RESTRICTION OF PLUTONIUM AND AMERICIUM MOBILITY IN CONTAMINATED AREAS OF BELARUS

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Abstract

Mobility of plutonium and americium in Belarusian soils has been investigated. The parameters of vertical migration, distribution coefficients between the solid phase and pore solution of soil and the transfer factors of ^{239,240}Pu and ^{241}Am in soil – plant system have been estimated. Influence of agrochemical characteristics of soil on migration properties of radionuclide has been analysed. It was shown that bioavailability of ^{239,240}Pu and ^{241}Am depends on their concentration in soil solution of root-inhabited layer. In the most cases portion of ^{241}Am in the soil solution is higher than that of ^{239,240}Pu. Chemical composition of soil solution significantly influences the redistribution of radionuclides in the solid phase – soil solution – plant system. It was established that in the most kinds of peat the mobility and biological availability of ^{239,240}Pu and ^{241}Am is significantly lower than that in mineral soils and the peat additives essentially decrease their mobility in soddy-podzolic sandy ones. This measure decreases the radionuclide mobility with the factor 2 - 4 due to formation of insoluble organic and organic-mineral complexes of transuranic elements (TUE) with humus components of peat additive. Enrichment of mineral soil with peat is ecologically safe and allows improving of soil fertility.

Introduction

Global fallouts from weapon testing and accidents of nuclear reactors have caused the contamination of the environment by transuranic elements. Chernobyl catastrophe of 1986 was the most significant source of TUE in Belarus. Before it, the level of soil contamination was 50 Bq/m^2, and after catastrophe inventory of TUE exceeded 50 kBq/m^2. The main part of TUE is localized in 30-km zone, nevertheless, TUE of Chernobyl origin were also identified at significant distance from Chernobyl NPP (ChNPP). Whilst 17 different transuranic radionuclides have been found in the fallout, ^{239,240}Pu and ^{241}Am are still the most important ones in the present time (1, 2).

Despite of the low mobility of TUE in ecosystems, they will remain in the environment during long period of time and the critical assessment of their behaviour should be done for estimation of the long-term radiological safety. Up to now, restriction of ^{239,240}Pu and ^{241}Am mobility in the contaminated regions is an actual problem (1 – 4).

Complex of different countermeasures is necessary to restrict radionuclide mobility in soil – plant system. The present study deals with the investigation of ^{239,240}Pu and ^{241}Am mobility in contaminated areas of Belarus. The emphasis was made on the factors, which mostly influence on the plutonium and americium migration in soils and their accumulation by plants. On the basis of obtained results the method consisting in application of natural peat additive to restrict plutonium and americium mobility in soil media is proposed.

Objects and Methods

The studied area (51° of north latitude, 29 – 30° of east longitude) is located in the southeast of Belarus. The control sites are situated in Gomel region within 10 – 50 km zone of the ChNPP. The soils and vegetation of the investigated area are characteristic for suffered region of Belarus that allows using obtained data for analysis of radionuclide behavior on the contaminated territory. Soddy-podzolic sandy and loamy sand, alluvial soddy loamy sand, and different varieties of peat have been studied. Vegetation was represented by the species such as *Carex, Elitrigia repens, Dactilus glomerata, Festuca pratensis, Festuca ovina, Urtica dioica, Rhinansus, Cirsium arvence, Corinephorus canesecus, Calamagrostis, Poa pratensis, Conium, Cirsium arvence*.

The soils were sampled in the uncultivated sites up to 40-cm depth, air-dried, sieved, homogenized and ashed. The plant samples were washed by distilled water, air-dried, cut into the pieces of 2 – 5 mm, mixed and also ashed. The soil solutions were obtained from water-saturated soil samples using high-speed centrifuge SIGMA–4–10. Content of ^{239,240}Pu and ^{241}Am in the samples of soils, soil solutions and plants has been determined by the method of radiochemical analysis, which includes the stages of ^{239,240}Pu and ^{241}Am separation from hindered nuclides by ion-exchange chromatography and coprecipitation of TUE with cerium fluoride according to the standard radiochemical procedure. Detection of ^{239,240}Pu and ^{241}Am has been carried out by alpha-spectrometer ALPHA–KING 676 A. The agrochemical characteristics of soils and soil solutions (water capacity, acidity, content of organic matter, exchangeable calcium, magnesium etc.) have been determined using generally accepted analytical methods.

