

Distribution and mobility of ^{137}Cs , ^{90}Sr , $^{239,240}\text{Pu}$ and ^{241}Am in solid phase-interstitial soil solution system

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Abstract. Analysis of the migratory properties of ^{137}Cs , ^{90}Sr , $^{239,240}\text{Pu}$ and ^{241}Am in different kinds of mineral and organic soils has been carried out. The radionuclide distribution coefficient between solid and interstitial water phases of soil was used as a parameter of radionuclide mobility. Redistribution of radionuclides in the soil profile, soil-plant, soil-environmental water systems to a great extent depends on this parameter. Determinations of ^{137}Cs , ^{90}Sr , $^{239,240}\text{Pu}$ and ^{241}Am in soils, interstitial soil solutions and estimation of distribution coefficients have been realized. The soil samples were taken in areas of Gomel, Mogilev and Brest regions of Belarus that were contaminated with radionuclides of Chernobyl origin. The influence of exchangeable radionuclide forms and moisture capacity of soil and such characteristics of the water phase as concentration of chemical analogs of radionuclides, organic matter and other components on distribution of ^{137}Cs , ^{90}Sr , $^{239,240}\text{Pu}$ and ^{241}Am in solid phase-interstitial water system has been analyzed.

1. INTRODUCTION

The mobility of radionuclides in the soils of the contaminated areas is one of the main factors determining the radiocological status of the environmental system as a whole. The interstitial (pore) soil solutions play an important role in the processes of radionuclide mass transfer in the soil profile and within soil-plant, soil-surface water and soil-ground water systems. The interstitial soil solution represents a specific kind of natural waters that is retained by surface interaction with soil particles. These waters differ from rapidly inflowing gravitational waters and are in contact with the solid phase of soil for relatively long periods of time. As a result the most mobile forms of chemical elements, including radionuclides, are found in the pore waters.

On the one hand, the interstitial soil solutions take part in biological migration of radionuclides. They define the accumulation of nutrients and also radionuclides by plants through their root system. On the other hand, they represent open thermodynamic systems, genetically bound with other kinds of natural waters, and play an important role in processes of geochemical migration of radionuclides.

Specific activities of soils and interstitial soil solutions, their chemical composition, physicochemical characteristics and distribution of ^{137}Cs , ^{90}Sr , $^{239,240}\text{Pu}$ and ^{241}Am between solid and water phases of different soil types have been determined. The distribution interphase coefficients of radionuclides for water saturated soils have been estimated. The data provide the means to classify the different soils according to the migratory ability of radionuclides.

2. OBJECTS AND METHODS OF INVESTIGATION

The points of observation are situated at the distances of 10 - 50 and 160 - 260 km from Chernobyl Nuclear Power Plant (CNPP). The radioactive contamination and soil cover (Table 1) is typical for the affected regions of Belarus. The upper (0-5)-cm soil layers of soddy-podzolic, peat and alluvial soddy soils from undisturbed lands were studied.

Table 1. Location of sampling sites, agrochemical characteristics and level of radioactive contamination of dry samples

