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## **ОСОБЕННОСТИ РАСПРЕДЕЛЕНИЯ <sup>90</sup>Sr В СИСТЕМЕ ПОЧВА–РАСТЕНИЕ В НАТУРАЛЬНЫХ УСЛОВИЯХ**

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*Было изучено распределение <sup>90</sup>Sr в системе «почва – подземная часть растения – надземная часть растения». Установлено, что удельная активность <sup>90</sup>Sr в подземной и надземной частях исследованных растений, а также во мху не зависела от удельной активности этого радионуклида в почве. Между удельной активности <sup>90</sup>Sr в подземной и надземной частях растений установлена прямая пропорциональность. Коэффициент накопления <sup>90</sup>Sr из почвы в подземную часть растений и в мох был прямопропорционален удельной активности этого радионуклида в исследованных растениях.*

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## **INVESTIGATION OF PLUTONIUM BEHAVIOUR IN ARTIFICIALLY CONTAMINATED SOIL**

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*The vertical migration and transformation of plutonium chemical forms artificially supplied to sandy loam columns after its exposure to natural conditions for about one year was investigated. An analysis of artificially contaminated samples after one year had shown that 81% of <sup>239</sup>Pu<sup>4+</sup> and 44% of <sup>239</sup>Pu<sup>3+</sup> were accumulated in the 0-5 cm layer of sandy loam. The data of sequential analysis of the same type of soil at the adequate artificial contamination level after one month exposure under laboratory conditions are presented as well. <sup>239</sup>Pu binding to soil geochemical fractions was rather uneven. The largest amount of <sup>239</sup>Pu (60 %) was determined in the residual fraction. Consequently, it can be assumed that organic substances and some inorganic compounds, which usually are the main components of a residual fraction, affects the retention and migration of plutonium in the soil.*

Investigation of plutonium as most hazardous and radiotoxic element behavior in the environment from radioecological and geophysical point of view is of particular concern. The diversity of the chemical forms being released from different sources – from insoluble oxides to soluble inorganic salts and organic complex compounds – is characteristic for plutonium. Because of the high temperature during the nuclear explosion, insoluble plutonium oxides, mixtures of oxides or compounds with the constructive elements (PuO<sub>x</sub>·MO<sub>x</sub>; PuO<sub>x</sub>·U<sub>3</sub>O<sub>8</sub>) are predominant. The soluble forms of plutonium (PuO<sub>x</sub>·nH<sub>2</sub>O; Pu(NO<sub>3</sub>)<sub>x</sub>, Pu organic complex compounds) are basically released to the environment from the nuclear technological cycle (1, 2). The physico-chemical forms of released radionuclides determine their interactions with soils and, thus, the degree to which soils can act as a sink or a diffuse source of contaminants (3)

Processes of migration and accumulation of plutonium in that environmental system are in tight connection with the stability and transformation of its chemical compounds because of a reciprocity between components of investigated soil and plutonium chemical species (4-7).

The aim of the present study was to determine vertical migration of <sup>239</sup>Pu in artificially contaminated sandy loam depending on the radionuclide initial chemical form and to evaluate <sup>239</sup>Pu binding to soil geochemical fractions.

### **MATERIAL AND METHODS**

Three series of field experiments with three columns of non-destroyed sandy loam were carried out. Experiments were designed as follows: the plastic columns (diameter-10cm, length-20cm) were stuck into the soil at all length. The sandy loam surface in a separate column was artificially contaminated with <sup>239</sup>Pu (30 Bq) in the form of <sup>239</sup>Pu (NO<sub>3</sub>)<sub>4</sub>, <sup>239</sup>PuCl<sub>3</sub> or <sup>239</sup>PuO<sub>2</sub>. The arranged experimental system retained the original microbiological and mechanical properties of soil. The interaction and vertical migration of added Pu species (III, IV) in a sandy loam has been followed as a function of contact time (326 days). After exposition each column was divided into 4 layers (5cm). The soil was dried at room temperature, the plants and roots were separated from soil. The soil was precisely ground and mixed. The amount of organic matter in the soil sample was obtained by a loss-on-ignition analysis (550°). The soil reaction (pH) was measured with a glass electrode in the 25 ml 1M KCl solution

which was intensively agitated with 10 g of soil. Radiochemical analysis of plutonium was carried out on an aliquot of 50 g of soil according to the method described by Druteikiene et al (8).

