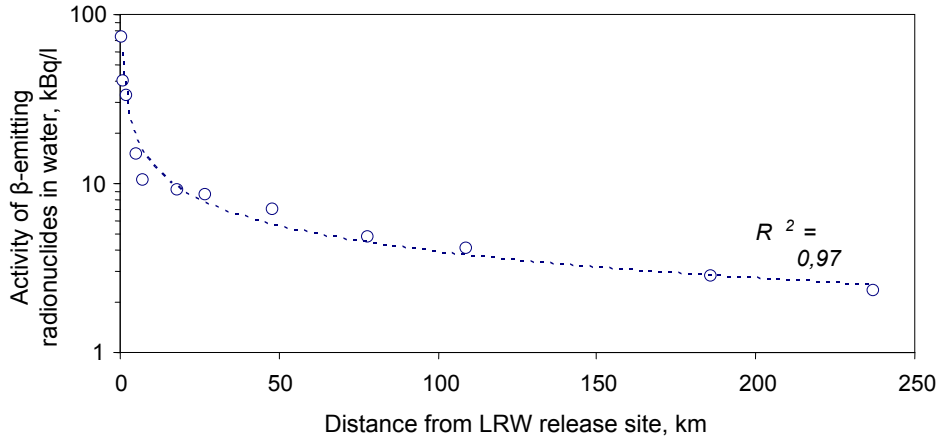

Fig. 3. Changes in volumetric activity of β-emitting radionuclides in Techa River water as a function of distance from LRW release site, 1952 (Marey A.N., 1959).

Nuclide composition of the river water sampled in the middle and lower reaches was for the first time determined in 1951 (Table 1). It was established that a significant proportion of activity of the radionuclides cesium, yttrium, cerium and plutonium is transported by the river stream down the rivercourse on clayey and sandy particles. The same applies to zirconium and niobium. The radionuclides strontium and ruthenium are transferred with river stream mostly in the dissolved state. The basic source of inflow of suspended particles is the surface-slope drainage from the catchment area.

Village name	β –activity in water, Bq/l	^{90}Sr	^{137}Cs	REE
Brodokalmak	$3.1 \cdot 10^4$	54.2	14.0	34.0
Bisserovo	$1.7 \cdot 10^4$	50.0	32.0	19.3
Pershinskoye	$2.1 \cdot 10^4$	55.8	24.0	15.2

Table 1. Radiochemical composition of the river water as of 5.08.1951, %

The results of the researches conducted in 1963 showed that small amounts of radionuclides (from 0.001 to 0.014%) (Yu.G. Mokrov, 2002) were carried by the bottom alluvium to the Techa River.

The construction of the Techa cascade of water reservoirs for storing low-level sewage water and re-directing medium-level waste to Karachai Lake resulted in reduced concentrations of radionuclides in river water and bottom sediments. By that time, the radionuclides ^{90}Sr and ^{137}Cs became the most important contaminants of the Techa River. The long-term dynamics of radionuclide content measured in river water (e.g., at Muslyumovo) up to 1990 was characterized by persistent reduction in ^{90}Sr and ^{137}Cs concentration. Instability and

periodical increases in radionuclide concentrations have been observed in river water (Fig. 4) since 1994. In addition to that, ^{90}Sr concentration is stably exceeding the currently permissible level of 4.9 Bq·l1. Concentrations of ^{137}Cs in river water are less stable along the watercourse (Fig. 5), but the values of the volumetric activity of this radionuclide does not exceed the permissible concentration for drinking water (11 Bq·l^{-1}), the role played by ^{137}Cs in radiation exposure of the riverside population is not very significant.

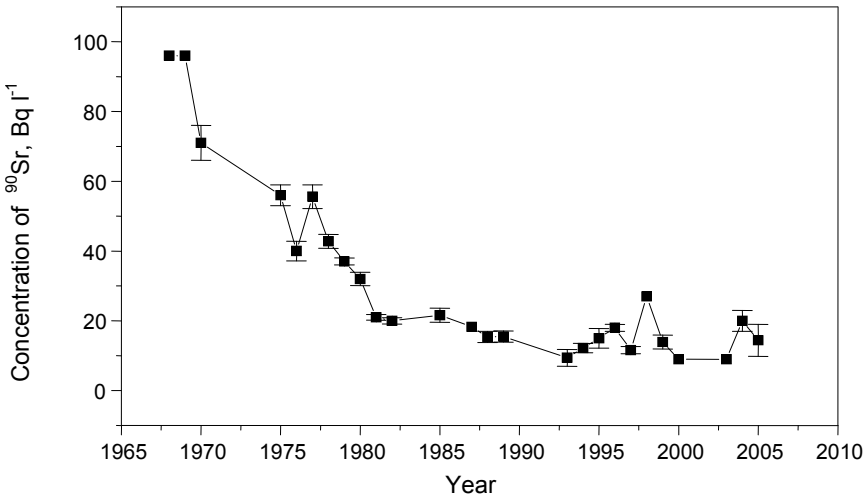

Fig. 4. Specific activity of ^{90}Sr in water of the river stretch vis-à-vis Muslyumovo.

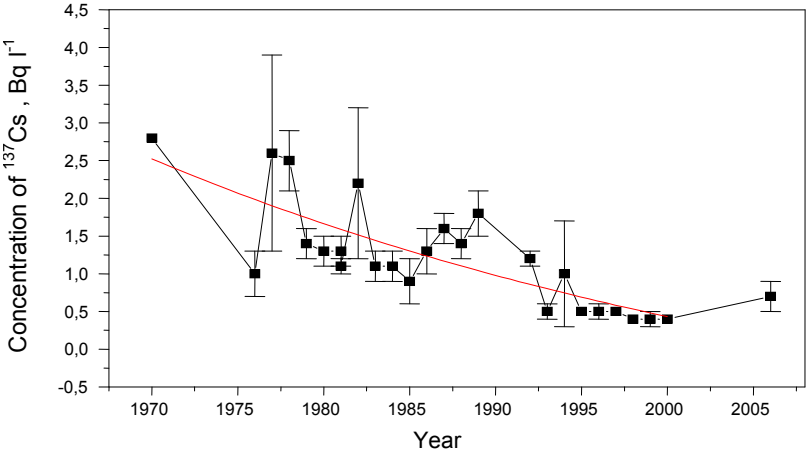

Fig. 5. Specific activity of ^{137}Cs in water of the river cross-section opposite Muslyumovo.

Data on contents of plutonium isotopes in Techa River water are scarce. Measurements of specific activity of $^{238,239,240}Pu$ in river water was initiated by the URCRM researchers in 1993

(Table 2). The main source of radioactive contamination of river water is the Techa cascade of reservoirs. Additional contamination is accounted for by desorption of radionuclides from the contaminated floodplain and the river bottom sediments.

In 2009, specific activity of tritium was for the first time determined in water of the Techa river (Fig. 6). Presented in the figure are concentrations of ^{90}Sr and tritium in water of the Techa River over its total length down to its confluence with the Isset River measured in samples taken within a week's time in August 2009.

Sampling site	Distance from dam 11, km	Years		
		1994	2000	2002
Assanov Bridge	4,5	3.2 ± 0.4 (3)	0.8 (2)	0.4 (2)
Chelyabinsk-Yekaterinburg Bridge	14	3.1 ± 0.8 (3)	0.46 ± 0.2 (4)	1.5 (2)
Muslyumovo	40	3.1 ± 1 (3)	0.14 (2)	-
Brodokalmak	71	-	-	-
Russkaya Techa	95	0.3 (2)	-	-
Nizhnepetropav-lovskoye	103	-	0.6 ± 0.2 (3)	-
Lobanovo	119	1.6 (2)	0.36±0.2 (3)	-
Verkhnyaya	141	2.5 (1)	1.0 (2)	-
Pershinskoye	170	2.7 (1)	-	-
Zatechanskoye	195	2.1 (2)	0.3 ± 0.1 (4)	-

Note: numbers of samples are given in brackets

Table 2. Specific activity of $^{238, 239, 240}Pu$ in Techa River water in 1994-2002, $Bq \cdot m^{-3}$

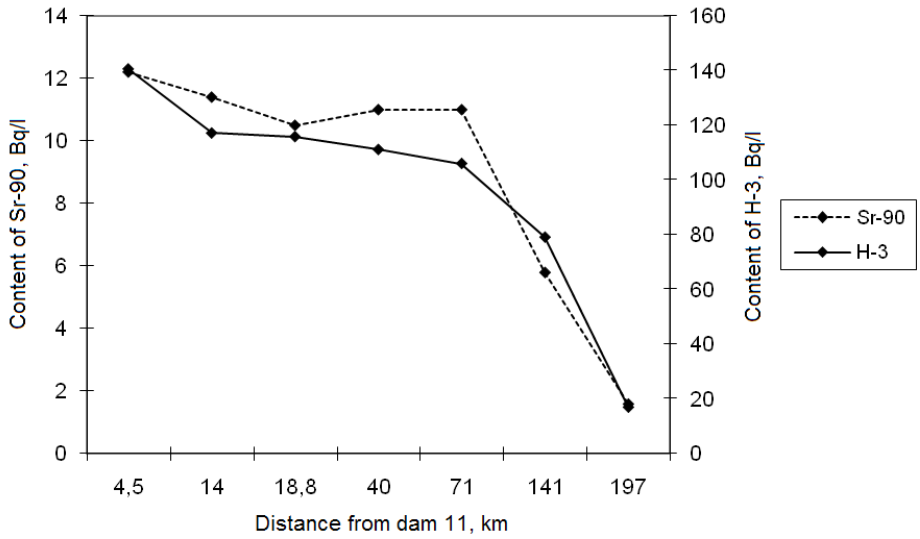

Fig. 6. Content of ^{90}Sr and ^{3}H in water of the Techa River

Concentrations of ^{90}Sr in water measured over the river length has changed from 12 to 1.6 Bq/l, tritium activity ranged from 140 to 17 Bq/l, which is 4-fold higher than the global level. The mean value of $^{3}H/^{90}Sr$ ratio in river water is 10.9±1.2. The total estimated carry-over of ^{90}Sr with the Techa river run-off into the Isset River over the period from 1958 through 2001 amounted 2.2×1014 Bq (Yu.A. Izrael, 2000).