Sample	Location of sampling site, distance from CNPP (km)	Soil type	Sampling date	kBq/kg			A ²⁴¹ Am	WC, %	OM, %	pH (pCC)	meq/kg			CEC
				A ¹³⁷ Cs	A ⁸⁹ Sr	A ^{239,240} Pu					Ca _{ex}	Mg _{ex}	K _{ex}	
SP-1	Masany, Gornel region, 10	Soddy-podzolic sandy loam	22.07.1997	74.9±4.4	27.1±2.2	0.410±0.021	0.504±0.025	33.5	2.9	5.5	24.0	5.04	2.92	57.7
P-1	Radin, Gornel region, 20	Peat gley	13.05.1995	427±24	17.7±1.4	0.214±0.011	0.257±0.013	171	41	4.4	246	41.1	17.7	676
SP-2	Borshovka, Gornel region, 17	Soddy-podzolic sand	22.07.1997	15.9±1.0	0.85±0.08	0.037±0.003	0.042±0.004	29.7	0.9	4.4	1.3	<0.3	0.4	51.5
S-2	Dovlyady, Gornel region, 20	Alluvial soddy sandy loam	13.05.1995	40.0±1.7	3.35±0.30	0.124±0.006	0.148±0.007	83.0	7.1	4.9	47.6	13.6	6.08	121
P-3	Pogonnoye, Gornel region, 30	Peat gley	17.09.1997	129±8	31.0±2.5	0.273±0.014	0.367±0.018	221	39	4.8	289	41.2	3.19	925
P-4	Novoselky, Gornel region, 46	Mineralized peat	25.10.1994	8.84±0.51	0.16±0.02	0.019±0.002	0.020±0.002	61.1	14.4	6.3	12.8	20.5	4.91	240
SP-5	Krasnoselye, Gornel region, 20	Soddy-podzolic sand	13.05.1995	46.2±2.4	0.89±0.09	0.099±0.005	0.116±0.006	25.7	1.3	3.9	1.3	<0.3	0.6	26.4
SP-6	Novoselky, Gornel region, 48	Soddy-podzolic sandy loam	13.05.1995	8.81±0.37	1.78±0.06	0.028±0.003	0.029±0.003	59.2	7.5	5.0	53.5	19.2	8.05	169
P-6	Besed, Gornel region, 158	Peat-boggy	17.10.1997	7.03±0.43	1.04±0.15	0.032±0.006	0.044±0.009	553	94.8	2.8	87.1	29.0	2.56	779
SP-7	Olimany, Brest region, 218	Soddy-podzolic sand	18.06.1996	1.01±0.05	NA	NA	NA	39.8	2.8	4.4	8.98	3.88	0.92	44.6
P-7	Olimany, Brest region, 218	Peat gley	18.06.1996	3.95±0.38	NA	NA	NA	221	62.4	6.6	257	99.9	4.11	912
P-11	Veprin, Mogilev region, 260	Alluvial peat gley	22.07.1997	78.5±4.5	NA	NA	NA	355	80.9	6.9	1220	106	12.4	2040

WC — total water capacity (maximum water retained by 100 g of soil dried at 378 K); % of dry soil mass; OM — organic matter content; Ca_{ex}, Mg_{ex}, K_{ex} — content of exchangeable calcium and magnesium and mobile potassium in soil; N.A. — radionuclide was not analyzed.

The interstitial solutions were obtained from the saturated soils using high-speed centrifuge SIGMA-4-10. Exchangeable forms of radionuclides (cationic and water-soluble complex) were extracted from soil with 1 M $\text{NH}_4\text{OOCCH}_3$. The content of ^{137}Cs in the soil and soil solution samples was measured by γ -spectrometer ADCAM-300. The activities of ^{90}Sr , $^{239,240}\text{Pu}$ and ^{241}Am were determined using the standard methods of radiochemical analysis. β -emitting radionuclides were detected by radiometer KRK-1-01, α -emitting radionuclides by α -spectrometer ALPHA-KING 676 A. The main agrochemical parameters of soils and soil solutions were estimated by standard methods [1].

3. RESULTS AND DISCUSSION

The locations of sampling sites, the levels of radioactive contamination of soil samples and their most important agrochemical characteristics are presented in Table 1. Specific activities of soil were in the range: 1.0 - 430 kBq/kg for ^{137}Cs , 0.2 - 31 kBq/kg for ^{90}Sr and 0.02- 0.5 kBq/kg for $^{239,240}\text{Pu}$ and ^{241}Am . The content of radionuclide in interstitial soil solution depends on the specific activity of the soil, the structure of absorbing soil complex and the chemical nature of the radionuclide. The radionuclide content in interstitial soil solution varied: from 0.6 to 77 Bq/l for ^{137}Cs , from 1.9 to 520 Bq/l for ^{90}Sr , from 0.02 to 1.1 for $^{239,240}\text{Pu}$ and ^{241}Am (Table 2).

Table 2. Chemical properties and specific activities (A, Bq/l) of interstitial soil solutions

