^{90}Sr and ^{137}Cs in flood-plain soils of the Techa river

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Abstract. The Techa river was contaminated in 1949-1956 from the nuclear enterprise "MAYAK". The investigations were carried out in flood plain of the Techa river in 1992-1999. In the upper and middle parts of the river the basic contaminator is ^{137}Cs; downstream - more active is ^{90}Sr. Density of contamination of soils with ^{90}Sr down the river changes from 600 to 200 kBq/m^2, and that with ^{137}Cs - from 2,000 to 80 kBq/m^2. Radionuclides content in soils of the central part of the flood plain (30-40 m from the river-bed) also decreases further from the discharge point 1-2 orders. The Techa flood plain may be considered to be a source of secondary radioactive contamination of the water ecosystem. We evaluated the absorption stability and water migration of radionuclides in its soils under experimental conditions. The main quantity of ^{90}Sr was in exchangeable (41-66 %) form. ^{137}Cs was in firmly fixed form (80-98 %). The study of radionuclides distribution in the liquid fraction of the soil showed that the main part of ^{90}Sr (84%) was found in gravitation moisture. ^{137}Cs (97%) was in a less mobile form of moisture which was connected with soil by capillary and sorption powers.

1. INTRODUCTION

The Techa river 243 km long originates in the lake Irtyash and flows on the territory of the nuclear enterprise "MAYAK" in its upper stream. Due to imperfect technologies of production and storage of radionuclides in 1949-1952 radioactive waste entered the open drainage network 6 km from the source of the Techa. Totally $76 \cdot 10^6$ m^3 of the waste water entered the river. The total discharge of radionuclides into the river estimated as $100 \cdot 10^{15}$ Bq. Long-lived ^{90}Sr and ^{137}Cs comprised 25 % of the total discharged radioactivity. The construction of the series of reservoirs in the upper part of the river has significantly restricted the entry of radionuclides into the river system, but bottom deposits, biota and the soil-vegetation cover of the flood plain were strongly contaminated over the whole length of the river. Such contamination of natural ecosystems has never been reported in the world, neither for the quantity nor for the isotope composition of the discharge [2,3].

Flood plain landscapes have become a source of secondary contamination of the river. The flood plain form and hydromorphic conditions of soil formation make landscape geochemical barriers where many macroelements, heavy metals and radionuclides are accumulated. An excessive moistening of the flood plain and anaerobic conditions of organic matter decomposition increase the transport of the pollutants from soils into the river with the surface and ground runoff. During the research in 1992-1999 we evaluated the peculiarities of the spatial distribution of ^{90}Sr and ^{137}Cs in soils of the Techa flood plain, radionuclides storage and the contribution of the liquid runoff to the secondary contamination of the river.

2. RADIONUCLIDES IN THE FLOOD PLAIN SOILS

2.1. Materials and methods

Soil profile cuts were studied on the flood plain plots on the right and left banks of the Techa in its middle and lower parts. The plots were located at various distance from the pollution source, the control soil profile cuts being outside the limits of its effect. The Techa flood plain 100-500 m wide, is rugged hilly in

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some places has many old river beds, lakes and hollows. Numerous bushes grow on the banks of the river. The morphological characteristics and chemical composition of the soils show periodicity of alluvial deposition with the flood water. The content of humus in them gradually decreases from 6.0 % in upper 0-5 cm layer to 1.4 % at the depth of 25-30 cm, pH of the water extract is about 6.5, pH of the salt extract is 5.8. The sum of exchangeable bases changes in the soil profile from 26.2 to 18.2 mg-eq/ 100 g air-dry mass in the upper and in the lower layers respectively. Samples (5 cm layers) were taken from soil profile with regard to the area. The maximum depth of sampling did not exceed 40 cm and, as a rule, corresponded to the level of the ground water. The above- ground mass of herbaceous plants removed in the immediate vicinity of the soil profile.

To estimate the strength of fixation of radionuclides in soil samples various physic-chemical forms were distinguished. The water-soluble form was obtained by distilled water, the ratio between the solid and liquid phases was 1:5. The exchangeable form was obtained using 1 N CH_3COONH_4 , the acid-soluble form - with 1N HCl (the ratio 1:10). The radionuclides which remained in the soil after this treatment were considered to be a fixed form. The ^{90}Sr content of the samples was estimated by radiochemical method by its daughter ^{90}Y. The concentrations of ^{137}Cs were measured by means of a multichannel gammacounter using a germanium detector. Sensitivity of the method for ^{90}Sr determination was 1 Bq, the minimum measured activity of ^{137}Cs was 3 Bq.

2.2. Distribution of radionuclides in the soil-vegetation cover of the Techa flood plain

The uneven character of the spatial contamination of the investigated area did not allow to reveal a distinct relationship the radionuclides content from their proximity to the river banks, therefore, table 1 gives the data only for the right river bank. Maximum ^{90}Sr and ^{137}Cs concentrations in the soil profiles near the river-bed were reported for the Brodokalmak village. On the other plots the quantity of radionuclides in the 0-10 cm layers varied from 195 to 1,500 Bq/kg depending on the remoteness of sampling from the pollution source. Concentrations of ^{90}Sr and ^{137}Cs were correspondingly 100 and 20 times more than in the control, even in the most remote plots.

