CHAPTER 25

STATE OF ECOSYSTEMS AT LONG-TERM CONTAMINATION WITH TRANSURANIUM RADIONUCLIDES

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Abstract: The levels of radioactive contamination with transuranium elements on territory of Belarus as a result of nuclear weapon tests and Chernobyl NPP accident have been assessed by 17 actinides.

The study of the atmosphere contamination with TUE in Republic of Belarus is being held since 1980 to now. A gradual decreasing of TUE content in the surface air to 3.2 nBq/m^3 in April 1986 as to $^{239+240}$ Pu. In the period of Chernobyl NPP accident (end of April - beginning of May, 1986) the $^{239+240}$ Pu content reached value of 120 μBq/m^3 in North part of Belarus. The period of half-removal of plutonium from nuclear weapon test and Chernobyl origin from the atmosphere according the observation period is the same and constitutes 14 ± 2 month. The mechanism of radioactive air pollution from April, 1986 is determined by dust transfer from radioactive contaminated regions. The value of this transfer is influenced considerably by agricultural activities on contaminated territory, forest fires and other anthropogenic factors.

A characteristic peculiarity of the Chernobyl NPP accident is the injection of small dispersed fuel particles containing TUE into biosphere.

The transfer coefficients in the soil- plant system vary from 7.1×10^{-6} to 4.0×10^{-3} for $^{239+240}$ Pu and 1.2×10^{-5} to 1.4×10^{-2} for ^{241} Am and have plant species dependence. The behavior of TUE in environment is discussed.

Keywords: transuranic elements; resuspension, plutonium; Chernobyl NPP accident; radioactive contamination; radionuclides

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Introduction

The nuclear weapon tests and different accidents at nuclear power plants caused the appearance of transuranium elements (TUE) in the environment. Their content is constantly increasing in separate components of biosphere. TUE have long half-life and include themselves into the circulation of substances by trophic chains. They will be radiologically dangerous for human during thousands of years. The alpha particles with energy higher than 5 Mev are present in the radiation spectrum of the majority of actinides. Therefore, by penetration of actinides into the organism, the alpha-radiation obtains the leading role by inducing of biological effects. High energy of alpha particles creates high ionization density in microvolumes of cells and tissues. Therefore the reparation processes are practically absent at the alpha-radiation action. As a result, the injuries caused by TUE sum up in the course of time. All this testifies the great danger of TUE incorporation into human organism and the necessity of thorough study of regularities of their behavior in the environment and biological effects.

Levels of Surface Soil Contamination and Isotopic Composition of Actinides Deposition

The territory of Republic of Belarus underwent the anthropogenic pollution with transuranium elements from two sources: firstly, the global fall-out after nuclear weapon tests, and, secondly, the fall-out as a result of Chernobyl NPP accident.

As a result of nuclear weapon tests, the principal levels of pollution of soil surface on Belarus territory were formed in the middle of 1970s and the level of pollution with $^{239+240}$Pu constituted 53 ± 17 Bq/m^2. Beside of $^{239+240}$Pu, other actinides precipitated also on the soil surface. Their relative composition is adduced on Table 1.

The Chernobyl NPP accident led to very uneven pollution of soil surface in Belarus with transuranium elements. The content of $^{239+240}$Pu of "Chernobyl" origin varied from 1.1×10^5Bq/m^2 on territories adjacent to Chernobyl NPP to average global levels in the north of Republic. At the active stage of Chernobyl NPP accident, the pollution of main components of biosphere was determined by short-living actinides ^{239}Np and^{242}Cm. Their contents were higher than the $^{239+240}$Pu activity 5.6×10^5 and 40 times respectively. In the global fall-out, these transuranium elements were practically absent. The constants growth of ^{241}Am contents in all components of Belarus ecosystem is observed as a result of radioactive decay of ^{241}Pu. The maximum level of pollution with ^{241}Am will be reached at 2060 and will exceed that of $^{239+240}$Pu 2.7 times. In our estimation, the areas with density of surface pollution of

TABLE 1. Composition of actinides on Belarus territory in the radioactive releases from nuclear weapon tests and Chernobyl NPP accident

		Relative content by the activity (at the Chernobyl NPP accident moment–26.04.1986)	
Radionuclide	Half-life	"Chernobyl" fall-out	Global fall-out from the nuclear weapon test
^{235}U	7.13×10^8 y	$(1.9 \pm 0.4) \times 10^{-4}$	–
^{236}U	2.39×10^7 y	$(1.0 \pm 0.7) \times 10^{-3}$	–
^{238}U	4.56×10^9 y	$(3.1 \pm 0.6) \times 10^{-3}$	–
^{237}Np	2.4×10^6 y	$(2.9 \pm 0.7) \times 10^{-4}$	–
^{239}Np	2.35 d	$(5.2 \pm 1.1) \times 10^4$	–
^{236}Pu	2.85 y	$(1.1 \pm 0.3) \times 10^{-4}$	–
^{238}Pu	87.1 y	0.99 ± 0.02	0.054 ± 0.010
^{239}Pu	2.41×10^4 y	1	1
^{240}Pu	6540 y	1.44 ± 0.04	0.58 ± 0.03
^{241}Pu	14.4 y	210 ± 10	6.4 ± 0.3
^{242}Pu	3.76×10^5 y	$(2.1 \pm 0.2) \times 10^{-3}$	$(2.4 \pm 0.1) \times 10^{-3}$
^{241}Am	452 y	0.17 ± 0.03	0.57 ± 0.15
^{242m}Am	152 y	$(2.9 \pm 0.7) \times 10^{-3}$	–
^{243}Am	7380 y	$(2.2 \pm 0.6) \times 10^{-3}$	–
^{242}Cm	163 d	28 ± 5	–
^{243}Cm	28.5 y	$(4.5 \pm 0.2) \times 10^{-3}$	–
^{244}Cm	18.1 y	0.15 ± 0.03	–