Zone	Volumetric activity (Bq l^{-1})								
	^{90}Sr			^{3}H			^{137}Cs		
	Range of values	Mean	Median	Range of values	Mean	Median	Range of values	Mean	Median
TCR	983-5149	3007	3162	763-5935	2881	3251	1.9-914.5	102.3	32.9
Techa River	3.8-32.9	14.6	11.8	31-263.2	129	116	0.028-2.4	0.39	0.17
Drained area	0.05-173.9	27	14.1	6-15.8	10.2	9.5	0.05-1.51	0.55	0.49
Tributaries	0.15-0.18	0.15	0.15	9.5-9.8	9.7	9.5	-	-	-

Table 3. Summarized statistical data on volumetric activities of ^{90}Sr, ^{3}H, ^{137}Cs in water bodies of different origin

Table 3 shows summarized statistical data on volumetric activities of the radionuclides measured in the Techa River, TCR, surface and ground waters of the catchment area in the upper reaches of the river for the period from 2008 through 2010. In our subsequent calculations, the median values of ^{90}Sr and ^{3}H were used for TCR water, as well as the $^{3}H/^{90}Sr$ ratio equal to 1, median values of ^{90}Sr and ^{3}H for Techa River water, $^{3}H/^{90}Sr$ radioactivity ratio equal to 10, rounded median values of volumetric activity of ^{3}H equal to 10 Bq l^{-1} in samples of surface and soil water taken in the catchment area of the upper reaches of the river. Concentrations of radionuclides in water of the tributaries corresponded to the background values obtained for contents of radionuclides in surface waterways in the area influenced by the Mayak PA: ^{90}Sr - 0.15 Bq l^{-1}, ^{3}H - 9.7 Bq l^{-1}. Concentrations of ^{137}Cs were found to be below the detectability level of activity for the volumes sampled. The comparisons of volumetric activity values for ^{90}Sr and ^{3}H allowed tracing a direct dependence of the concentrations in river as well as in TCR. The only mechanism determining concentrations of ^{90}Sr in water is the process of intermix of waters with different initial radionuclide concentrations in different proportions. Also, uranium isotopes (^{234}U, ^{238}U) were used as a radionuclide of reference in relation to ^{90}Sr (^{234}U, ^{238}U). Uranium was selected because, unlike ^{3}H, its isotopes represent ions dissolved in water, as is ^{90}Sr (Mokrov Yu.G., 2000); and also because under oxidizing conditions characteristic of surface waters, uranium existing in the form of uranyl-ion (UO_2^{+2}) is weakly sorbed by floodplain soils and the river's bottom sediments. The difference between the values of the ratio $^{3}H/^{90}Sr$ for river and TCR waters (10 and 1) is accounted for by dilution of TCR effluent seepage with bypass canal waters in the proportion 1 to 10.

In order to assess the role played by the catchment area of the upper reaches and the river bottom sediments (0-40 km) in contamination of water with ^{90}Sr, we applied the two-component mixing model ($X_M=X_A\times f + X_B\times(1-f)$, where X_M is the end mixture, X_A and X_B are components, and f is the compound coefficient) using mixture parameters obtained for ^{3}H. ^{3}H and ^{90}Sr volumetric activities were measured at cross-sections located at 3.5 km and 40 km from dam 11, respectively. The values of ^{90}Sr volumetric activities for waters flowing

into the river from the catchment area of the upper reaches range from 4.3 Bq L^{-1} to 19.3 Bq l^{-1}, the average value amounting to 9.24 Bq l^{-1}. It was concluded based on the calculations that entry of 70% of the total activity of ^{90}Sr into the Techa watercourse results from TCR effluent seepage drained through the bypass canal system. The proportion of ^{90}Sr activity contributed by washing out of radionuclides from the floodplain and by desorption from bottom sediments accounts for 30%.

2.3 Contamination of bottom sediments with ^{90}Sr and ^{137}Cs

According to the data of the first investigations, the highest concentrations of radionuclides in bottom sediments were observed in the reaches close to the release site and in the area of Assanov swamps (30 km from the release site). The lowest level of contamination was registered over the last 40 km stretch down to the outfall. A large amount of activity was accumulated in the surface layer (Table 4) (Glagolenko Yu.G., 1966).

Distance from the release site, kn	Depth, cm		
	0-1	5-7	10-12
Release site	103045-486550	703-77071	262-62678
7	50098	8621	703
18	47101	16539	10730
33	6734	4625	37
48	1961	1184	59
78	2035	292	44
109	888	540	373
138	1221	355	222
186	255	215	133
223	307	252	-

Table 4. Specific β-activity of bottom sediments, August 1952, kBq·kg^{-1} (Glagolenko Yu.G., 1966).

Distance from the dam 11, km	^{137}Cs	Σ Pu
1	2	3
1.8	27491	61
2.2	14874	33
3.8	45103	133
5.8	23199	27
6.7	7104	24
13.0	15392	57
14.0	17020	60
23.0	32190	78
41.0	666	4
44.0	8325	11

Table 5. Distribution of contamination density in bottom sediments along the length of the river in 1991-1992, kBq·m^{-2}

It was established that radioactivity is best of all accumulated by silt and clay material, while sandy soils manifest a lower rate of accumulation. It was demonstrated that ^{137}Cs and plutonium isotopes were intensively sorbed by all varieties of soils. ^{90}Sr, too, is actively sorbed by soils, but it easily enters into exchange reactions which determines its high migration potential (Saurov M.M., 1968). ^{90}Sr and ^{137}Cs contamination densities in bottom sediments in the early 1990s are shown in Table 5. During spring floods, silt sediments contaminate the surface of the floodplain which maintains the high levels of contamination of the flooded riverside valley. The levels of silt and water radionuclide contamination are interdependent over the river course. The contents of radionuclides in silt and in water, from dam 11 up to the river outfall are steadily decreasing (Figure 7). Specific activity of ^{137}Cs in silts is about 5-fold higher than in water, that of ^{90}Sr if 3-fold higher. Compared to ^{90}Sr, ^{137}Cs contamination densities for silts are about 2-fold higher over the total length of the river.

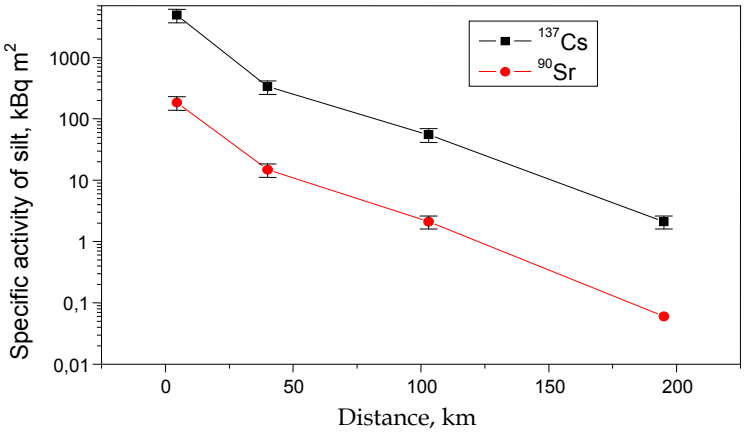

Fig. 7. ^{90}Sr and ^{137}Cs contamination densities measured in Techa River silts at different distances from dam 11.

Four decades after termination of intensive discharges of radioactive waste into the river, radionuclides deposited in sandy and silty soils migrated to the depth of over 35 cm. The results of the vertical distribution of the radionuclides of interest in the upper reaches of the river are presented in Figures 8 and 9. Compared to ^{137}Cs, the distribution of ^{90}Sr in the bottom soil profiles is more uniform. Maximum values of contamination densities for these radionuclides are in general observed in 0-10 cm layers of soil. ^{137}Cs is characterized by a more dramatic decline of contamination density values in lower layers of soil (at the depth of 20-35 cm). In the upper reaches, additional inflow of radionuclides due seepage from TCR and washout of radioactivity from the lands adjacent to the river is observed; in the mid-stream area the contribution of desorption processes from the upper layer is larger.

With distance from the release site, the proportion of exchangeable and mobile forms of ^{90}Sr is increasing, on the contrary, the proportion of poorly-accessible forms of ^{90}Sr is decreasing.

2.4 Contamination of the floodplain with ^{90}Sr and ^{137}Cs

Spring overflows of the Techa, and particularly the flood of 1951 contributed to intensive radioactive contamination of the riverside area. The studies of the contents of radioactive substances in floodplain soils started in 1951. The results obtained allowed an insight into the patterns and intensity of the riverside contamination (A.N. Marey, 1959). The width of the floodplain where radioactive contamination was detected did not usually exceed 150-200 m (Table 6). In the upper reaches, in the Assanov swamps area, the overflow reaches 3000 m. A consequent decrease in the levels of floodplain contamination with increasing distance from the release site, and a decrease in the ^{137}Cs/^{90}Sr ratio should also be noted (Table 7).

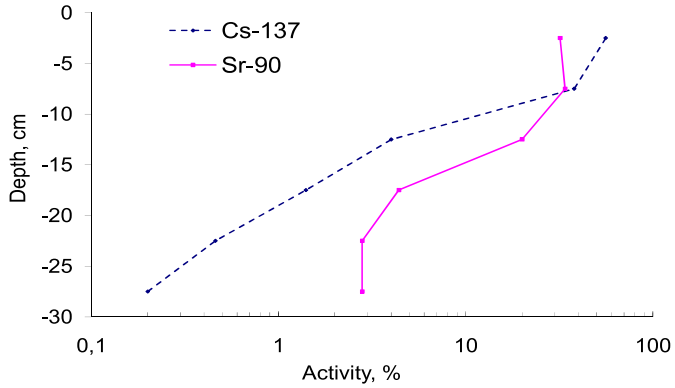

Fig. 8. Distribution of radionuclides along the depth of sandy bottom sediments in the upper reaches of the river.

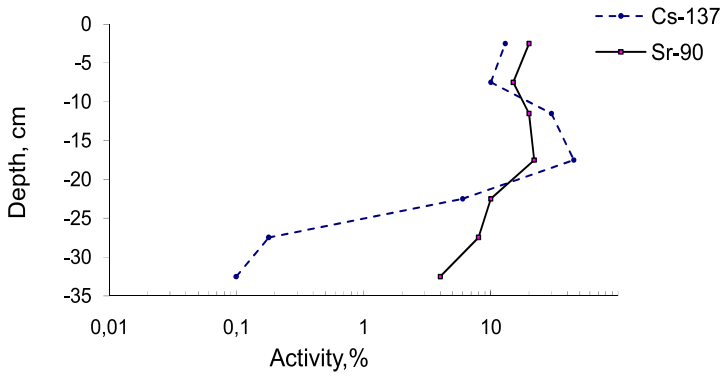

Fig. 9. Distribution of radionuclides along the depth of silty-clayey bottom sediments in the upper reaches of the river.

