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Mobility of Technogenic Radionuclides in the Soil–Plant System

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Global radioactive contamination of the biosphere took place in the mid-20th century as a result of large-scale nuclear weapons tests. Radiochemical and nuclear power industries in normal operation are also major sources of technogenic radionuclides, to say nothing about radiation accidents. With respect to ecological consequences, accidents at the Mayak Radiochemical Plant (today, PO Mayak) in the Southern Urals in 1957 and at the Chernobyl Nuclear Power Plant (NPP) in 1986 are considered most serious. One of these consequences is the establishment of corresponding alienation zones, the Eastern Ural State Reserve (EUSR) and the 30-km zone around the Chernobyl NPP.

In previous studies, we estimated spatiotemporal characteristics of distribution, direction, and intensity of long-liver radionuclide migration flows in terrestrial ecosystems (Aarkrog et al., 1992, 1997; Karavaeva et al., 1994; Molchanova et al., 1998; Chyna et al., 2000; Molchanova and Karavaeva, 2001). Their results show that the physicochemical state of radionuclides and, therefore, the strength of their fixation in soils depend on conditions of radioactive contamination, chemical properties of emitters, the time of their presence in natural media, and the properties of these media (Mikhailovskaya et al., 2004). The rate of radionuclide input into plants through the roots depends in a number of physicochemical and ecological factors which are difficult to control under natural conditions.

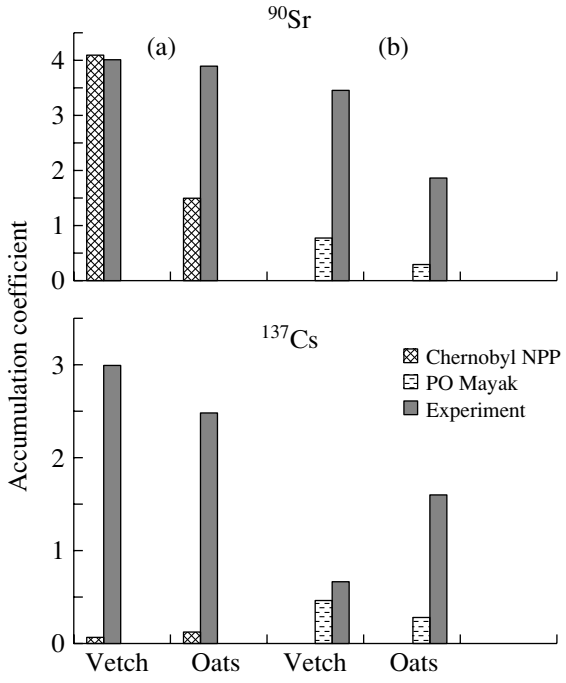
The purpose of this study was to analyze the mobility of different technogenic radionuclides in a model soil–plant system depending on the time of their presence and physicochemical state in the soil.

Two series of greenhouse experiments were perfumed. In the first series, we used samples of soddy podzolic soil (horizon A2) collected in the Chernobyl zone, 3 km west of the damaged reactor unit, two years after the accident. The second series was performed with samples of gray forest soil (horizon A) collected in the EUSR, 12 km northwest of PO Mayak, 47 years after the accident. These soils differed in physicochemical parameters (Table 1). The podzolic A2 horizon had an acid pHH_2O, and the contents of humus, exchangeable bases, and clay fraction in it were an order of magnitude lower than in the A horizon of gray forest soil. Radionuclide concentrations reached 414.0 kBq/kg ^{90}Sr and 1112.0 kBq/kg ^{137}Cs in the Chernobyl soil and 77.7 kBq/kg ^{90}Sr and 1.8 kBq/kg ^{137}Cs in the EUSR soil.

In each series, we performed one experiment with ^{90}Sr or ^{137}Cs chlorides added to the final concentrations of 100 or 200 kBq/kg, respectively. For this purpose,

Table 1. Physicochemical characteristics of soils studied

Sampling site	Soil, horizon	pH_{H_2O}	Carbon according to Tyurin, %	Exchangeable $Ca^{2+} + Mg^{2+}$, mg-equiv/100 g	Particle-size composition		
					1.0–0.01 mm	0.01–0.001 mm	<0.001 mm
30-km Chernobyl zone	Soddy podzolic soil, A2	4.7	0.4	4.7	76.9	19.8	3.3
EUSR	Gray forest soil, A	7.1	7.0	67.3	41.2	18.1	40.7
Background sites	Soddy podzolic soil, A2	5.3	0.14	4.7	63.9	22.8	13.3
	Gray forest soil, A	6.8	13.1	52.0	56.9	19.0	24.1



1 Radionuclide accumulation by plants from (a) soddy podzolic and (b) gray forest soils.

soil types with physicochemical properties corresponding to those of the test soils were selected in control areas with the background radionuclide contents (Table 1). After adding radionuclides, the soil was thoroughly mixed and dried to an air-dry state. In all experimental variants, the soil was placed in 0.5-l polyethylene vessels and planted with germinating seeds of vetch or oats, ten seeds per vessel. Soil moisture in the vessels was maintained at 50% full water capacity. The experiment was performed in three to five replications. Plants were removed from the vessels after 30 days (oat plants were at the phase of the fourth true leaf, and vetch plants, of the fifth true leaf). Their above-ground parts were weighed, dried, and ashed at 450–500°C.

In soil samples contaminated after radiation accidents, we determined the contents of water-soluble, exchangeable, and acid-soluble forms of radionuclides using sequential extraction with distilled water, 1 N CH_3COONH_4, and 1 N NCl. Radionuclides remaining in the soil after this procedure were classified as fixed forms, and acid-soluble radionuclides were also included in this category. Water-soluble and exchangeable radionuclides were classified as movable.