Sample	pH	OM	Fe	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	A^{137}Cs	A^{90}Sr	$\text{A}^{239,240}\text{Pu}$	A^{241}Am
		mg/l							Bq/l		
SP-1	7.18	80	5.1	0.01	30.0	3.04	20.0	58.2±4.1	521±62	0.30±0.03	0.44±0.04
SP-2	6.63	17.5	4.3	0.02	4.7	<0.4	10.0	54.8±6.1	10.0±4.0	0.32±0.03	0.52±0.05
P-1	6.26	2030	77.5	0.12	67.5	27.3	90.2	76.9±6.7	197±20	0.74±0.07	1.13±0.11
S-2	6.32	300	NA	NA	31.5	NA	30.1	8.59±0.88	49.7±6.9	0.13±0.02	0.61±0.06
P-3	4.57	190	3.0	0.02	15.0	18.2	80.2	10.6±0.9	183±23	0.083±0.008	0.13±0.02
P-4	6.16	50	NA	NA	1.17	NA	10.0	3.17±0.54	NA	0.015±0.002	NA
SP-5	6.22	94	6.6	0.01	7.95	<0.4	2.00	32.4±3.2	19.6±2.3	0.29±0.04	0.90±0.09
SP-6	6.98	574	22.0	0.03	9.9	21.3	65.1	6.92±0.84	28.8±5.6	0.07±0.01	0.13±0.02
P-6	3.48	158	NA	0.01	1.8	<0.4	10.0	24.6±2.3	1.9±0.8	0.068±0.013	NA
SP-7	5.53	158	NA	NA	2.83	NA	35.1	1.83±0.25	NA	NA	NA
P-7	6.95	208	NA	43.3	14.8	NA	71.1	0.64±0.08	NA	NA	NA
P-11	8.18	30	NA	74.0	54.0	NA	371	7.02±0.93	NA	NA	NA

A close correlation between the content of radionuclide in interstitial soil solution and the total reserve of its exchangeable form in soil has been found. The maximum fraction of ^{90}Sr in the most soil solutions (up to 1.9 % of its total content in soil) corresponds to the maximum content of its exchangeable form in soil (up to 80 %). In the majority of uncultivated soils the fraction of the exchangeable form of ^{137}Cs is 0.6 - 5 %. Only in highly organic soil P-6 (OM is 95 %) does the fraction of the exchangeable ^{137}Cs form reach 30 %. In mineral soils a higher fraction of ^{241}Am in interstitial solution (0.03 - 0.8 %) in comparison with $^{239,240}\text{Pu}$ (0.02 - 0.6 %) corresponds to higher portion of exchangeable americium in soil. The average content of exchangeable forms of ^{241}Am and $^{239,240}\text{Pu}$ in the studied mineral soils is 6 % and 4 %. In organic soils fractions of plutonium and americium in interstitial soil solutions are comparable as well as portions of exchangeable forms of radionuclides in soils. Therefore, the physicochemical state of the radionuclides significantly influences their migratory properties. In general, an increase in the content

of mobile exchangeable forms of radionuclide in soil promotes the higher radionuclide content in interstitial soil solution and its higher mobility in environmental systems.

After the Chernobyl catastrophe the physicochemical state of radionuclides has changed, particularly in the areas with significant content of fuel particles in the soils (10-50 km from CNPP). This was a result of dispersion of the fuel particles and their destruction as well as interaction of radionuclides with natural components of the soil complex [2]. In the most investigated soils the physicochemical state of radionuclides, in principle, has stabilized and the properties of soil medium and chemical nature of radionuclide are the main factors determining their state and behaviour in soil. Only at the areas near to the CNPP (≤ 10 km) can the properties of fuel particles significantly influence the physicochemical state of radionuclides (for example, in the soil SP-1).

The coefficient of radionuclide distribution between solid phase and interstitial soil solution (K_d) is an important parameter describing radionuclide behaviour at the solid-liquid interphase. The distribution coefficient is a ratio of specific activities of radionuclide at equilibrium in solid and liquid phases. It is an important factor defining the sorption ability of soil. A higher K_d corresponds to a higher sorption ability of soils and lower specific activity of interstitial soil solution. This parameter can be used as a measure of radionuclide mobility in those systems where soil solution plays an important role in radionuclide migration. In such cases the higher distribution coefficient corresponds to a lower mobility of the radionuclide.