Distance from the release site, km	Bank	End of the inundable floodplain
Release site	7548	141
18	2442	18
33	407	22
48	74	15
78	481	4
105	200	60
109	63	33
128	33	4
138	4	23
212	3	22

Table 6. Specific β-activity in the surface layer in 1952 - 1954, $kBq \cdot kg^{-1}$ (Marey A.N., 1959)

Changes in the width of the riverside area is, as a rule, determined by soil relief; maximum concentrations of radionuclides are registered in lowland areas.

Distance from dam11, km	^{137}Cs	^{90}Sr	^{137}Cs / ^{90}Sr
67	210	150	1.4
102	95	60	1.5
135	80	80	1.0
152	35	100	0.3
237	5	42	0.1

Table 7. Changes in contents of ^{137}Cs and ^{90}Sr in floodplain soils along the river course, $Ci \cdot km^{-2}$ (Marey A.N., 1959)

The first maximum level of floodplain contamination density is observed in the area of Assanov swamps: from dam 11 to the distance of 7.5 km. The second peak takes place in lower reaches, at the distance of 37 km from the dam in the area of Muslyumovo swamps. In these areas, the incessant winter run-off of radioactive substances with TCR waters is accumulated. Most thoroughly the floodplain soils are washed during high waters. Surface-downslope waters wash upper layers of the soil, flood waters wash upper layers of the flooded river bank, and ground waters wash deeper layers of the floodplain soils.

With distance along the watercourse, the levels of floodplain contamination with ^{137}Cs are appreciably decreasing. In the swamps of the upper reaches, the values of contamination with the radionuclides of interest amount to 150-550 $Ci \cdot km^{-2}$, the values for the middle reaches are 20-30 Ci km^{-2}, and the respective value for the area close to the estuary is 5 $Ci \cdot km^{-2}$. The dynamics of reduction in ^{90}Sr contamination density values assumes a more monotonous character. The results of measurements of ^{137}Cs and ^{90}Sr contamination densities conducted by us in the floodplain in 2005 are presented in figure 10. It can be seen that the level of floodplain contamination with ^{137}Cs is higher than the respective value obtained for ^{90}Sr, with the exclusion of the last locality (village Zatechanskoye).

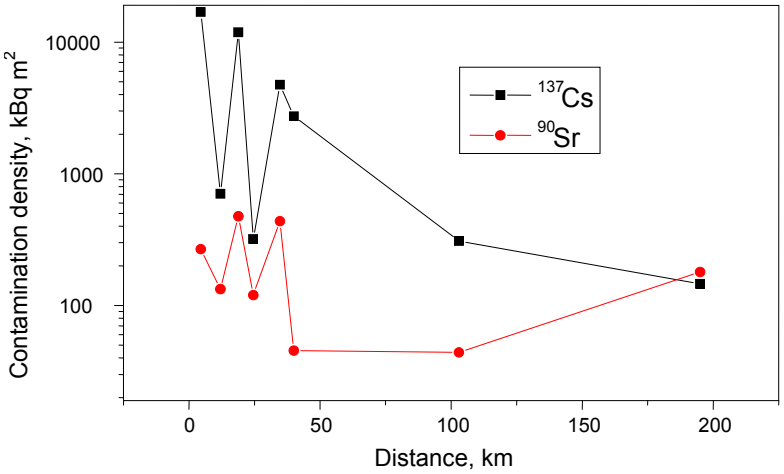

Fig. 10. ^{137}Cs and ^{90}Sr contamination densities in the floodplain along the watercourse, kBq·m^{-2}

The study of ^{137}Cs distribution by layers of the floodplain along the watercourse (Table 8) has demonstrated that the upper 20 cm layer is contaminated to the highest extent. However, the measurements conducted in the village Zatechenskoye where sandy soils predominate, the concentrations of radionuclides in the lower and surface layers are actually the same. As has been shown by measurements at most of the sampling sites, the upper 0-5 cm soil layer contains a smaller amount of ^{137}Cs as compared to the underlying layer. This fact indicates that there has been no intensive contamination of the floodplain with ^{137}Cs in the recent years. An extreme non-uniformity in the distribution of ^{90}Sr by soil profiles (Table 9) in different reaches of the river can be accounted for by different physical-chemical and morphological properties of the floodplain soils.

Sampling site	Distance from dam 11, km	Position of layers, cm			
		0-5 см	5-20 см	20-40 см	40-60 см
Assanov bridge	4.5	1438.7	1011.0	1595.3	1059
Maloye Taskino	12	230.7	61.1	7.9	6.9
Nadyrov bridge	18.8	593.2	10084.4	1281.3	439.0
6 km below Nadyrov bridge	24.5	129.6	28.4	53.7	44.9
Buslyumovo swamp	34.7	660	62.1	784.3	354
Muslyumovo	40	277.1	95.4	17.7	10.9
Nizhne-Petropavlovskoye	103	8.1	65.4	19.3	6.4
Zatechenskoye	195	11.3	99.8	31	101.7

Table 8. Distribution of ^{137}Cs by floodplain soil layers over the length of the river, kBq·m^{-2}

Most of the floodplain soils (Assanov Bridge, Nadyrov Bridge) are characterized by depletion of the upper 0-10 cm layer, presence of maximum ^{90}Sr concentrations at the depth of 10-30 cm with a dramatic drop in concentrations measured deeper along the profile. This is caused by washing of the upper layer with the surface waters. In the locality of Muslyumovo, the distribution of ^{90}Sr by soil layers is influenced by the continuous run-off from the overlying swampy layers. Further along the watercourse, the processes of cleaning radionuclides from the upper soil layers and a more uniform distribution along the depth of the soil become prevalent. (Nizhnepetropavlovskoye and Zatechenskoye).

Sampling site	Distance from dam 11, km	Position of layers, cm			
		0-5 см	5-20 см	20-40 см	40-60 см
Assanov bridge	4,5	20	78,5	112,6	49,4
Maloye Taskino	12	51,8	143,8	12,6	2,1
Nadyrov bridge	18,8	27,2	394,1	136,9	37,6
6 km below Nadyrov bridge	24,5	46,2	39,2	26,6	8,3
Buslyumovo swamp	34,7	27,7	72,1	233,9	103,7
Muslyumovo	40	10,7	33,8	6,5	2,1
Nizhne-Petropavlovskoye	103	2,5	16,4	3,6	0,74
Zatechenskoye	195	26,3	239,8	321,1	24,5

Table 9. Distribution of ^{90}Sr by floodplain layers along the watercourse, kBq·m^{-2}

From 1991 through 2005, the total content (reserve) of ^{137}Cs at the sampling site (Fig. 11) decreased from 574.5 MBq·m^{-2} to 52.8 MBq·m^{-2}, accumulating mainly at the depth of 30 cm, and lower, at 40 cm above the aquifer. The total content of ^{90}Sr in soil samples taken in the floodplain (Fig. 12) decreased from 14.4 to 8.8 kBq·m^{-2}. Over this period, equalization of the contamination was noted at the depth of 55 cm, while in the lower layer an increase in contamination level was observed, with radionuclides moving all the way to the aquiferous layer, obviously due to intensive seasonal washouts of the bog soils.

In 2009, samples were taken in the floodplain area close to Assanov swamps at the depth of 80 cm within 30, 70, 106 and 250 m of the shoreline. Below, an aquifer composed of blue clay was situated. It was shown that ^{137}Cs and ^{90}Sr contamination densities in the floodplain soil (figure 13) were declining depending on distance from the shoreline. The reserve of ^{137}Cs exceeds that of ^{90}Sr at any distance, and their ratio changed at different distances from 22-2 times. The distance from 30 to 70 m is characterized by the lowest levels of contamination due to wash-off by ground waters.

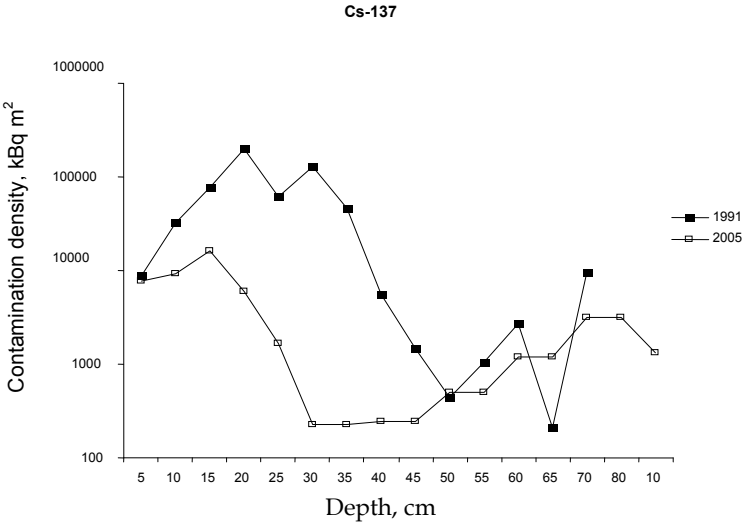

Fig. 11. Contamination densities for ^{137}Cs in the Techa floodplain area at sampling site "Assanov swamps" within 6.5 km of dam 11, 10 m from the shoreline.

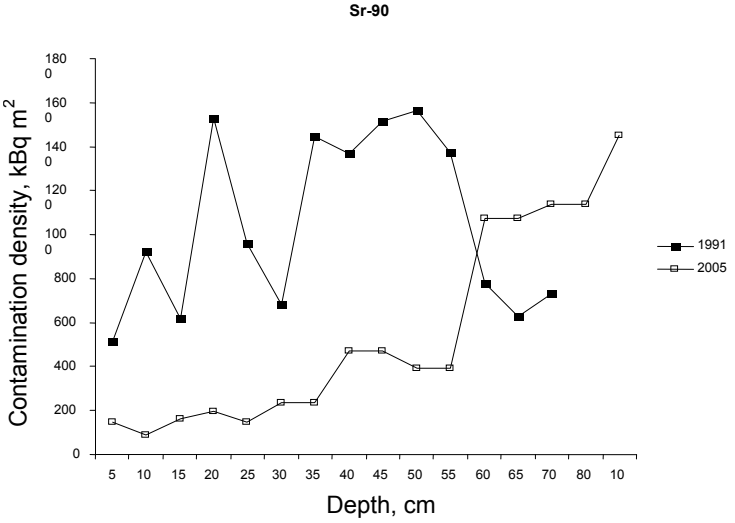

Fig. 12. ^{90}Sr contamination densities in the Techa River floodplain at 6.5 km from dam 11, left bank, within 10 of the shoreline.