The soil-vegetation cover of the periodically flooded central part of the flood plain (30-40 m from the bed) had also a higher concentration of radionuclides compared to the control. Comparison of the data shows that in some cases accumulation of ^{90}Sr and ^{137}Cs in soils of the central flood plain was higher because during the high water of the Techa the dispersed material rich with radionuclides is deposited in the stagnant water. Maximum ^{90}Sr concentration in the above ground mass of herbaceous plants comprised 2,000 Bq/kg air-dry mass (Bugaevo village), which 200 times exceeds the control. ^{137}Cs content in plants was ca. 10 Bq/kg on the control plot and no more that 40 Bq/kg air-dry mass in the Techa flood plain. Differences in ^{90}Sr and ^{137}Cs accumulation in the herbaceous plants while in soil their contents were similar might be connected with predomination of ^{90}Sr mobile forms in hydromorphic soils and their more active accumulation in the vegetation cover. In the flood plain soils 1.0-2.6 % ^{90}Sr were in the water-soluble form, 41-66 % in the exchangeable form, 30-52 % in the acid-soluble form. ^{137}Cs accumulated by soils much more firmly, the content of its water-soluble forms comprised 0.02-0.5 %, that of its exchangeable forms - 1-16 % and 80-96 % was in the fixed form. Enrichment of the lower soil layers with mobile radionuclides forms was observed in the process of vertical migration. Corresponding to various fixation of radionuclides by soils ^{90}Sr accumulation coefficients (C_{ac} - ratio between radionuclide concentration in plant and its average concentration in soil)) were 25-300 times higher in herbaceous plants than ^{137}Cs accumulation coefficients (0.5-27 for ^{90}Sr and 0.02-0.07 for ^{137}Cs depending on the plant species peculiarities).

The vertical distribution of radionuclides in soils of the pre-bed and central flood plain was similar (table 2). The distribution in depth was even. Migration ability of ^{90}Sr and ^{137}Cs leveled off in the flood plain as a results of changes in their physic-chemical state and movement in the water-soluble compounds and fine dispersed particles with the surface and ground runoff.

Table 1. Radionuclides in the soil-vegetation cover in the pre-bed (1) and central (2) flood plain, Bq/kg air-dry mass

Location	Layers, cm	^{90}Sr		^{137}Cs	
		1	2	1	2
v. Brodokalmak	Plants	-	64	-	30
	0-10	1,500	60	7,900	2,200
	10-20	1,300	40	7,300	1,300
	20-30	1,500	38	3,000	140
	30-40	1,650	18	5,600	40
v. Anchogova	Plants	1,230		39	
	0-10	932	145	1,200	157
	10-20	608	195	670	127
	20-30	562	117	340	153
v. Bugaevo	Plants	2,016	2,090	10	10
	0-10	195	1,470	275	1,320
	10-20	266	2,080	170	1,250
	20-30	388	1,270	370	1,310
v. Pershino	Plants	745	375	22	10
	0-10	280	250	242	585
	10-20	75	80	39	30
	20-30	34	18	21	5
v. Zatechenskoe	Plants	40	97	40	40
	0-10	1,500	190	775	490
	10-20	277	55	60	54
	20-30	212	13	13	43
Control	Plants	10	39	10	10
	0-10	16	32	30	60
	10-20	8	10	22	10
	20-30	4	5	10	10

Table 2. The vertical distribution of radionuclides in the flood plain soils, % of the total content in soil profile (1 – pre-bed, 2 – central parts)

Location	Layers, cm	^{90}Sr		^{137}Cs	
		1	2	1	2
v. Brodokalmak	0-10	38.0	26.1	33.8	47.1
	10-20	34.8	34.2	51.2	46.7
	20-30	27.2	39.7	15.0	6.2
v. Bugaevo	0-10	28.7	21.1	38.4	24.4
	10-20	33.7	46.2	25.9	35.6
	20-30	37.6	32.7	35.7	40.0
v. Pershino	0-10	43.2	48.8	48.0	68.8
	10-20	29.7	33.3	30.9	17.1
	20-30	27.1	17.9	21.1	14.1
v. Zatechenskoe	0-10	77.0	72.5	72.4	81.0
	10-20	11.7	24.2	26.4	10.3
	20-30	11.3	3.3	1.2	8.7

2.3. Estimation of the storage of radionuclides in the Techa flood plain

It is seen from table 3, which gives the data for the whole research period, that the storage of radionuclides in soils decreased farther away from the pollution source. This reduction is well described by the function $y=e^{a}\ x^{b}$, where **y** is contamination density, kBq/m^2 , and **x** is the distance from the MAYAK enterprise, km. Coefficients **a** and **b** for various elements were obtained empirically [1]. In the pre-bed flood plain the values $^{90}Sr/^{137}Cs$ increase with the distance from the discharge point, and downstream ^{90}Sr is the main pollutant. It is also the main pollutant of the central flood plain (as a rule

$^{90}Sr/^{137}Cs > 1$). The pre-bed landscape sites enriched with ^{137}Cs can be considered to be natural barriers which restricted its spatial distribution.