In the majority of soils the distribution coefficient (K_d) for ^{90}Sr is significantly lower and varies in a more narrow range than the corresponding coefficients for ^{137}Cs , $^{239,240}\text{Pu}$ and ^{241}Am (Fig. 1). This indicates much higher mobility of ^{90}Sr in comparison with other radionuclides in the most kinds of soil. Only in the highly organic soil, P-6, does the distribution coefficient for ^{90}Sr (540 l/kg) predominate over the K_d for ^{137}Cs (280 ± 40 l/kg). The soil type also influences the migratory properties of radionuclides. In soddy-podzolic soils, SP-1 - SP-7, the mobility of ^{137}Cs is higher (K_d : 290 - 1420 l/kg) than in alluvial soddy soil, S-2, (K_d : 4650 ± 670 l/kg). In accordance with the lowering of ^{137}Cs migratory properties, the organic soils can be arranged in the following order: highly organic peat, P-6, (280 ± 40 l/kg) \gg mineralized peat, P-4, (2790 ± 590 l/kg) $>$ peat gley, P-1 and P-3, ($5550 - 12400$ l/kg).

Cationic exchange plays an important role in the redistribution of ^{137}Cs between the solid phase and interstitial soil solution. Strong sorption of ^{137}Cs in soils is connected with its selective sorption at the specific Frayed Edge Sites (FES) and further fixation in the interlayer space of clay minerals of the illite group. In highly organic soils a nonselective ion-exchange sorption of the radioisotope plays a significant role. This is sorption of caesium cations at Regular Exchange Sites (RES) of the soil complex: carboxil, hydroxil and other functional groups of organic substances belonging to the solid phase and planar sites of soil minerals [3].

The mobility of ^{137}Cs essentially depends on the concentration of K^+ and NH_4^+ in water phase [3, 4]. These cations compete with ^{137}Cs for FES and increase caesium transfer from solid phase into interstitial soil solution and NH_4^+ is more selective to FES than K^+ . In the majority of soils, K^+ concentration in water phase is much higher than NH_4^+ (Table 2), but in humid soils with poor aeration there are good conditions for generation of NH_4^+ and its effect on behavior of ^{137}Cs significantly increases [3, 5].

Characteristic of ^{90}Sr is the predominance of ionexchangeable, unselective sorption by components of the absorbing soil complex. The behavior of ^{90}Sr mainly depends on organic matter content, water and cationic exchange capacity of soil, ionic strength, acidity (pH) of solution and concentration of calcium and magnesium in the water phase. In general, the mobility of ^{90}Sr in organic soils is lower (K_d : 87 - 540 l/kg) than in mineral ones (K_d : 45 - 67 l/kg, Fig. 1). It also noticeably depends on the ratio of mobile and immobile components of humus substances. Among organic soils with comparable organic matter content (for example, P-1 and P-3, Table 1) the higher concentration of water-soluble humus substances promotes the higher mobility of ^{90}Sr . In the soil P-1, with concentration of organic substances in interstitial solution 2030 mg/l the distribution coefficient of ^{90}Sr is 87 ± 16 l/kg and in the soil P-3 where organic matter concentration is 190 mg/l, it is 167 ± 33 l/kg.

The highest mobility of transuranic elements was found in soddy-podzolic friable sand, SP-2 and SP-3, (K_d : 80 - 340 l/kg), it being less in soddy-podzolic, SP-1, SP-6, and soddy, S-2, sandy loam ones (K_d : 230 - 1370 l/kg). The lowest mobility of $^{239,240}\text{Pu}$ and ^{241}Am (K_d : 1150 - 1370 l/kg) was found at the plot SP-1 nearest to CNPP (~ 10 km), where fuel particles were detected in the soil cover (Fig. 1).