To investigate the possibility to restrict the ^{239,240}Pu and ^{241}Am mobility in the soils the following experiment has been carried out: samples of soddy-podzolic sandy soil contaminated by TUE of Chernobyl origin have been mixed with uncontaminated peat in ratio of 10 : 1 and 10 : 3 by mass. The obtained soil samples were homogenized, subjected to water saturation for further separation of pore soil solution in two weeks after moistening. Finally, the values of radionuclide distribution coefficients between solid phase and pore solution of soil have been estimated and compared with corresponding coefficients of soddy-podzolic sandy soil without peat additives.

Results and Discussion

Inventory and vertical migration of TUE in soils

Inventory of TUE in soils within 10 – 50 km zone and the main agrochemical characteristics of investigated soils are summarized in Tables 1, 2. It has been shown that inventory of ^{239,240}Pu and ^{241}Am in the soil-vegetation cover is 1.0 – 38 kBq/m^2 and significantly depends on the distance of site from ChNPP. Investigated soils are mainly presented by acidic and slightly acidic soils (pH_{KCl} 3.9 - 6.5). Content of organic matter (OM) is 1.0 – 7.5% of dry mass for mineral soils and 14 – 80% for organic ones. The mineral soils have much lower cationic exchange capacity (CEC — 30 – 120 meq/kg) than organic ones (230 – 1700 meq/kg).

Table 1. Inventory of ^{239,240}Pu and ^{241}Am in soils depending on distance from ChNPP. 2002.

Distance from CNPP, km	Average radionuclide inventory in soil-vegetative cover, kBq/m^2	
	^{239,240}Pu	^{241}Am
~ 10	28 ± 7	38 ± 10
20 – 30	4.7 ± 1.7	6.3 ± 1.4
40 – 50	1.0 ± 0.3	1.3 ± 0.5

Table 2. The main agrochemical characteristics of soils

Soil type	Water capacity, %	pH_{KCl}	OM, %	CEC, meq/kg
Mineral soils				
Soddy-podzolic sand	25 - 35	3.9 - 4.4	1.0 - 1.5	30 - 50
Soddy-podzolic loamy sand	30 - 60	5.0 - 6.6	2.5 - 7.5	60 - 170
Alluvial soddy loamy sand	80 - 90	4.8 - 5.1	6.5 - 7.5	100 -120
Organic soils				
Peat (highly mineralized)	60 - 65	6.2 - 6.5	14 - 30	230 - 600
Peat	140-260	4.0 - 6.0	40 - 80	600 - 1700

On the basis of data on vertical redistribution of radionuclides in the soil profiles the migration rates of ^{239,240}Pu and ^{241}Am in soils have been estimated. It has been shown that the variation ranges of migration linear rates are 0.15 – 0.40 cm/year for ^{239,240}Pu and 0.25 – 0.50 cm/year for ^{241}Am. In a whole, the migration ability of radionuclides in mineral soils is higher than that in organic ones.

Behavior of plutonium and americium in the solid phase – soil solution system

Sorption-desorption of TUE in the solid phase - soil solution system affects significantly their mobility in ecosystems. In this connection, information on radionuclide distribution in such system is useful for