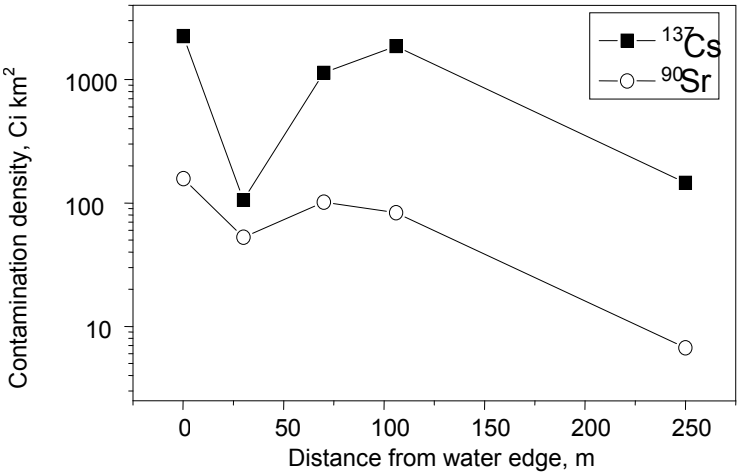

Fig. 13. ^{137}Cs and ^{90}Sr contamination densities for soils of Assanov swamps, Ci km^{-2}

In samples of floodplain taken in the area of Assanov swamps at different depths and sites, prevalent are water-soluble forms of radionuclides deposited in deep layers (70-80 cm) of riverside soil (up to 26%) in localities where they migrate in the direction of the watercourse. A slight increase in exchangeable and mobile forms of ^{137}Cs in the lower layer was observed.

In 2005 and 2006, samples of ground and surface waters were taken in the area of Assanov and Muslyumovo swamps with the aim to determine the actual scope of influence exerted by deposited radionuclides on concentrations of radioactivity in swamp water. During cold seasons, when migration processes are at their minimum, concentration of ^{90}Sr in flow channels is high, reaching 187 Bq l^{-1}, the concentration level registered in ground water is 14-19 Bq·l^{-1}. In spring, in the portion of the swamp area, where there is no distinctly outlined water flow, ^{90}Sr concentrations in water amount to 80-100 Bq·l^{-1} in localities far removed from the river banks. In flow channels with a sufficiently dynamic water flow, radionuclide concentrations reach about 40 Bq·l^{-1}. In Muslyumovo swamps, water sampled in dead channels contains 11-25 Bq·l^{-1} of ^{90}Sr with radionuclide concentration level in river water of 6.2 Bq·l^{-1}. The data obtained point out to the fact that it is mainly ^{90}Sr which is leached from the floodplain to water, the same applies to ^{137}Cs, but to lesser extent.

Thus, it was demonstrated that the floodplain and bottom deposits are the key source of secondary contamination of the river water with radionuclides. The level of radioactive contamination of these river components is, in its turn, determined by the radioactive run-offs from the Techa cascade of reservoirs.

3. East-Urals Radioactive Trace

3.1 Radioactive contamination of soil, vegetation, food products with ^{90}Sr and ^{137}Cs in the early years after the 1957 accident

A distinguishing feature of the radioactive emission on the EURT is the presence in it of all basic uranium fission products and a minimum content of the ^{137}Cs (Table 10).

Radionuclide	%	Scope of release, PBq
^{89}Sr	traces	-
$^{90}Sr+^{90}Y$	5.4	2.0
$^{95}Zr+^{95}Nb$	24.8	18.4
$^{106}Ru+^{106}Rh$	3.7	2.7
^{137}Cs	0.36	0.26
$^{144}Ce+^{144}Pr$	66.0	48.7
^{147}Pm, ^{155}Eu	traces	-
^{239}Pu	traces	0,0014

Table 10. Characterization of the radioactive releases and the initial reserve of radionuclides on the EURT outside the Mayak PA industrial site (Avramenko V.I. et al., 1977)

The settling of the radioactive mixture from the cloud which was wind-drawn in the north-eastern direction from the explosion site resulted in the formation of the East-Urals Radioactive Trace (EURT). The Trace encompassed parts of Chelyabinsk, Sverdlovsk and Tyumen oblasts. The length of the EURT is 300 km, its width is 30-50 km. According to refined data, (Korsakov Yu.D. et al., 1996), in 1957 the area with contamination density (η) for ^{90}Sr >2 Ci km^{-2} was 560 km^2, that with η>12 Ci km $^{-2}$ was 230 km^2, with η>50 Ci·km^{-2} – 120 km^2, with η>200 Ci·km^{-2} –50 km^2, with η>800 Ci·km^{-2} – 16 km^2, and with η> 2 000 Ci·km^{-2} – 8 km^2. There were over 200 populated localities in the Trace area, including several towns and industrial communities. At early time after the accident, the population of the EURT area was exposed to radiation-related hazards, including, in the first place, external exposure to γ-radiation due to prevalence of γ-emitting nuclides in the deposited mixture and, in the second place, internal exposure resulting from intakes of radionuclides contained in food products produced in the localities. As the activity of γ-emitting nuclides (which decayed almost completely 6-7 years after the accident) decreased, the radiation hazards were mostly determined by the radionuclide ^{90}Sr.

The first measurements conducted on the contaminated territory showed that the γ-radiation dose rate was proportional to the distance from the accident site. According to measurements conducted during the first year after the accident in the area with contamination density amounting to 1 Ci km^{-2} for^{90}Sr, the dose in air due to γ-radiation was 1R (G.N. Romanov, 1963). Direct exposure to β-radiation only occurred in areas with contamination density of over 1 500 Ci·km^{-2} for ^{90}Sr. According to data presented in (A.Ya. Kogotkov, 1968), a decrease in relative contents of ^{90}Sr in the composition of the mixture depending on distance from the accident site, and, respectively, an increase in the contents of ^{144}Ce and ^{137}Cs. Soil contamination densities in some localities may differ by an order of magnitude, or greater.

The coniferous woods were affected most heavily. At the distance of 12.5 km from the contamination source, a total loss of pine woods was registered in the summer of 1958. Mass loss of birch forests were only observed in areas with contamination densities of over 4 000 Ci·km^{-2}. Migratory birds were only affected in the spring of 1958, after dose rates in the tree crowns had decreased 10-fold. A reduced number of bird's nests was noted in the areas with contamination level of over 2 000 Ci·km^{-2} for ^{90}Sr. No loss of animals was registered. In the

ensuing years, due to the fact that the contaminated area was made into a sanitary-protection zone, the number of hares, roes and elks increased considerably.

During the first days after the accident, radioactive contamination of grass estimated relative to 1 Ci/km^2 of soil contamination was $1.5 \cdot 10^6$ decay min^{-1}·kg^{-1}. Radioactive contamination of individual food products was very high (Table 11). Since cattle and other domestic animals were fed contaminated forage, contamination of milk and meat was of structural rather than superficial nature. It should be noted that contamination of milk was registered as early as the first 2-3 days after the accident, and that of meat on days 10-12 (R.M. Alexakhin et al., 2001).

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Locality	Soil	Water from water sources	Grass	Milk	Meat	
					muscles	bones
1	2	3	4	5	6	7
Berdyanish	21016	52	360380	115	58	92
Saltykova	33760	104	97162	218	-	-
Galikayeva	2405	104	34262	-	6	74
Kasli	18	-	32	-	-	-
Russ. Karabolka	4810	42	22200	-	6	137
Yugo-Konevo	192	0.02	15207	-	1.2	10
Yushkovo	3	-	-	3.1	3	71
Boyevskoye	266	-	755	-	-	-
Bagaryak	74	-	8399	4.4	-	-
Kamensk-Uralsky	-	0.004	-	-	2.8	1.5
Pozarikha	-	-	440	-	-	-

Note: the results include ^{40}K activity

Table 11. β-activity in food products and environmental entities in the EURT area as of 20.10.1957, kBq/kg, l

Food products were the main contributor to radiation exposure of the population. The main cause of the reduction in contents of radionuclides in food products over time is the reduction of radionuclide contents in soils. Table 12 shows the basic composition of radionuclides observed in food products and the dynamics of cleaning the food from radionuclides. Among those radionuclides, ^{144}Ce+^{144}Pr prevailed during the first 3 years, later on ^{90}Sr gained priority.

Beginning from the spring-summer season in 1958, an additional contamination of vegetation and agricultural plants occurred due to a downwind migration of radionuclides from areas with a higher level of contamination density. The proportion of surface contamination of grain crops accounted for 10-15% (P.P. Lyarsky, 1962).

Contamination levels measured for food products of the first post-accident harvest reaped in the fields situated in the Trace zone with contamination densities of 0.2-0.5 Ci·km^{-2} for ^{90}Sr or higher, were higher than the permissible limit legally valid at that time: 1300 decay min^{-1}·kg^{-1} (22 Bq·kg^{-1}).

Time after the accident, years	Key radionuclides, %					Other
	$^{90}Sr+^{90}Y$	^{137}Cs	$^{144}Ce+^{144}Pr$	$^{95}Zr+^{95}Nb$	$^{106}Ru+^{106}Rh$	
1 (1957)	10.1	1.5	76.9	10.1	1.3	<1%
2 (1958)	12.9	2.8	82.4	0.2	1.5	
3 (1959)	7.0	8.0	80.6	-	4.2	
5 (1961)	90.3	2.7	6.2	-	0.6	
10 (1966)	98.8	1.2	-	-	-	

Table 12. Composition of radionuclide observed in food products at different time after the accident

3.2 Current levels of soil contamination

The studies conducted in 2006-2009 in the villages Allaki, Bagaryak, Bulzi, Tartar Karabolka and Yushkovo located around the perimeter of the EURT of sanitary-protection zone. The soils of the pasture lands adjacent to these villages are mainly dark-grey clay and leached chernozem.