The contents of ^{90}Sr in the samples were determined using a Progress computer-aided spectrometer with the Прогресс-2000 software and also radiochemically, by daughter ^{90}Y, with radiometry in a UMF-2000 instrument. The contents of ^{137}Cs was determined in multichannel gamma-analyzers with a NaI(Tl) scintillation detector (for the samples from Chernobyl) or a Ge semiconductor detector. The statistical error of measurements was no more than 15%.

To estimate the ability of plants to accumulate radionuclides, we used the accumulation coefficient (AC) accepted in radioecological studies, which is calculated as the ratio of radionuclide concentration in the plant to that in the soil. The standard error of estimate in AC calculations did not exceed 15%. The results (figure) showed that experimental plants in all variants accumulated ^{90}Sr more actively than ^{137}Cs. In only one variant (^{90}Sr, soddy podzolic soil, vetch), AC values for water- 1
soluble radionuclides added to the soil were higher than those for radionuclides in soils contaminated after radiation accidents (by factors of 3.0–6.0 for ^{90}Sr and 20050 for ^{137}Cs).

A comparative analysis of radionuclide accumulation in vetch and oat plants showed that they took up ^{90}Sr more actively from the Chernobyl soil than from the soil contaminated after the accident at PO Mayak. The values of AC for ^{137}Cs did not exceed 0.2 in the former case and reached 0.5 in the latter case. As follows from the figure, water-soluble radionuclides added to the soils simultaneously were accumulated by plants more actively from the soddy podzolic soil than 1
from the gray forest soil. This is explained by differences in the contents of exchangeable Ca^{2+} and Mg^{2+} in the soil absorbing complex as well as in the contents of organic matter and the fraction of particles less than 0.001 mm in size. Correlation analysis confirmed a significant inverse correlation between these parameters and AC values for ^{90}Sr and ^{137}Cs ($r = -0.95$; $t_{exp} = 5.3$, $t_{St} = 3.2$). As a rule, the capacity for accumulating ^{90}Sr was higher in vetch than in oats, with this difference being less apparent in the case of ^{137}Cs. In general, differences in the mobility of radionuclides in the soil–plant system are reflected in the following ascending series: for ^{90}Sr, accidental discharge from PO Mayak < accidental discharge from Chernobyl NPP < experimental contamination; for ^{137}Cs, accidental discharge from Chernobyl NPP < accidental discharge from PO Mayak < experimental contamination.

Table 2 shows the results of study on the physicochemical state of radionuclides in soils from the zones of radiation accidents. It is noteworthy that these soils proved to differed significantly in the contents of acid-soluble ^{90}Sr and ^{137}Cs and of fixed ^{90}Sr, with the relative contents of movable (water-soluble and exchangeable ^{90}Sr и ^{137}Cs being 37–50 % и 1.7 %, respectively. Total contents of ^{90}Sr и ^{137}Cs in the soils were used to calculate absolute amounts of their movable forms (Table 3), since they largely characterize the pool of radionuclides accessible to plants (Shcheglov, 1999; Mamikhin, 2004). On this basis, we then calculated the intake coefficients (IC) of radionuclides as the ratios of their concentrations in plants to the total contents of their movable forms in the soils.

The results showed that IC values were not proportional to the contents of movable radionuclides in the

Table 2. Contents of different physicochemical forms of ^{90}Sr (above the line) and ^{137}Cs (below the line) in soils from nuclear accident zones, %

Radionuclide source	Soil, horizon	Physicochemical form			
		water-soluble	exchangeable	acid-soluble	fixed
1 Chernobyl NPP	Soddy podzolic soil, A2	$\frac{0.3 \pm 0.1}{0.1 \pm 0.02}$	$\frac{36.9 \pm 4.7}{1.5 \pm 0.2}$	$\frac{7.7 \pm 0.9}{11.3 \pm 1.8}$	$\frac{55.1 \pm 6.0}{87.1 \pm 7.0}$
PO Mayak	Gray forest soil, A	$\frac{0.5 \pm 0.01}{0.1 \pm 0.01}$	$\frac{50.1 \pm 0.4}{1.6 \pm 0.7}$	$\frac{44.5 \pm 0.7}{2.3 \pm 0.5}$	$\frac{4.9 \pm 0.5}{96.0 \pm 8.0}$

Table 3. Coefficients of radionuclide input (IC) from soils into plants

Radionuclide source	^{90}Sr			^{137}Cs		
	Movable forms in soil, kBq/kg	IC		Movable forms in soil, kBq/kg	IC	
		vetch	oats		vetch	oats
Chernobyl NPP	154.1	11.0 ± 1.10	4.0 ± 0.70	17.8	3.8 ± 0.6	8.3 ± 0.3
PO Mayak	39.3	1.6 ± 0.10	0.6 ± 0.02	0.03	27.0 ± 1.7	18.0 ± 2.3

soil. Their intake by plants apparently depends on a number of factors that were not controlled in this study, such as the presence in soil solutions of competing cations, stable nonisotope carriers, and organic matter or changes in their pH. Moreover, plants can take up radionuclides not only from the soil solution but also through direct contact of the root system with soil particles.

Thus, the mobility of technogenic radionuclides in the soil–plant system depends on their physicochemical properties and the time of contact with the soil. Soil properties, including physicochemical features, have a significant effect on the accumulation of radionuclides by plants. Their absorption through the roots is a complex multifactor process. Therefore, in assessing the mobility of radionuclides in the soil–plant system, it is necessary to take into account their physicochemical state in the soil and specific features of transfer to the soil solution.

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SPELL: 1. soddy