Laboratory experiments have been performed where the tracer contaminated ( $^{239}\text{Pu}$  30 Bq) the same sandy loam as in the field experiments was subject to sequential extraction with some characteristic chemical reagents. Columns prepared in this way were left under laboratory conditions for 30 days. By the sequential extraction method (9) water-soluble, ion-exchangeable, acid-soluble, and residual fractions of  $^{239}\text{Pu}$  were separated.  $^{242}\text{Pu}$  as yield monitor was applied.

Plutonium isotopes were determined by alpha-spectrometry using a Canberra PD type detector (area 450 mm<sup>2</sup>, resolution 17 keV (FWHM) at 4-6 MeV). Alpha-efficiency was 25%, the detection limit to the counting time of 86400 seconds was about 10<sup>-3</sup> Bq of  $^{239,240}\text{Pu}$ .

## RESULTS AND DISCUSSION

The highest  $^{239}\text{Pu}$  activity concentration was obtained in the upper 0-5 cm sandy loam layer contaminated with  $^{239}\text{PuCl}_3$ ,  $^{239}\text{Pu}(\text{NO}_3)_4$ ,  $^{239}\text{PuO}_2$  after almost one year exposition under natural environmental conditions (Fig.1). Accumulation of plutonium activity concentration from  $^{239}\text{Pu}(\text{NO}_3)_4$  in the top soil horizon of the sandy loam came up to 81.0 %, while in the 15-20 cm layer it amounted only to 2.5 %. The data presented as percentage amount everywhere are relative to initial contamination. A smaller amount of  $^{239}\text{Pu}$  from chloride (44.1%) was observed in the 0-5 cm soil layer, and through all deeper layers its quantity was very similar to that of  $^{239}\text{Pu}$  in nitrate form. Vertical migration of insoluble  $^{239}\text{PuO}_2$  had an analogous tendency as in the case of soluble  $^{239}\text{Pu}$  compounds. The largest share of  $^{239}\text{Pu}$  activity in  $^{239}\text{PuO}_2$  form was obtained in the upper 0-5 cm sandy loam layer (92 %). An obvious decrease in the plutonium activity concentration was determined in the lower layers. The activity of whole layers constituted only to 1-6%. It can be assumed that reduced mobility of  $^{239}\text{Pu}$  in soluble nitrate form can be related to the formation of insoluble products of hydrolysis and Pu(IV) interaction with the mineral and organic fractions of soil. A smaller amount of  $^{239}\text{Pu}$  (III) in chloride form (44%) was observed in the 0-5 cm sandy loam soil layer. Through all deeper layers its quantity was very similar to that of  $^{239}\text{Pu}$  in nitrate form. It allows us to assume that Pu(IV) is the most stable valence form which forms stable complex compounds immediately influencing vertical migration. Referring to (2) the mobility of valence forms decreases in the following order: Pu(V) > Pu(VI) > Pu(III) > Pu(IV). On the other hand, the constants of the stability of complex compounds decrease in the cation (metals) line M(IV) > MO<sub>2</sub>(II) > M(III) > MO<sub>2</sub>(I) and in the anion line F<sup>-</sup> > NO<sub>3</sub><sup>-</sup> > Cl<sup>-</sup> > ClO<sub>4</sub><sup>-</sup> (1). Partition of insoluble  $^{239}\text{PuO}_2$  through all studied soil horizons (0-20 cm) confirms the statement that insoluble and heavily mobile plutonium compounds released onto the soil and affected by various environmental factors are transformed to mobile forms. Some part of them migrates to depth.