The mean dose-rate value of gamma-radiation in the localities of Karabolka, Musakayeva and Bagaryak is 0.12 μSv·hr^{-1} which is comparable to natural background values for Chelyabinsk oblast. The mean pasture land contamination density for Sr^{90} in the localities of Tatarskaya Karabolka and Musakayeva is 5.9 kBq·m^{-2}, that for Bagaryak is 2-2.5 times higher. Pasture land contamination density for ^{137}C is 12.9-24.8 kBq·m^{-2}. Mean contamination densities measured in soils of kitchen gardens attached to houses in Bagaryak was 77.5 kBq·m^{-2}. Mean specific activity in grass measured in different areas along the EURT axis ranged from 11 to 15 thousand Bq·kg^{-1} for Sr^{90} and from 3 to 60 Bq·kg^{-1} for ^{137}C. Forms of Sr^{90} and ^{137}C encountered in soils sampled in the frontal portion of the Trace (close to Alabuga Lake) have been identified (Table 13). Attention is drawn to the increased number of exchangeable forms of Sr^{90} and mobile forms of ^{137}C. During the early years after the fallout, the total amount of Sr^{90} was only encountered in soluble state. A decade later, the proportion of exchangeable forms accounted for 65-75%, and the value has not changed since then.

According to data presented by V.V. Martyushov et al. (1996) 36 years after the contamination, the content of exchangeable forms of ^{137}C and plutonium did not exceed 3% and 1%, respectively. The proportion of poorly accessible forms of ^{137}C and plutonium reached 95-98%. The content of water-soluble forms of ^{137}C and plutonium accounts for less than 1%. Water-soluble forms of Sr^{90} are mostly found in cationic compounds (72-76%).

Radionuclide	Physical-chemical forms of radionuclides , %			
	Water-soluble	Soluble in 1H CH_3COONH_4 (exchangeable)	Soluble in 1H HCl (mobile)	Solid residue (poorly accessible)
^{90}Sr	2.8 ± 0.5	73.2 ± 0.4	16.3 ± 0.6	7.7 ± 0.4
^{137}Cs	2.7 ± 0.4	3.1 ± 1.0	19.4 ± 1.5	74.8 ± 2.5

Table 13. Forms of radionuclides identified in the upper 5-cm layer of soil in the vicinity of Alabuga Lake

In 2009, soil samples were taken at 16 sampling sites at a distance of 20 km from the Mayak PA perpendicular to the EURT axis. Five of those sites are situated in a birch forest. The total contamination density in the soils of the rhizogenic layer and forest litter was, on the average, equal to 737 kBq/m^2 for ^{90}Sr, and 41.2 kBqк/m^2 for ^{137}Cs. It should be noted that 10.5% of the contamination density for ^{90}Sr and 14.1% of contamination density for ^{137}Cs are contributed by the debris layer. At 10 points situated in the hayfield extending from the forest up to Alabuga Lake ^{90}Sr contamination density ranged from 161 to 350 kBq/m^2, ^{137}Cs contamination density measured in the hayfield ranged from 34 to 93 kBq/m^2. Contamination densities measured in the wood were higher than those measured in the hayfield.

At a distance of 30 km from the Mayak PA in an area along the Trace axis samples of soils were taken at a depth of 10 cm. ^{90}Sr contamination density was found to range from 2.2 kBq/m^2 to 55.9 kBq/m^2, that for ^{137}Cs ranged from 2.2 to 50.7 kBq/m^2. At the periphery of the Trace, contamination density for ^{137}Cs exceeded that for ^{90}Sr 2-5-fold. The contribution made by the debris layer varies significantly. In afforested and steppified areas with well developed steppe debris layer, the contribution of the debris layer to the contamination density for ^{90}Sr reaches 37%, for ^{137}Cs – 8.8%.

At a distance of about 40 km from the Mayk PA at a right angle to the Trace axis, ^{90}Sr and ^{137}Cs contamination density distribution was determined for the ploughed layer (0-20 cm). As of the date of the measurements, there were only arable lands that had no sod cover were available for measurements. The highest measured contamination density was 162 kBq/m^2 for ^{90}Sr and 24.4 kBq/m^2 for ^{137}Cs

Samples of sod-podzol soil were taken on the Trace axis close to Bolshoi Irtysh Lake (about 55 km from the Mayak PA). Contamination densities in the upper 0-20 cm layer and in the ground litter was 308 kBq/m^2 for ^{90}Sr, and 20 Bq/m^2 for ^{137}Cs. It is noteworthy that 11.2% of ^{90}Sr and 18.0% of ^{137}Cs are contributed by the ground litter. The total contamination densities in soil (0-20 cm) and ground litter along the Trace axis are presented in Figure 14.

The studies performed have shown that soil contamination densities close to the EURT axis are still high, even at the present time; besides, in a number of cases a considerable portion

of radionuclides is contained in forest debris layer or steppe litter. Contamination levels mostly depend on the distance from the contamination source, i.e. from the Mayak PA. The highest levels of radioactive contamination were identified in the 0-5 cm layer of soil. Below the root-inhabited layer, 2.7%-57.2% of ^{90}Sr and 28.4%-41.1% of ^{137}Cs are deposited. In all types of soil, ^{90}Sr is encountered in biologically accessible forms, and, ^{137}Cs is contained in poorly-soluble compounds.

3.3 Vertical migration of ^{90}Sr and ^{137}Cs through soil profiles

To allow assessment of soil contamination levels, two main parameters are used, viz., specific activity of the radionuclide in soil (Bq·kg^{-1}) and contamination density (Bq·m^{-2}) which takes into account the total contamination density in all *n* soil layers. There is no direct relationship between these two parameters. It was established that over the time period since the 1957 accident the radionuclides had migrated to a significant depth. Actually, samples taken at all the sampling sites showed that the highest level of contamination with ^{137}Cs and ^{90}Sr was detected in the upper level of soil and in debris layer (in meadow soils it was found in sod cover and in thick felt of the steppe). The ratio of radionuclide specific activity in the 0-10 cm layer to activity in the 10-20 cm layer did not depend on the summarized contamination density. The value of this ratio is mostly influenced by the type of the ecosystem: the mean value of this ratio for ^{137}Cs in forest ecosystems is 20.4±4.4, and in meadows it is 2.9±1.6. For ^{90}Sr, the differences are insignificant: 3.4±0.9 in forests, and 2.8±1.5 in meadows. The ratio of specific activity of ^{137}Cs in debris cover to activity in the 0-10 cm soil layer amounted on an average to 0.5±0.1, and for ^{90}Sr to 1.5±0.2. Although the specific activity of ^{137}Cs and ^{90}Sr in the litter layer is sufficiently high, it does not exert substantial influence on the summarized contamination density since the volume weight of the litter layer is by two orders of magnitude lower than the volume weight of soil.

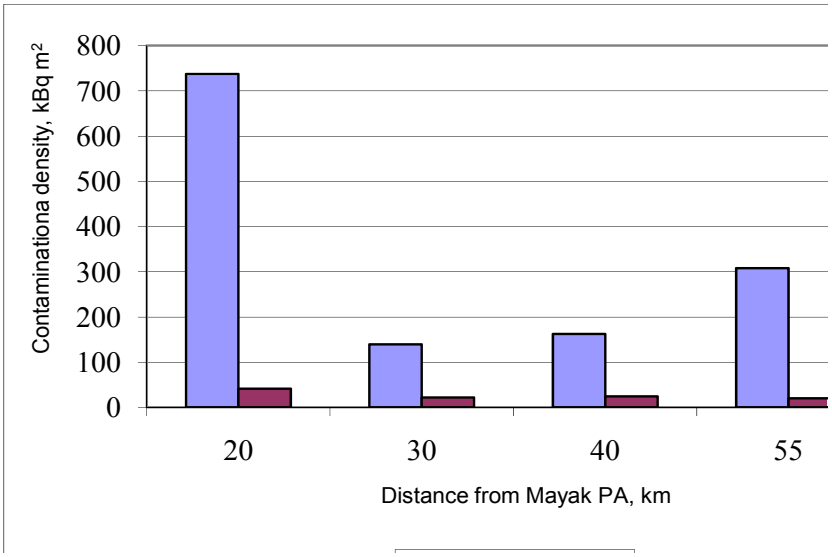

Fig. 14. Contamination density of soil (0-20 cm) and litter layer along the EURT axis as a function of distance from the source of contamination.

Distribution of ^{90}Sr and ^{137}Cs in soil profiles was studied in 2008-2010 for 3 types of soils: gray forest, sod-podzol and chernozem. Gray forest soils are most prevalent in the forest-steppe zone. They are formed in leaf woods and mixed woods of the Trans-Urals region. Distribution of ^{90}Sr and ^{137}Cs in gray forest soil profiles was determined at a sampling site within 20 km of the Mayak PA. The highest specific activity of ^{90}Sr (1.6×10^4 Bq·kg^{-1}) and ^{137}Cs (1.3×10^3 Bq·kg^{-1}) was measured in the lower layer of the forest litter which contains a half-decayed tree waste. ^{90}Sr activity in the upper layer of the forest litter is actually equal to activity found in the 0-5 cm soil layer, ^{137}Cs activity is 2.7 times lower. However, the volume weight of the forest litter is low that is why contamination density summarized for its upper and lower layers is lower than that in the underlying 0-5 cm soil layer where the main amounts of ^{90}Sr (60%) and ^{137}Cs (50%) are deposited. The tree waste of 2008 sampled on October 31 demonstrated an increase in contamination density of the tree waste by 2.4 kBq·kg^{-}m^{-2} for ^{90}Sr which accounts for 2.1% of the tree waste contamination measured in spring of 2008. Over the 50 years since the accident, no shift of the maximum values with increasing depth through the soil profiles has taken place. At the same time, both ^{90}Sr, and ^{137}Cs, though in small amounts, have at least, reached the depth of 170-175 cm. It should be added that ^{137}Cs which is considered to be less capable for vertical migration, is evidently migrating more actively than ^{90}Sr, 2.7% of ^{90}Sr, and 28.4% of ^{137}Cs are deposited at a depth of over 20 cm.

Sod-podzol soils which most often are found in the northern part of the Trace, are formed under coniferous woods. The distribution of ^{90}Sr and ^{137}Cs along the profile of sod-podzol soils is comparable to the distribution observed for gray forest soils. The highest specific activity of ^{90}Sr (1.0×10^4 Bq·kg^{-1}) and ^{137}Cs (1.2×10^3 Bq·kg^{-1}) was found in the lower part of the forest waste, the highest contamination density for ^{90}Sr (39.6%) and ^{137}Cs (40.4%) was measured in the upper 0-5 cm soil layer. The forest waste and soil layer up to 20 cm in depth contain 96.2% of ^{90}Sr and 58.9% of ^{137}Cs. At the depth of over 20 cm 3.7% of ^{90}Sr and 41.1% of ^{137}Cs are deposited below 20 cm. As can be seen from the comparison of the distributions, the mobility of ^{90}Sr in the sod-podzol soils differs insignificantly from that registered in gray forest soils, the mobility manifested by ^{137}Cs is significantly higher. Already at the depth of 25-30 cm, the specific activities of ^{90}Sr and ^{137}Cs actually differ one from the other, however, in lower layers ^{137}Cs takes the first position.