soil with $^{238+239+240}Pu$+ ^{241}Am up to 3.7 kBq/m^2 will expand out of the limits of alienation zone in west and north-west direction by 20–30 km.

The Air Contamination in Belarus with Transuranium Elements

The pollution of air in the Republic of Belarus with transuranium elements before the Chernobyl NPP accident took place as a result of nuclear weapon tests in the atmosphere. After the intensive nuclear weapon tests in 1961–1962, the maximum annual contents of $^{239+240}Pu$ in the air of Belarus reached the value of 0.21 mBq/m^3 in 1963.

The study of radioactive pollution of the atmosphere in the Belarus with transuranium elements was carried out from 1980 to the present (Fig. 1). In the dynamics of radioactive pollution of the near ground air with $^{239+240}Pu$ before the Chernobyl NPP accident, the tendency is registered to the decrease of $^{239+240}Pu$ content from 57 nBq/m^3 in 1980 to 3.2 nBq/m^3 in

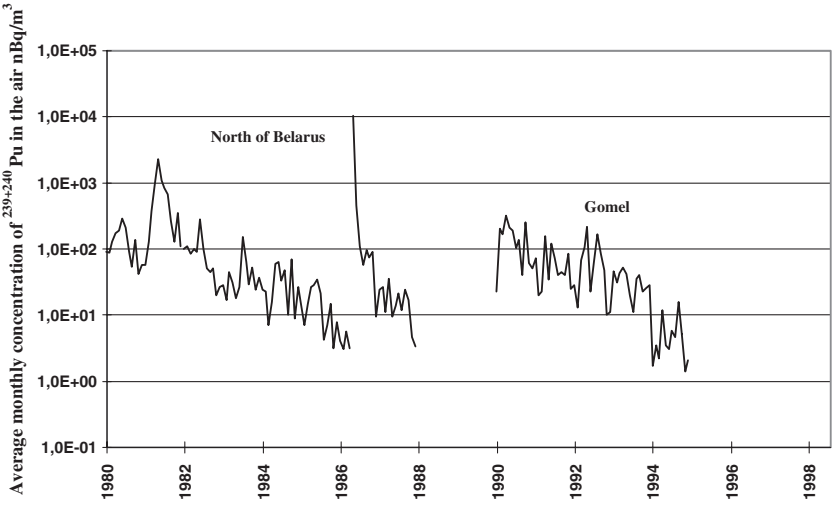

Fig. 1. The dynamic of the plutonium content in near ground air of Belarus.

April 1986. The increase of $^{239+240}$Pu in near ground air up to 3.5 μBq/m^3 in May, 1981 was the result of stratospheric fall-out from 26th Chinese nuclear test in atmosphere in October 1980.

There are three stages in the dynamics of radioactive pollution in the near ground air with $^{239+240}$Pu after Chernobyl NPP accident:

I. Sufficient increasing of radionuclides content in surface air during transfer of the radioactive cloud. At this period (end of April–beginning of May 1986) the $^{239+240}$Pu content has increased approximately by a factor of 5×10^4 in the north of Belarus and has reached the value of 120 μBq/m^3. The ratio of activities of ^{238}Pu/$^{239+240}$Pu has increased from 0.031 to 0.47.

II. Fast decreasing of radionuclides content in the air. This phase has lasted approximately till the mid of July 1986. At this period the levels of soil radioactive contamination have been arranged. $^{239+240}$Pu content in the air has decreased to 300 nBq/m^3 in the north of Belarus.

III. Slow decreasing of radionuclides content in the surface air. This stage has lasted and is being lasted up-to the present and is attributed to resuspension processes mainly.

The primary pollution of ground surface occurred in approximately two first weeks from the moment of accident. The overwhelming majority of radionuclides was released from the active zone of reactor into the atmosphere in that period. The dry precipitation and washout of radioactive aerosols from atmosphere formed the primary pollution of littering surface.

The systematic control of radioactive air pollution on sites within the alienation zone and adjacent zone is carried out. The observations of radioactive air pollution were carried out on the territories with different pollution density of soil (from 0.04 to 14 kBq/m^2 of $^{239+240}$Pu). The samplers were stood in the regional centers of Belarus (Gomel, Mogilev, Minsk, Mozyr, Brest), in the alienation zone of Chernobyl NPP and in the places where the agricultural and other activities are being held and the people are living.