prediction of its behavior in the soil-vegetation cover. Content of ^{239,240}Pu and ^{241}Am in the soil solutions has been established. In the (0 – 15)-cm layers of soil of 60-100%-water capacity it varies in the range: 0.005 – 0.88 Bq/l for ^{239,240}Pu and 0.006 – 1.03 Bq/l for ^{241}Am. In the majority of soil solutions the americium content is higher than that of plutonium and does not exceed 1.5% from the total content of radionuclide in soil. The main properties of the soil solutions have also been determined. Among organic soils the group with the extra high content of water-soluble organic components (2000 – 2300 mg/l) has been found. To reveal the most important solution characteristics, which influence on radionuclide transfer from solid phase to soil solution, the received data have been statistically processed. It has been shown that in composition of soil solution content of water-soluble organic components was the most important. There is a direct interrelationship between activity concentration of ^{239,240}Pu or ^{241}Am and content of organic substances in soil solution.
An important parameter characterizing sorption capacity of soil to radionuclide and its mobility in soil is the distribution coefficient (K_d) that represents the ratio between activity concentrations of radionuclide in the solid phase and equilibrium soil solution. The higher K_d corresponds to the higher sorption of radionuclide by the solid phase and its lower mobility in soil. The average values of K_d for various types of soil with different total content of organic matter are presented at Fig. 1. There is a close correlation between K_d and organic matter content, especially for mineral soils, where plutonium and americium are more mobile (K_d are smaller) than in organic ones. Analysis of obtained data for organic soils revealed the influence not only the quantitative, but also the qualitative composition of organic matter on TUE behaviour. In organic soils with extra high content of water-soluble organic components the values of K_d are much lower in comparison with typical peat depleted in soluble organic constituents.

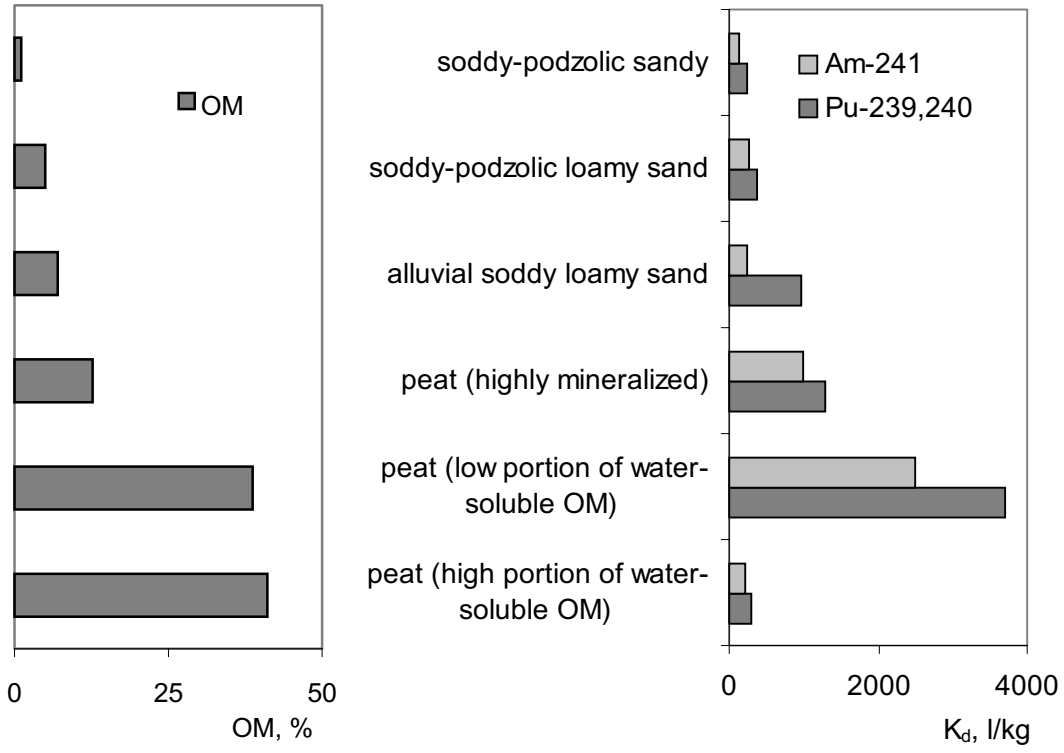

Fig. 1. Content of organic matter in soils (OM) and distribution coefficients of ^{239,240}Pu and ^{241}Am in the solid phase – pore solution system of water-saturated soils (K_d)

Soil humidity and acidity also significantly influence on the radionuclide redistribution in the solid phase – soil solution system. The typical soil humidity dependencies of K_d for mineral and organic soils have been discussed in our previous work (4). Interrelationships between coefficients K_d and pH_{KCl} of soil are presented on Fig. 2. Acidification of soil decreases the stability of organic-mineral complexes of TUE and, as a result, mobility of plutonium and americium increases. The relationship between K_d of plutonium and pH_{KCl} of soil is closer and more pronounced than that of americium. The possible reason of that is the higher ability of plutonium to form complexes with insoluble components of soil humus in comparison with that of americium. The influence of organic matter on migratory properties of radionuclides has been discussed in more detail in our previous work (5).