There occur in steppefied areas of the EURT weak northern or leached chernozems which have been ploughed up for a long time period. It should have been expected, therefore, that the distribution of radionuclides through the 0-20 cm plough-layer would be more uniform and speedy. The site for taking soil samples from the chernozem profile is located close to the EURT axis, however, the contamination density for ^{90}Sr turned out to be low, viz., 17.8 kBq·m^{-2} for ^{90}Sr and 40.2 kBq·m^{-2} for ^{137}Cs over the whole profile. ^{137}Cs was found to be significantly prevalent in each layer.

The highest specific activity for ^{90}Sr (92.2 Bq·kg^{-1}) and ^{137}Cs (161 Bq·kg^{-1}) was observed in the debris cover which was characteristic of other soil types too. The highest contamination density was measured in the 0-5 cm layer for ^{90}Sr (10.4%) and in the 5-10 cm layer (10.9%), and for ^{137}Cs in the 0-5 layer (21.4%). At the depth of over 20 cm, 57.2% of ^{90}Sr, and 37. 4% of ^{137}Cs were deposited.

The analysis of ^{90}Sr distribution patterns in the EURT soils were made using data of researches conducted in 1963-2008 (figures 15-17). It can be seen that the distribution of ^{90}Sr in the profile of the 30 cm layer is well described by the following exponential function: $y=ae^{-bx}$, where y is the content of radionuclides calculated as percentage of the total contamination density in the 30-cm layer, x is reference number of sample taken in the 5-cm soil layer. Using the coefficient b it becomes possible to calculate the depth at which a decrease in contamination density to a preset level takes place. ^{90}Sr which settled on the soil surface is slowly migrating to deeper layers, and the coefficient b is decreasing (Table 14).

In gray forest soils, the value of coefficient b correlates with the number of years that have passed since the accident ($r = -0.94$, $p = 0.02$). In sod-podzol soils, especially intensive ^{90}Sr migration is going on.

So far, no shift in maximum ^{90}Sr and ^{137}Cs activities down the soil profile has been observed in any soil types of interest (gray forest, sod-podzol, chernozem). The highest specific activity of both ^{90}Sr, and ^{137}Cs in natural and fallow lands is retained in the lower layers of forest litter and or steppe debris. High specific activity of ^{90}Sr and ^{137}Cs is also retained in 0-5 soil layer. In deeper soil layers, the activity of these radionuclides is decreasing rapidly and reaches the minimal values in the 25-40 cm layers of eluvial horizons. In illuvial horizons, radionuclide activity is slightly decreased. In general, the specific activity of ^{90}Sr and^{137}Cs in natural soils, beginning at a depth 20-25 cm is relatively stable. The calculations have shown that the 180-300 cm soil layer contains about 28% of ^{137}Cs and 18% of ^{90}Sr of the total contamination density in soil layer 0-300.

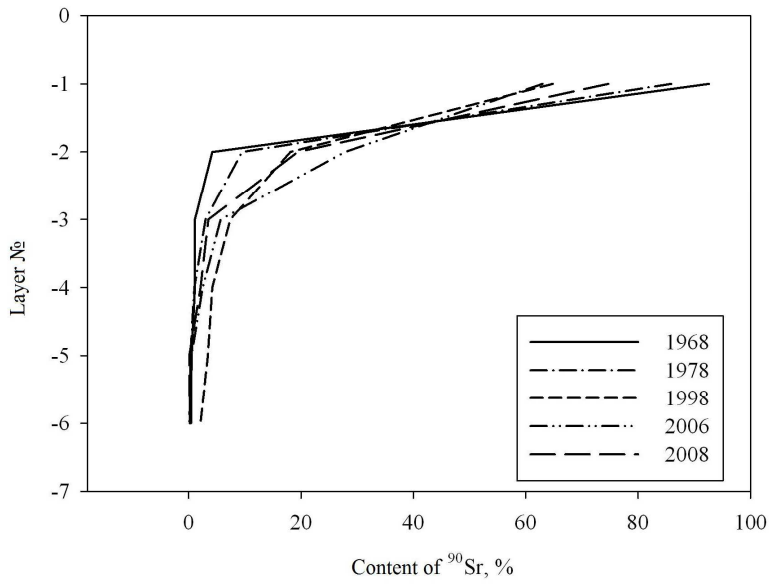

Fig. 15. Dynamics of ^{90}Sr distribution in 30-cm layers of gray forest soil

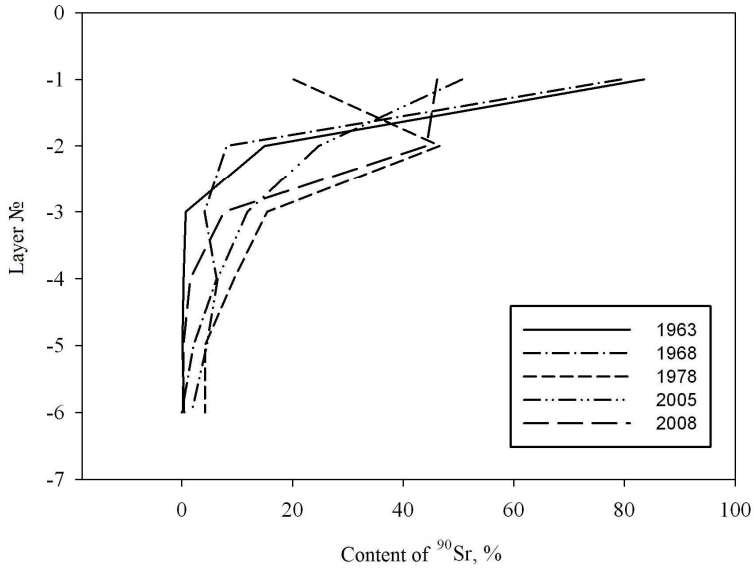

Fig. 16. Dynamics of ^{90}Sr distribution in 30-cm layers of sod-podzol soil

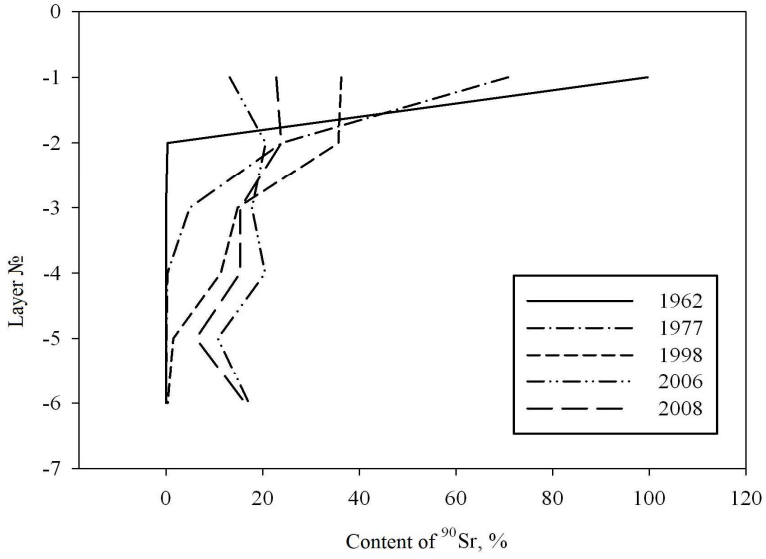

Fig. 17. Dynamics of ^{90}Sr distribution in 30-cm layers of chernozem

Year of sampling	b	R2
Gray forest		
1968	3.07	0.999
1978	2.15	0.999
1998	1.17	0.993
2006	0.96	0.991
2008	1.38	0.999
Sod-podzol		
1963	1.76	0.993
1968	2.17	0.989
1978	0.30	0.435
2005	0.70	0.998
2008	0.62	0.831
Chernozem		
1962	5.78	1.000
1977	1.16	0.997
1998	0.45	0.874
2006	0.01	0.008
2008	0.16	0.555

Table 14. Parameters of equations describing ^{90}Sr distribution in the profile of the 30-cm soil layer

Most of the researchers engaged in studies of radionuclide distribution in soil have noted that in the first years after atmospheric fallouts the highest amounts of ^{90}Sr and ^{137}Cs settled in sod cover or forest litter. It is known, however, that vegetable waste of herbaceous type decays within one season, while leaf wood waste decays within 3-4 years. It takes longer for waste of coniferous forests to decay, however, over the 50 years since formation of the EURT the forest litter contaminated by atmospheric fallouts should have decayed long ago, while the steppe cover of chernozem did not develop until 1991. Thus, the high specific activity currently observed in the forest litter is due not only to the initial fallouts in 1957 and 1967, but also to a high rate of local radionuclide turnover and continuous fallouts of activity.

The reserve of radionuclides in the vegetation mat and upper soil layers is constantly replenished due to vegetation waste. The waste cover is, in its turn, contaminated due to atmospheric fallouts and transfer of radionuclides to the top by the root system. Since actually the total surface biomass of vegetation is transferred to the waste cover at the end of the year, the yearly carry-over of ^{90}Sr to the surface is determined by the annual productivity of the ecosystem.