The ratio of activities of ^{238}Pu/$^{239+240}$Pu in near ground air on all territory of Belarus correspond to "Chernobyl" one and constitutes 0.47 ± 0.05. This means that the pollution of air with transuranium elements after Chernobyl NPP accident is determined by the processes of transfer from radiocontaminated areas adjacent to Chernobyl NPP. As a result, the$^{239+240}$Pu content in near ground air in cities situated near alienation zone (Gomel, Mozyr) is 2–3 times as high as in other cities.

It can highlight the next components in the dynamics of average monthly levels of radioactive contamination of atmosphere: the determined trend of mid level, the cyclic component conditioned by seasonal variations and the occasional component which is the result of influence of multiple factors of anthropogenic and natural origin.

In the dynamics of average monthly levels of $^{239+240}$Pu content, there is seasonal cyclic component which conditions the spring growth of $^{239+240}$Pu content in near ground air as 4–5 times as compared with average annual level.

The forest fires on radioactively polluted territory in 1992 led to the increase of transuranium elements contents in air on all territories in the summer. Whereas the average annual content of $^{239+240}$Pu in Mozyr and Gomel in 1992 exceeded the level of 1991 and constituted 80 nBq/m^3.

As a result of study of mechanism of formation of radioactive pollution with transuranium elements, it was found that the levels of radioactive pollution of air with transuranium elements in the alienation zone and adjacent areas were determined both by the content of dust in the air and its specific activity. In the alienation zone, due to the cessation of economic activity, the concentration of dust in the air is approximately equal in all sites and does not exceed 10 μg/m^3. Therefore the transuranium elements content in the air of this locality depends linearly on the density of surface pollution of soil. In the areas adjacent to the alienation zone, the intensive agricultural activity in spring period leads to significant increase of dust content in the air (up to 1 mg/m^3). As a result, the transuranium elements content in the air of these area reaches and, in separate time periods, exceeds their content in the mostly polluted sites of alienation zone. The analysis of dynamics of average annual concentrations of radionuclides in the air of Belarus cities testifies

to the tendency of slow decrease of atmosphere pollution with transuranium elements. This process may be described quantitatively with the period of semi-cleaning of atmosphere.

The half-life in the atmosphere of the plutonium of "Chernobyl" origin and from nuclear weapon tests by the data for the period of 1980–1999 was equal for all observation sites and constituted 14 ± 2 months.

It was revealed by the methods of α-autoradiography and neutron-fragment radiography that there is inversely proportional square dependence of the specific activity of transuranium elements containing aerosol particles on their diameter. The existence of such dependence indicates that transuranium elements containing aerosol particles are not fuel ones at all, but they are non-radioactive aerosol particles, on the surface of which the smallest fuel particles are distributed. The dimension of elementary fuel particle on the average constitutes 0.1 μm, and the specific $^{239+240}$Pu activity in such a particle is ~10^8 Bq/g.

The Accumulation of Transuranium Elements by Plants and Animals

The range of varying the coefficients of plutonium and americium accumulation in wild species of plants on forest and meadow sites constituted by our data from 3.0×10^{-5} to 4.3×10^{-4} for $^{239+240}$Pu and from 1.2×10^{-4} to 1.8×10^{-3} for ^{241}Am in forest phytocoenosis and 1.9–6.7×10^{-4} for $^{239+240}$Pu and 4.6×10^{-4}–2.7×10^{-3} for ^{241}Am in meadow phytocoenosis.

The radiochemical analysis of transuranium elements content in above-ground phytomass of sowed fodder grasses in the alienation zone of Chernobyl NPP has shown in field experiment that biological mobility of transuranium elements in condition of cultivated soil increases as compared with non-cultivated soil of natural complexes: the values of transfer coefficients in cultivated species of grasses are higher by 10–100 than these in wild species.

It has been found the dependence of level of transuranium elements accumulation on the species of both wild and agricultural plant: the bush dominants – *Cytisus ruthenicus*, *Vaccinium myrtillus*, *Calamagrostis epigeios* – are accumulators in forest phytocoenoses, and among meadow grasses (in agrocoenoses and natural communities) - sedges and legumes.

The value of transfer coefficients of "Chernobyl" transuranium elements from soil to plants is within the limits determinated for "bomb" transuranium elements. The transfer coefficients from soil to plant of ^{241}Am are higher that those for plutonium because the americium is more soluble. It is necessary to note that the transuranium elements presently are transported to plant via the root system.

The specific activity in water of mostly radioactively polluted Belarus lakes reaches 0.98 Bq/m^3 for $^{239+240}$Pu and 11 Bq/m^3 for ^{241}Am. The main forms of plutonium in water are colloidal particles with adsorbed Pu(IV) and complex compounds of Pu(V) and Pu(VI).

The concentrations of $^{239+240}$Pu and ^{241}Am in wild animal body are 3.1 ± 1.4 mBq/kg and 1.2 ± 0.4 respectively, for global fall out, and reaches values 120 mBq/kg for $^{239+240}$Pu and 1300 mBq/kg for ^{241}Am for some "hot" regions in Belarus after Chernobyl NPP accident.