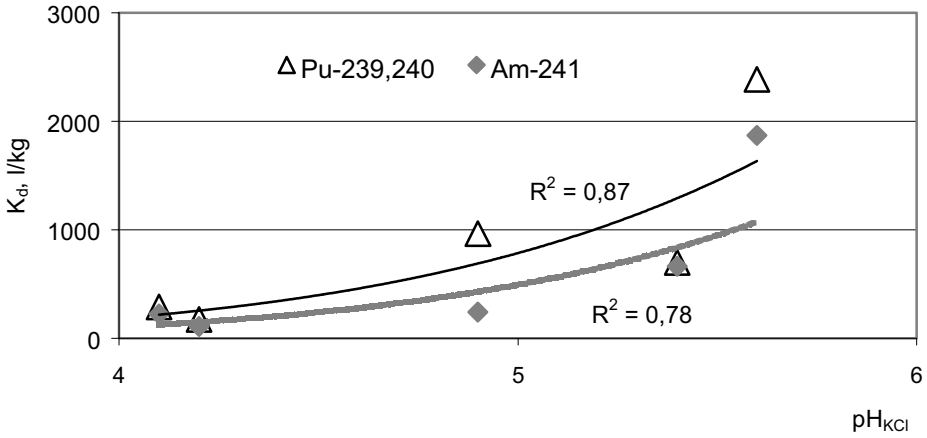

Fig. 2. Distribution coefficients of ^{239,240}Pu and ^{241}Am (K_d) in the solid phase – soil solution system depending on acidity of soil (pH_{KCL})

Soil – plant transfer of plutonium and americium

The activity concentration of ^{239,240}Pu and ^{241}Am in plants varies from 0.2 to 9.0 Bq/kg and is higher for americium. It depends on the plant specie, contamination level of soil and soil solution.

The accumulation coefficient (K_a) has been used for characteristic of soil - plant transfer of radionuclide. It is equal to the ratio between activity concentrations of radionuclide in plant (A_p, Bq/kg) and root-inhabited layer of soil (A_s, Bq/kg). The studied soils can be arranged in the decreasing order of radionuclide uptake by vegetation of meadow sedge-herbaceous association: soddy-podzolic sand (average value of K_a is 0.025 for ^{239,240}Pu and 0.048 for ^{241}Am) > soddy-podzolic loamy sand (0.012 and 0.026) > alluvial soddy loamy sand (0.010 and 0.020) > peat (0.004 and 0.010). The coefficients K_a of plutonium and americium correlate inversely with the radionuclide distribution coefficients K_d (Fig. 3).

