STUDIES OF THE PHYSICOCHEMICAL SPECIATION AND MOBILITY OF PLUTONIUM AND AMERICIUM IN BELARUSIAN SOILS

Iryna M. KIMLENKA, Svetlana V. OVSIANNIKOVA

Radiation Chemistry and Chemical Technology Department
Belarusian State University, Minsk, Belarus

The behavior of plutonium and americium of Chernobyl origin in soil media has been investigated. The studied area is located in the southeast of Belarus. Soddy-podzolic sandy and sandy-loam, alluvial soddy sandy-loam and peaty soils have been analyzed. Soil solutions were obtained from different types of soils by means of high-speed centrifuge SIGMA-4-A. The method of radiochemical analysis and α-spectrometer ALPHA-KING 676 A were used for the determination of ^{239,240}Pu and ^{241}Am content in the samples. The agrochemical properties of soils were determined by generally accepted analytical methods. Chemical fractionation, membrane filtration, ion exchange and gel-chromatography methods were utilized for the characterization of plutonium and americium species.

The distribution coefficients of plutonium and americium (K_d) between the solid phase and soil solution (a criterion of radionuclide mobility) have been evaluated under different humidification conditions characteristic for Belarus. The higher K_d corresponds to the lower mobility of radionuclide in soil. Results of our research demonstrate clearly that humidity, acidity, quantitative and qualitative composition of organic matter of soils influence significantly on Pu and Am behavior in soil media. An important finding is that the humic substances (HS) present govern the mobility of radionuclides. The soil organic matter promotes the fixation of plutonium and americium in the solid phase. Pu and Am were found mostly in the complexes with high molecular HS associated with Ca, stable hydroxides of Fe, Al and clay minerals. In most kinds of peat the mobility of Pu and Am is significantly lower (K_d reaches 3500 l/kg) than in mineral soils (K_d is less than 1000 l/kg). In the most of soils americium is more mobile than plutonium. Significant differences in radionuclide mobility in two distinct varieties of peat have been observed. In organic soils with extra high content of water-soluble organic components K_d is about 10 times less than in typical peat depleted in soluble organic constituents.

Particular attention has been focused on the investigation of the Pu and Am physicochemical speciation in soil waters. The soil solutions play an important role in the radionuclide transfer within the *soil-plant soil-surface water* and *soil-ground water* systems. A significant positive correlation between the content of the most mobile HS fractions in soils and the amount of radionuclides in respective soil solutions has been observed. The content of ^{239,240}Pu and ^{241}Am in the colloidal particles and organic complexes of different electrostatic charge has been evaluated. In soil waters 2-24 % of radionuclides were found to be associated with the particles in the 50-450 nm size range. HS with a mass > 2400 D sorbs most plutonium and americium present in soil solutions. The high percentage of Pu and Am (70-90 %) was found in the anionic and neutral organic complexes of soil solutions. The predominance of americium anion complexes in soil waters may be one of the reasons of its higher mobility in soils in comparison to plutonium.

Possibilities of the *soil – water – carboxylic resin* model system have been shown for the investigation of the physicochemical speciation, migration ability and biological availability of plutonium and americium. Carboxylic resin has been used as a model of the plant root system. The total amount of Pu and Am potentially available for plants and mobile in soils has been evaluated. This part of radionuclides can be remobilized if soil-climatic conditions alter. It has been shown the influence of HS on the radionuclide behavior. Experimental data are in good agreement with our findings for the *soil – soil solution – plant* system.