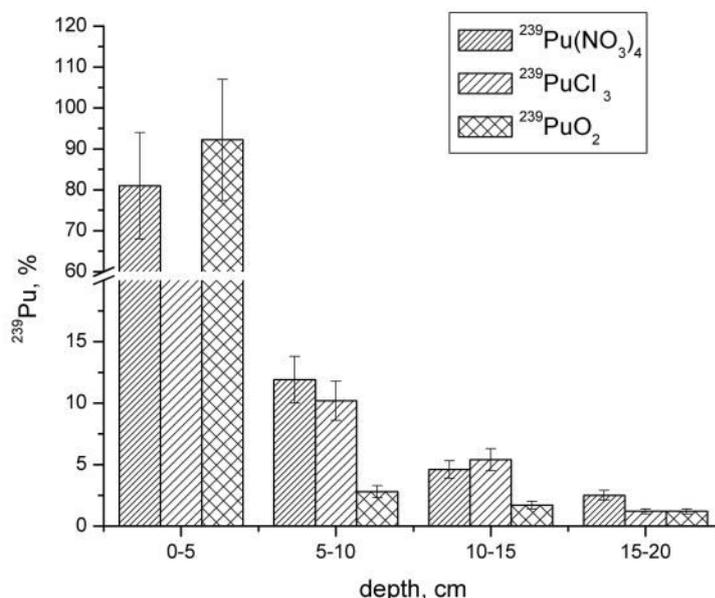


Fig.1. Downward migration of different chemical forms of plutonium in sandy loam

Series of another design experiments allowed us to compare  $^{239}\text{Pu}$  binding to soil geochemical fractions. After 30 days exposition artificially contaminated soil columns were analyzed for  $^{239}\text{Pu}$  using the sequential extraction method (10-13). The largest amount of  $^{239}\text{Pu}$  (60%) was determined in an acid-soluble fraction. A residual fraction contained almost 20% of supplied  $^{239}\text{Pu}$  activity concentration (Fig.2). The presence of radionuclides in these fractions is conditioned by their binding to organic matter, to oxides or amorphous silicic acids.  $^{239}\text{Pu}$  is rather irreversible in crystal lattice of soil minerals. Conjunction with organic part of soil and sorption into insoluble compounds or Fe, Al, Mn oxides are more characteristic of  $^{239}\text{Pu}$ . The results of laboratory experiments allow understanding the reasons of  $^{239}\text{Pu}$  accumulation in the upper soil layer under field conditions. Furthermore, being in the soil matrix organic matter, various iron compounds and other chemical substances can bind plutonium and make difficult downward migration of this radionuclide in environmental soil as well.

## CONCLUSIONS

The long – range field study has shown that soluble and insoluble forms of plutonium ( $^{239}\text{PuCl}_3$ ,  $^{239}\text{Pu}(\text{NO}_3)_4$ ,  $^{239}\text{PuO}_2$ ) to a significant extent (from 44 to 92 %) were retained in the top (0-5 cm) horizon of the undisturbed sandy loam. However, the percolation of  $^{239}\text{Pu}$  chloride into the deeper soil was observed to be higher. Insoluble  $^{239}\text{PuO}_2$  showed the lowest mobility. The following mobility rank of studied  $^{239}\text{Pu}$  compounds according to our investigations was composed:  $^{239}\text{PuCl}_3 > ^{239}\text{Pu}(\text{NO}_3)_4 > ^{239}\text{PuO}_2$ .

The laboratory experiments demonstrated that  $^{239}\text{Pu}$  is basically bound with the residual and acid-soluble fractions, where on the average 80% of plutonium were leached. By explaining the plutonium retention in the upper soil layer in field experiments and in environmental samples, its significant binding to the residual fraction referring to the laboratory experiments should be taken into consideration.

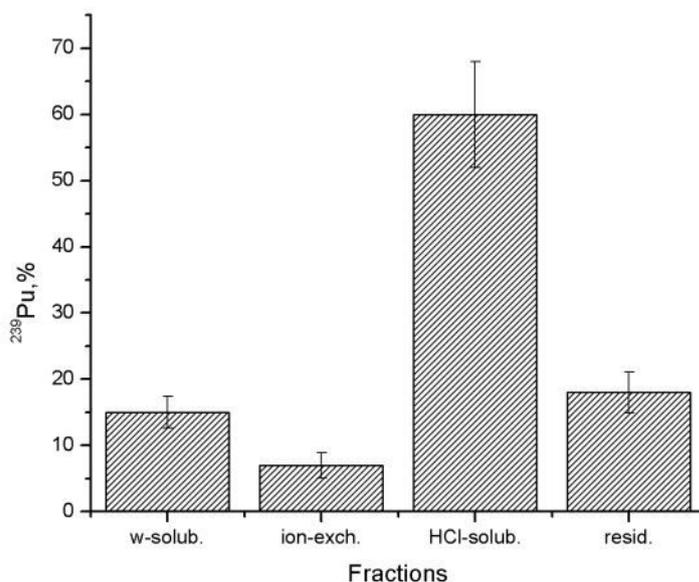


Fig.2. The distribution of  $^{239}\text{Pu}$  in geochemical fractions (water-soluble; ion-exchangeable; HCl-soluble; residual)

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