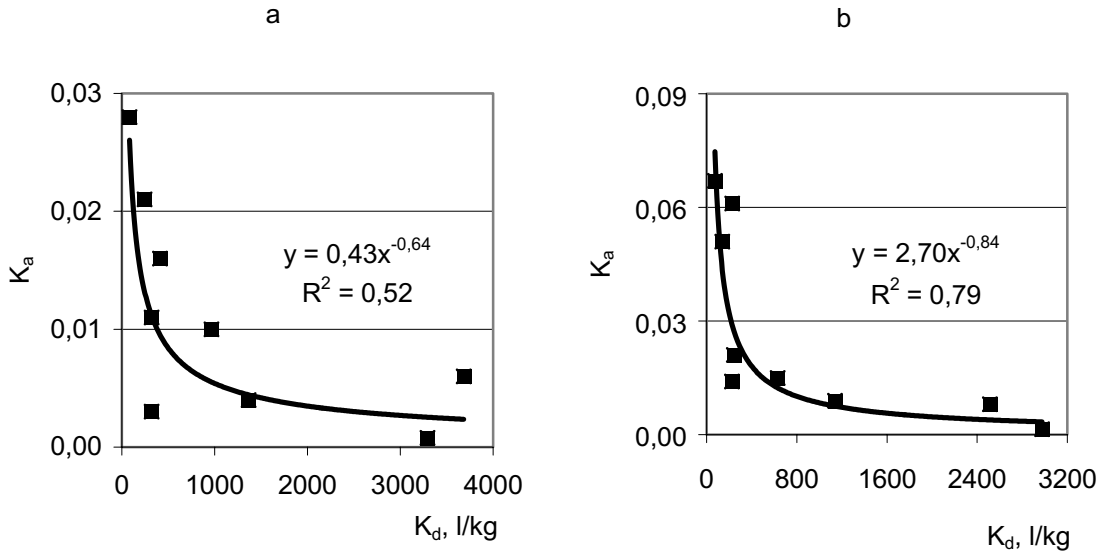

Fig. 3. The interrelationship of the coefficient K_a and coefficient K_d for ^{239,240}Pu (a) and ^{241}Am (b)

The higher K_a corresponds to the lower K_d. In the K_d range of 100-1000 l/kg the coefficient K_a decreases significantly (by a factor 3 - 5), and does not reduce noticeably in the K_d range of 1000-3000 l/kg. During uptake of radionuclides by root system of plant the dynamic equilibrium between mobile forms of radionuclides in the solid phase - soil solution system is kept. It means that solid phase of soils contaminated by TUE is a potential long-term source of radionuclides transfer from soil into the soil solution and further into the plants.

Effect of peat additive to restrict the mobility of plutonium and americium

In contaminated regions of Belarus acidic soddy-podzolic sandy soils are widespread. They are characterized by not high productivity due to the low content of humus and nutrients. The different countermeasures for decreasing of radionuclide mobility should be implemented on a cost-benefit basis. To restrict the mobility of plutonium and americium in the soil – plant system we suggest the method of natural peat additives with the low content of water-soluble organic components, taking into account above-mentioned data. This measure can allow decreasing the plutonium and americium mobility in the soils due to the radionuclide fixation by peat organic components of low solubility. The method of peat additive has been tested in laboratory conditions as described before, and appropriate values of K_d have been estimated (Table 3). The coefficients K_a have been predicted using regression equations characterizing relationship between K_a and K_d (Fig. 3).

Table 3. Experimental values of K_d and predicted coefficients K_a for $^{239,240}Pu$ and ^{241}Am

Experiment	$^{239,240}Pu$		^{241}Am	
	K_d, l/kg	K_a (predicted)	K_d, l/kg	K_a (predicted)
Control (soddy-podzolic sandy soil)	171	0.016	98	0.057
Soddy podzolic sandy soil + peat (10:1)	325	0.011	182	0.034
Soddy podzolic sandy soil + peat (10:3)	673	0.007	322	0.021

It has been established that after application of peat additives K_d of TUE changed by a factor of 2 - 4 in comparison with control sample of sandy soil. For the soil samples with additives of peat the soil-plant transfer of radionuclides as estimated has to be lower in 2 - 3 times.

To conclude that the method works properly it must be tested in the field conditions. In any case, the obtained results allow making the preliminary conclusion that application of peat additives decreases the mobility and bioavailability of plutonium and americium in the soil – plant system. This measure can also improve fertility of soil that has evident economical profit in regions with soils of low productivity.

Conclusions

Results of our study demonstrate clearly that:

- Mobility and bioavailability of plutonium and americium are low and in the most of the soils they are higher for americium than those of plutonium.
- Water capacity, acidity, quantitative and qualitative composition of organic matter of soil influence significantly on TUE behaviour in a soil – plant system.
- In organic soils mobility and bioavailability of plutonium and americium are much lower in comparison with those in mineral ones.
- Application of peat additives to soddy-podzolic sandy soil allows restricting of plutonium and americium mobility by a factor of 2 - 4.

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