Waste of woody vegetation accounts for just a portion of the yearly increment, in addition to leaves and grass, the waste composition includes slowly decaying wood, needles and strobiles. That is why radionuclides are deposited not only in soil but also in wood and litter cover. Mineralization of decayed biomass involves an increase in the relative contents of radionuclides in the waste litter. Specific activity of ^{90}Sr in the ground litter was 2.2±0.7 times higher than in grass at a distance of 20 km from the contamination source, and 6.4±36 times higher at a distance of 30 km; the respective values for ^{137}Cs were 14±8 and 12±8. Table

15 presents specific activity values for radionuclides deposited in litter cover and the upper layer of soil. ^{90}Sr activity measured in the upper layer of the litter cover is actually the same as that in the upper layer of the soil, and activity in the lower layer is substantially higher. The difference between levels of ^{137}Cs activity in the upper and lower layers of the litter cover is even more substantial than that found for ^{90}Sr. Ratio of ^{90}Sr specific activity to the activity in the 0-10 cm soil layer at a distance of 20 km from the Mayak PA is 0.7±0.3, and that at 30 km is 2.1±1.0. The ratio for ^{137}Cs is 0.08±0.04 and 0.4±0.4. Since contamination levels of grass are higher at longer distances, it can be assumed that currently the uptake of ^{90}Sr and ^{137}Cs by the biomass is going on through the root system. ^{90}Sr activity in leaves which contribute the largest portion of the vegetation mat is actually the same as that identified in the 0-5 soil layer. However, as was mentioned above, the weight of the vegetation waste per 1 m^2 was not large. On the EURT axis, within 20 km of the Mayak PA, samples of 2008-vegetation waste were taken in a birch forest. Specific activity of ^{90}Sr measured in the waste was 5904 $Bq \cdot kg^{-1}$, that of ^{137}Cs 54 $Bq \cdot kg^{-1}$. The fall of the aboveground phytomass to the forest floor contributed 2.3 $kBq \cdot kg\ m^{-2}$ of ^{90}Sr and 028 $kBq \cdot kg\ m^{-2}$ of ^{137}Cs. This accounted for 2.1% and 3.4% of contamination density in forest litter for ^{137} and ^{90}Sr, respectively, and for 0.36% and 0.8% of the total contamination density in the 0-20 cm layer of forest litter and soil for ^{90}Sr and ^{137}Cs, respectively.

Distance from Mayak PA, km	Soil	Upper layer of litter cover		Lower layer of litter cover		0-5 cm soil layer	
		^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
20	gray forest	7.1×10^3	0.18×10^3	16.3×10^3	1.3×10^3	7.8×10^3	0.48×10^3
	chernozem	0.6×10^3	0.02×10^3	1.2×10^3	0.2×10^3	1.1×10^3	0.3×10^3
30	chernozem	29	5	92	161	31	143
55	Sod-podzol	2.8×10^3	0.06×10^3	10×10^3	1.2×10^3	1.9×10^3	0.2×10^3

Table 15. Soil and litter contamination levels, $Bq \cdot kg^{-1}$

Therefore, the plant litter vegetation waste contributes an insignificant proportion of contamination density of the plant litter and upper layer of soil.

It was found out as a result of measurements of the proportions of radionuclides occurring in different forms and having different degrees of accessibility to plants that ^{90}Sr is mostly encountered in exchangeable form (64-85%), a larger amount of exchangeable strontium being deposited in sod-podsol soils than in gray forest and chernozem. The largest amount of ^{137}Cs is strongly bound together and it occurs in acid-soluble form (4-36%) or as a solid residue (27-82%). In all cases, the content of ^{137}Cs accessible to plants in the 5-10 layer proved to be higher, and the content of ^{137}Cs in the form of solid residue was significantly lower.

Thus, as of today, the studies of all the soil types have shown that the major part of ^{90}Sr and ^{137}Cs activity is deposited in the upper layer.

3.6 Dependencies governing migration of ^{90}Sr and ^{137}Cs in the soil-grass chain and forest products

In 2005-2006, samples of grass and soils taken in the most heavily contaminated areas of the EURT were processed, following which proportionality factors were assessed (PF). Specific activity of ^{90}Sr in grass ranged from 70 to10940 Bq·kg^{-1}, that of ^{137}Cs from 10 to 997 Bq·kg^{-1}. Proportionality factor (Bq/kg in grass/Bq/kg in soil) was within the range 0.2-2.1, the mean value of transfer factor calculated as a ratio of specific activity of radionuclides in grass to soil contamination density, Bq·kg^{-1}/ Bq·kg^{-2}, was 15.9±8.5 for ^{90}Sr, while for ^{137}Cs it was lower: 3.1±3.4. The difference in transfer factor values may depend on soil type and diversity of plant species composing the cover layer. It should be noted that transfer factor values are slightly higher for sod-podzol soils in the EURT zone.

Transfer factor estimated for grass in the EURT zone has changed insignificantly over the recent 10 years, it is 10.3 Bq·kg^{-1} / Bq·kg^{-2} for ^{90}Sr. The mean transfer factor calculated in 1997 for ^{90}Sr in pasture soils in the EURT zone was 13.5 Bq·kg^{-1}/Bq·kg^{-2}. Transfer factor values calculated for ^{90}Sr relative to standard soil contamination of 1 Ci·km^{-2} (37 kBq·m^{-2}) amounted to 230-270 Bq·kg^{-1} / 37 Bq·kg^{-2}.

Contamination with ^{90}Sr of all sampled wild-growing berries exceeds the permissible limit 3.5-13.5 times, ^{90}Sr proportionality factor is higher than that determined for vegetables and grain (Table 16). Specific activity of ^{137}Cs in grass and berries does not exceed the permissible limit. It should be taken into consideration that wild-growing berries in the EURT zone present the highest hazard compared to other food products in view of the contribution they can make to dietary intakes of ^{90}Sr.

Sampling site	Species	Specific activity of fresh berries,Bq·kg^{-1}		proportionality factor, (Bq·kg^{-1} in berries)/ (Bq·kg^{-1} in soil)	
		^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
20 km from Mayak PA	Wild strawberry	813	5.2	0.140	0.0165
	stone bramble	243	2.4	0.042	0.0076
30 km from Mayak PA	Wild strawberry	213	1.0	0.254	0.0054

Table 16. Contamination of wild-growing berries with ^{90}Sr and ^{137}Cs

Table 17 presents data on levels of contamination of fresh mushrooms. In 2008, specific activity of ^{90}Sr and ^{137}Cs in all samples was found to be significantly lower than the permissible limits (50 Bq·kg^{-1} for ^{90}Sr and 500 Bq·kg^{-1} for ^{137}Cs). Proportionality factors and transfer factors were significantly lower than those obtained for grass, vegetables and grains sampled in 2007. It can be assumed that mushrooms growing in the EURT zone present no hazards for the population as their contribution to internal dose is insignificant.

Sampling site	Species	Specific activity of fresh mushrooms, Bq ·kg^{-1}		Accumulation factor, (Bq ·kg^{-1} in the fruit body)/(Bq ·kg^{-1} in soil)		Proportionality factor, (Bq ·kg^{-1} in fruit body) / (Bq ·kg^{-2} in soil)	
		^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
50 km from Mayak PA	Birch mushroom	0.47	0.58	0.0039	0.0113	0.0328	0.0941
	Coral milky cap	1.2	0.97	0.0101	0.0189	0.0839	0.1573
	Mix	0.7	0.9	0.0059	0.0175	0.0489	0.1460
	Milk mushroom yellow	0.34	4.6	0.0041	0.0176	0.0305	0.1447
	Blewits	0.59	19.9	0.0071	0.0762	0.0529	0.6259
	Arachnoid muchroom, excellent	0.63	18.5	0.0076	0.0709	0.0565	0.5819
	Mix	1.4	8.2	0.0169	0.0314	0.1255	0.2579
	Blewits white	0.7	3.9	0.0092	0.0169	0.0749	0.1383
	Mix	1.7	18.8	0.0531	0.0450	0.2798	0.3705
20 km from Mayak PA	Honey agaric summer mushroom	9.8	39.0	0.0017	0.124	0.0133	0.9476
30 km from Mayak PA	Yellow boletus	5.5	10.9	0.0065	0.0586	0.0396	0.4950
	Mix of the lamellar	8.9	2.0	0.0106	0.0108	0.0641	0.0908

Table 17. Contamination of mushrooms with ^{90}Sr and ^{137}Cs

3.7 Migration of radionuclides along food chains

Levels and dynamics of ^{90}Sr specific activity in milk produced in villages at the periphery of the EURT in 1958-2006 are presented in figure 24. A dramatic decrease in contents of ^{90}Sr in milk had occurred before 1963 while later on a slow decrease was observed overtime. Since 1960 the period of a 2-fold decrease in ^{90}Sr contents in milk ($T_{1/2\ eff.}$) was 23 years. The content of ^{137}Cs in milk produced in the villages studied was 2 times lower as compared to that of ^{90}Sr. Currently, this value is1.1 ±0.4 Bq/l for ^{137}Cs.

In 1960, the transfer factor in the soil-milk chain was 340 μBq·l^{-1}/Bq·m^{-2}, in 2006 it was 45 μBq·l^{-1}/Bq·m^{-2}. A reduction in the transfer factor resulted from a decrease in biological accessibility of the radionuclide in the soil-pasture grass chain.

Potatoes take the second position after milk in terms of ^{90}Sr contributed by food produce. The dynamics of reduction in specific activity of ^{90}Sr in potatoes has been the sane over the total period of observations, viz., from 7.0 to 0.9 Bq·kg^{-1}, the respective value for ^{137}Cs is from 1.5 to 0.7 Bq·kg^{-1}. Within a number of years, stable values of transfer factor have been established for agricultural products (Table 18).

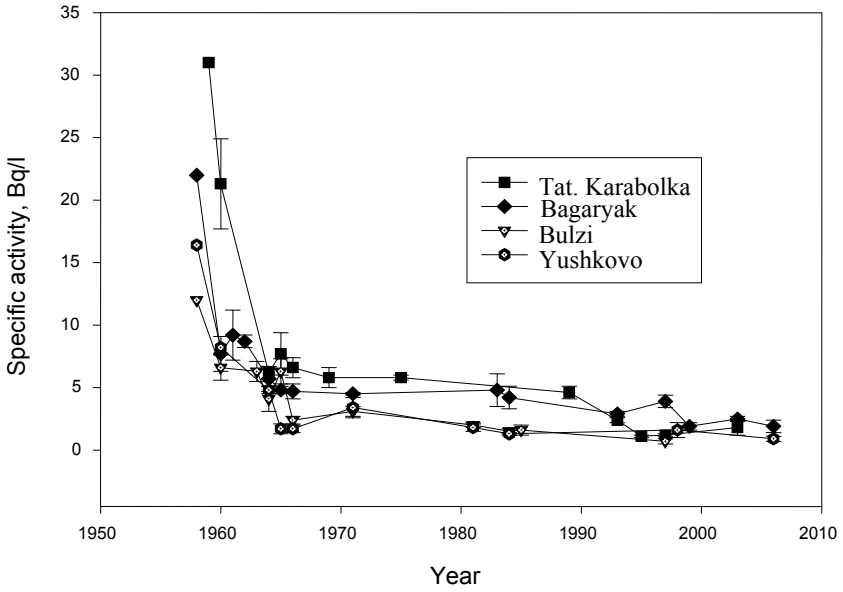

Fig. 24. Dynamics of specific activity of ^{90}Sr in milk produced in EURT villages in the period 1958-2006

Crops	Products	Proportionality factors	
		^{90}Sr	^{137}Cs
Peas	Grain	11.4±6.6	15.2±5.2
Wheat	Grain	14.4±9.1	6.6±1.9
Oats	Grain	5.7±2.5	9.9±4.6
Barley	Grain	6.5±2.1	4.4±1.3
Cabbage	Head of cabbage	4.7±1.6	3.0±1.4
Carrots	Root crop	5.2±1.1	2.2±0.8
Beet	Root crop	43.3±44.7	16.5±15.7
Potato	Tubers	4.8±1.3	2.3±0.5
Corn	Paste (dry)	1.4±0.6	3.6±0.5
Sudanese grass	Paste (dry)	9.0±7.2	46.0±36.7

Table 18. Mean values of proportionality factors for ^{137}Cs and ^{90}Sr measured in the portion of cash crop grown in 0-20 cm layer of gray forest soils

Values of proportionality factor decrease overtime and, as a result, it becomes possible to grow agricultural product with admissible level of conatimation in soils with a higher contaminated level.