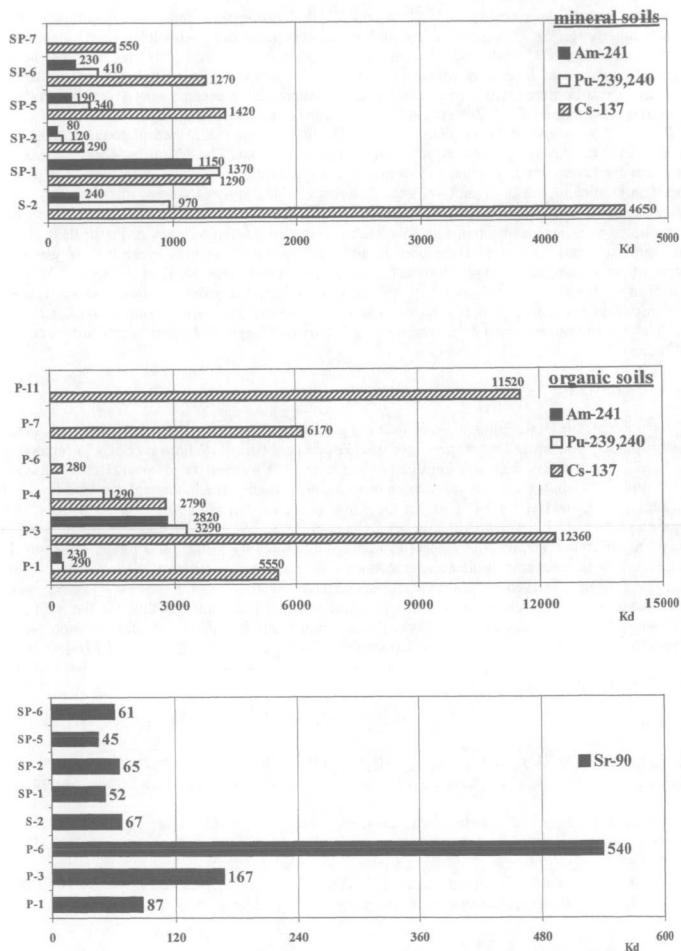


Figure 1: Distribution coefficients of radionuclides (Kd, l/kg) in the "solid phase - interstitial solution" system of soils

With respect to the mobility of $^{239,240}\text{Pu}$ and ^{241}Am , organic soils can be differentiated into two groups. Similarly to ^{90}Sr , in organic soils with high concentration of water-soluble organic components (as P-1) the mobility of $^{239,240}\text{Pu}$ and ^{241}Am is relatively high (K_d : 290 and 230 l/kg), and in soils, which are poor in water-soluble organic matter (as P-3), their mobility is extremely low (K_d : 3290 and 2820 l/kg). It can be noted that organic soils poor in water-soluble organic matter are more typical of Belarus and weak migration of $^{239,240}\text{Pu}$ and ^{241}Am in organic soils is more common.

There is a tendency for lower distribution coefficients of americium than plutonium, especially in mineral soils. This difference between radionuclides in organic soils is, as a rule, does not exceed the error in the coefficient determination. Consequently, the mobility of americium in mineral soils, as a rule, higher than plutonium, but in organic soils the difference in migratory properties of radionuclides is not so noticeable. The behavior of $^{239,240}\text{Pu}$ and ^{241}Am is related with chemical peculiarities of the radionuclides. They are in different oxidation states in the same solution, they differ in their ability to form complexes, and their similar compounds differ in solubility and hydrolyzability. In general, the entrance of americium into the soil solutions proceeds even more intensively than ^{137}Cs and $^{239,240}\text{Pu}$. The distribution coefficients of ^{137}Cs and $^{239,240}\text{Pu}$ are often of the same order. But the lower content of transuranic elements and their mobile forms in soils and significantly lower penetration of radionuclide through cell membranes of plants essentially restrict mobility and biological availability of $^{239,240}\text{Pu}$ and ^{241}Am .

4. CONCLUSION

Sorption of ^{90}Sr by the solid phase of soils is significantly lower than ^{137}Cs and transuranic elements and sorption of ^{241}Am by soils is respectively weaker compare with $^{239,240}\text{Pu}$, especially in mineral soils. As a result, ^{90}Sr transfers from the solid phase into interstitial soil waters more intensively than ^{137}Cs , $^{239,240}\text{Pu}$ and ^{241}Am . In a great extent it determines a higher mobility and biological availability of ^{90}Sr in ecosystems. A higher fraction of ^{241}Am in the water phase determines a higher mobility of ^{241}Am in comparison with $^{239,240}\text{Pu}$. The distribution coefficients of ^{137}Cs , ^{90}Sr , $^{239,240}\text{Pu}$ and ^{241}Am that were found allow us to differentiate soils with respect to radionuclide mobility in the solid phase - interstitial soil solution system. The separate soils corresponding to decrease in migration of ^{137}Cs , ^{90}Sr , $^{239,240}\text{Pu}$ and ^{241}Am have been observed. The influence of different factors on the physicochemical state of radionuclides is the main reason for specific distribution of individual radionuclides in the solid phase - interstitial soil solution system. The observed data expand our knowledge of migratory properties of radionuclides in the environment and give a scientific basis for development of new, and improvement of existing methods of restricting radionuclide migration into surface and underground waters and radionuclide accumulation by vegetation.

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