Table 3. The storage of radionuclides in soils of the pre-bed (1) and central (2) flood plain, kBq/ m^2

Location	Distance from the MAYAK	^{90}Sr		^{137}Cs		$^{90}Sr/^{137}Cs$	
		1	2	1	2	1	2
v. Brodokalmak	107	625	1,272	2,886	467	0.2	2.7
v. Lobanovo	158	85	-	184	-	0.5	-
v. Anchogovo	169	161	337	190	2,000	0.8	0.2
v. Bugaevo	189	158	928	128	440	1.2	2.1
v. Pershino	214	204	375	124	125	1.6	3.0
v. Zatechenskoe	237	191	25	84	28	2.3	0.9
Control	Outside of the influence	3.8	4.2	8.9	5.0	0.3	0.7

According to calculations [1] based on assumptions and direct measurements of radionuclides concentrations, ^{90}Sr storage in the investigated soils accounted for $75 \cdot 10^{12}$Bq, that of ^{137}Cs – $198 \cdot 10^{12}$ Bq.

3. MIGRATION OF RADIONUCLIDES FROM SOILS BY WATER

In conditions of flooding mobile radionuclide compounds can transfer from soil to the liquid form and enter the river system again in the liquid runoff. We experimentally estimated the secondary contamination of the river with radionuclides entering it with the liquid runoff from the flood plain soils.

3.1. Material and methods

An upper 10-cm layer of soils from the pre-bed flood plain (Brodokalmak-Anchogovo region) was used in the experiments. Soil samples (3 kg) were put into special containers (d=18 cm, h=30 cm) which had a drain in the column bottoms. During the first 3 months the soil was found in conditions of damping, in every column the water layer was 5 cm above the soil surface (we controlled ^{90}Sr and ^{137}Cs concentrations in this water layer during the whole period of the experiment). To imitate washing water regime every 2 weeks we opened the clips in the column bottoms and collected the filtrates, then restored the primary levels of water in the containers. Then the soil was kept in conditions of variable moistening for 3 months, alternating drying and moistening up to 60% from the maximum water holding capacity. The soil kept in such conditions was again flooded as in the beginning of the experiment.

During the experiment the water on the surface of the flooded soil (above-surface water) and the filtrate were sampled. In the end of the experiment, the last filtrate portion having drained off, the water kept by capillary-sorption forces was extracted from the soil on a centrifuge. It was assumed that the excessive above soil water imitated the surface runoff and the filtrate – the ground runoff.

3.2. Results

The results of the experiments have shown that differences in the strength of ^{90}Sr and ^{137}Cs fixation in soils are reflected in indices characterizing their return into the liquid phase. The entry of radionuclides into successive portions of the filtrate decreased in different measure. Thus, 1.3% of ^{90}Sr and 0.002% ^{137}Cs left the soil with the first filtrate portion. After the 6-fold change of water the quantity decreased 2 and 4 times correspondingly. Alternating moistening and drying of the soil resulted in the recovery of the pool of mobile radionuclide compounds up to the original level.

The research of radionuclides distribution in the soil liquid phase showed that the main quantity of ^{90}Sr was removed with freely filtered water, less – with the water bonded by capillary-sorption forces and still less with the water above the surface of the flooded soil (Table 4). ^{137}Cs was basically kept in the

water bonded by capillary-sorption forces. Similar data for ^{137}Cs had been obtained earlier, in the experiments with other soils [4]. The analysis of the data allows to conclude that movement with mobile forms of moisture more characteristic for ^{90}Sr than for ^{137}Cs.

Table 4. Distribution of radionuclides in the soil liquid phase, %

Forms of moisture	Volume	^{90}Sr	^{137}Cs
Gravitation (the water above the surface of the flooded soil)	52 (40)	1.46 (0.16)	0.006 (0.004)
Capillary-sorption	48	0.28	0.184

The obtained characteristics of the transfer of radionuclides to various forms of the soil moisture allowed to estimate the possible contribution of the liquid runoff to the secondary contamination of the river. Calculations showed that ca. 0.16% of ^{90}Sr and 0.004 % of ^{137}Cs from their storage in the flood plain soils could enter the river with the surface runoff. Up to 1.46% of ^{90}Sr and 0.006 % of ^{137}Cs could move in the soil profile with the ground runoff and partially enter the river. Besides, about 0.28 % of ^{90}Sr and 0.18% of ^{137}Cs could be in the form of soil water bonded by capillary-sorption forces.

Thus , although ^{137}Cs storage in the flood plain soils ca. 2.5 times exceeded that of ^{90}Sr, the contribution of the last to the secondary contamination of the river ca. 100 times exceeded that of ^{137}Cs. Changes of moistening regimes, seasonal drying and over moistening of soils could result in higher estimates.

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