4. Karachai Radioactive Trace

In 1997 the area covered by the Karachai Radioactive Trace (KRT) delineated by ^{90}Sr contamination isoline of 7.4 kBq/m^2 (0.2 Ci/km^2) amounted to 1660 km^2. The total amount of the radionuclides deposited on this territory was 800 Ci. Radinuclide composition of dust fallouts was as follows: 32 % of ^{90}Sr+^{90}Y, 47% of ^{137}Cs, 21% of ^{144}Ce+^{144}Pr. Biological accessibility to plants was 90 % for ^{90}Sr, and 12% for ^{137}Cs (Yu.D. Korsakov et al., 1996).

In 1967 about 97% of the total ^{137}Cs deposited in pasture soils settled in the upper 0-3 cm layer. Currently, 38 years after the fallout, ^{137}Cs is accumulated in the soil layer at a depth of 13 cm, and ^{90}Sr is mostly deposited in the soil layer to a depth of 0-20 cm (89.5%). A small portion of radionuclides which settled in the layer at a depth of 0-70 cm migrated to a depth of 70 cm.

In 1967, the transfer factor for contents of ^{90}Sr in grass and soil was 0.09 Bq·kg^{-1} / kBq·m^{-2}. A sharp decline in transfer factor values for ^{137}Cs within the soil-grass chain occurred one year later after it had been cleaned of surface contamination, and in the subsequent years no changes in transfer factor values were noted. During early years, the uptakes of the radionuclide were going on from sod cover and soil, and transfer factor values fluctuated between 0.0025 and 0.005 Bq/kg : Bq/m^2.

Long-term studies of ^{90}Sr and ^{137}Cs transfer from soil to grass were conducted in the grazing land of Sarykulmak village where dairy cattle was grazing (Figure 25).

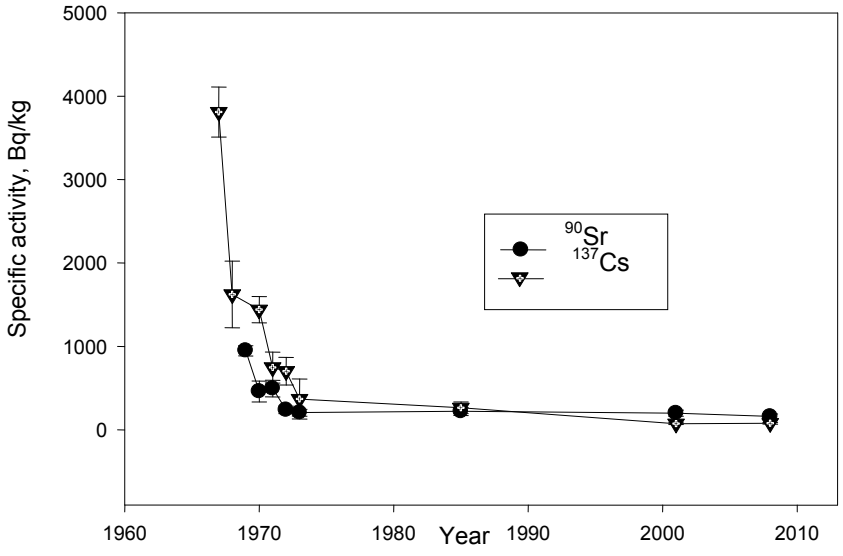

Fig. 25. Levels and dynamics of decrease in radionuclide contents measured in pasture grass

A reduction in specific activity of ^{137}Cs in the pasture grass over the period from 1967 through 2009 is described by the two exponential dependences: $T_{1/2}$ was 1.8 years in the initial period after deposition of radionuclides, and in the subsequent period it was 3.8 years. The first exponent describes the cleaning of grass from surface contamination, the second describes the reduction in uptakes of radionuclides by grass from soil. Over the period from 1967 through 2009, the specific activity of ^{137}Cs in grass decreased 100-fold , that of ^{90}Sr 10-fold.

The initial specific activity of ^{90}Sr in milk late in April, 1967, was 140 Bq/l, that of ^{137}Cs - 237 Bq/l. From 1967 through 2009, the average values of ^{90}Sr and ^{137}Cs specific activity in milk decreased 30-fold. The decrease in ^{90}Sr and ^{137}Cs levels in milk was determined based on the decrease in grass contamination. Beginning from 1970, the period of half-cleaning of milk from ^{90}Sr was 20 years, and from ^{137}Cs 10 years. Proportionality factors for ^{90}Sr in the soil-milk chain estimated on the Karachai Radioactive Trace were on the average 5-fold lower than the values estimated for the EURT for the same time periods elapsed after the radioactive fallouts on the soil. Evidently, the long period for which ^{90}Sr and ^{137}Cs remained deposited in silt and soil on the shores of Karachai led to a reduction in biological accessibility of radionuclides to plants and, as a result, to a reduced uptake of radioactivity by milk. Specific activities of ^{90}Sr in milk are more correctly described by the log-normal distribution. After the accident, the contents of ^{137}Cs in potatoes was insignificant, and overtime it decreased about 10-fold, while the contents of ^{90}Sr decreased 3-fold.

5. Conclusion

1. Major radiation accidents that took place in the Southern Urals during the period 1949-1967 brought about contamination of vast territories with radionuclides and exposure of the local population. The situation on the Techa River involved contamination of river water, bottom sediments and the floodplain with ^{137}Cs and ^{90}Sr. On the East-Urals Radioactive Trace ^{90}Sr was the prevalent contaminant, while on the Karachai Trace biologically poorly-accessible compounds of ^{137}Cs and ^{90}Sr prevailed.
2. The key mechanisms by which contamination of the environment can be eliminated include radioactive decay, reduction in biological accessibility and deepening of radioactive substances in soil. Pronounced sorption capabilities and poor solubility hamper migration of ^{137}Cs in the environment, while ^{90}Sr is more mobile.
3. Due to a number of protection measures implemented from 1965 through 2004 on the Techa River, concentrations of ^{90}Sr and ^{137}Cs in river water have decreased, and currently the specific activity of ^{90}Sr amounts to 10-15 Bq $\cdot l^{-1}$, and that of ^{137}Cs to 0.5-1.5 Bq $\cdot l^{-1}$. Radionuclides deposited in bottom sediments migrated to the depth of over 35 cm. Contamination density in the 0-10 cm layer depends on the concentration of radionuclides in the watercourse. ^{90}Sr and ^{137}Cs are revealed in floodplain soil at a depth of over 70 cm where a decrease and averaging of concentrations overtime has been observed. At the present time, it is expedient to retain the constraints on the use of river water.

   After the use of river water was banned in 1956, the major pathway for contribution of radioactivity to dietary intakes has been made by milk (87-95 %) and vegetables. Since 1967, the content of ^{90}Sr in milk, with rare exception, has not exceeded the permissible limit (25 Bq $\cdot l^{-1}$).

4. The rate of vertical migration of ^{90}Sr in EURT soils ranges from 0.25 to 0.35 cm·year^{-1}, the largest amount of radionuclides which remains deposited in the upper part of the soil profile (0-20 cm) is decreasing with increasing depth. Nonmobile ^{137}Cs is mosty retained in the upper 10-cm layer. Biological accessibility of ^{90}Sr has decreased over the past period 7-10 times and it has not actually changed over the recent years. Currently, 40 years after the accident, the content of fixed forms of ^{90}Sr in soil has reached 34%, that of ^{137}Cs and plutonium 95-98%.
   The period of half-cleaning of milk from ^{90}Sr was 2-3 years during the first years after the accident, and during the subsequent period it amounted to 15 years.
5. On the Karachai Trace, due to the prevalence of ^{137}Cs and ^{90}Sr with poor biological accessibility, the reduction in contamination of soils, grass and food products was going on more speedily (than that registered on the EURT). Specific activity of ^{137}Cs and ^{90}Sr in milk exceeded the permissible limits only during the first month after the accident.
6. The prognosis for the further development of the radiation situation on the Techa River is determined predominantly by the radioactive runoff from the Techa cascade of reservoirs. With the lapse of time, the part of the EURT territory where the use of agricultural lands is restricted, will diminish. There are no such restrictions in the Karachai Trace area.

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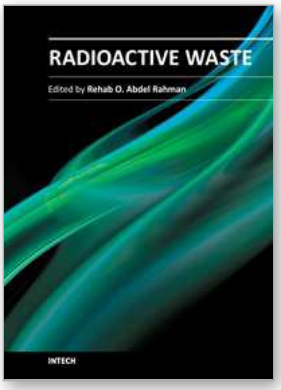

Radioactive Waste

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The safe management of nuclear and radioactive wastes is a subject that has recently received considerable recognition due to the huge volume of accumulative wastes and the increased public awareness of the hazards of these wastes. This book aims to cover the practice and research efforts that are currently conducted to deal with the technical difficulties in different radioactive waste management activities and to introduce to the non-technical factors that can affect the management practice. The collective contribution of esteem international experts has covered the science and technology of different management activities. The authors have introduced to the management system, illustrate how old management practices and radioactive accident can affect the environment and summarize the knowledge gained from current management practice and results of research efforts for using some innovative technologies in both pre-disposal and disposal activities.

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