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Simulation of Entry and Propagation of Pu Isotopes and ^{241}Am on Ukraine Territory

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Abstract: Since the entry of transuranium elements (TUE) into the environment, the ^{241}Am content in different ecological system objects continues to increase as a result of ^{241}Pu beta decay. The ionizing radiation of these isotopes and their impact on the ecological situation in the propagation areas give rise to grave concern. The soil contamination with TUE radionuclides in Ukraine has stemmed mainly from two sources: global fallouts and Chernobyl nuclear power plant (NPP) accident. The ChNPP accident has led to nonuniform contamination with transuranium radionuclides, including Pu isotopes and ^{241}Am. To determine the contamination source characteristics, it was used the Receptor Modeling technique. For computing the trajectories of radionuclide propagation in atmosphere, and for preparing the radionuclide fallout maps, the HYSPLIT program was used. In consequence of the present studies it was constructed the maps of ^{241}Pu concentrations in air and soil in the territory of Ukraine in 1986. The forecast model has been created for ^{241}Am distribution at the time of the maximum isotope activity in 2059. The given results point to the necessary of further investigation of the data on the ^{241}Am accumulation and its effect on different ecosystems. This will permit the improvement of forecast models in emergency planning for protection of population health and rehabilitation of affected territories.

Keywords: Nuclear Power Plant, Contamination Source Identification, Transuranium Elements, Half-life Period, Radionuclide Distribution

1. Introduction

The development of nuclear power industry calls for the use of modern methods to control the environmental contamination with radionuclides of technogenic origin. Environmental monitoring is required to control the ecology state of the areas adjacent to nuclear power stations, spent nuclear fuel reprocessing plants, facilities of temporary radioactive-waste storage and disposal. In this respect, the high-priority goals are: to determine the sources of TUE entry to the territory of Ukraine, to analyze the TUE behavior, to estimate the risk of their entry to the human body.

The Chernobyl accident of 26 April 1986 considerably changed the radiation environment not only in Ukraine, but in Europe and in the world at large, too [1, 2]. One of the top-priority tasks of the accident management was to determine the radionuclide composition of the emission and the space distribution of radioactive contamination. For the 33 years that have passed since the accident, studies were made on the processes of radionuclide migration inside Ukraine, viz., in landscapes, soils, biological chains, water and geological environments. Relying on the data obtained, maps were constructed for the Ukrainian areas contaminated with radionuclides of vital importance for the man.

Specific circumstances of radioactive contamination formation, involving the peculiar properties of the source, the long-term character of its action, the changing radionuclide composition and the rate of emission, the impact of dynamic meteorological factors and diversified land forms, have resulted in a sharply heterogeneous pattern of fallouts, and thus, have complicated the solution of the assigned task. As a result of the Chernobyl NPP accident, a considerable part of North Ukraine has been contaminated with TUE, including

the ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{241}Am radionuclides [3].

To investigate the entry of the Pu and ^{241}Am isotopes inside the Ukraine, it is necessary to determine their content in the soil, to construct the maps of the levels of area contamination, and also, to study their migration process in the "soil-plant-human" chain. The aim of the present work has been to investigate the process and the sources of Pu isotopes and ^{241}Am entry on the territory of Ukraine.

2. The Subject of Research and Input Data

The paper is concerned with the data on the global and local contamination of Ukraine with Pu isotopes and ^{241}Am.

The accumulation of TUE in the environment is attributed to atmospheric nuclear weapon tests. The tests have been performed since early sixties by such countries as the USA, USSR, Great Britain, and later on, by France and China. This has led to TUE contamination of the environment on a global scope.

Locally, the environmental contamination with TUE is due to emission by nuclear-fuel cycle enterprises, operation of nuclear power units, and NPP accidents.

The Chernobyl NPP (ChNPP) accident has led to the formation of local regions with extremely high levels of radionuclide contamination (including Pu isotopes and ^{241}Am).

The isotopic composition of TUE produced on nuclear weapon testing differs from that resulting from NPP accidents. In global fallouts, the main radioactivity of TUE is contributed by ^{239}Pu (15-20%), ^{240}Pu (3-5%), and insignificant amount of ^{241}Am, with no Cm isotopes at all. The emissions from the destroyed 4th nuclear power unit of the ChNPP showed a wide range of transuranium isotopes: ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{242}Cm, ^{243}Cm, ^{244}Cm [4].

The amount and distribution of contaminant fallouts after the ChNPP accident were nonuniform, and were determined in many instances by the airstream strength and direction. For the period of active emission from the reactor (26 April to 6 May, 1986), the wind around Chernobyl had turned through 360 degrees with the result that the radioactive releases (of different compositions at different days) covered a great area (Figure 1). Under the action of wind, the radioactive releases were distributed over the territories of Ukraine, Belarus and Russia [5].

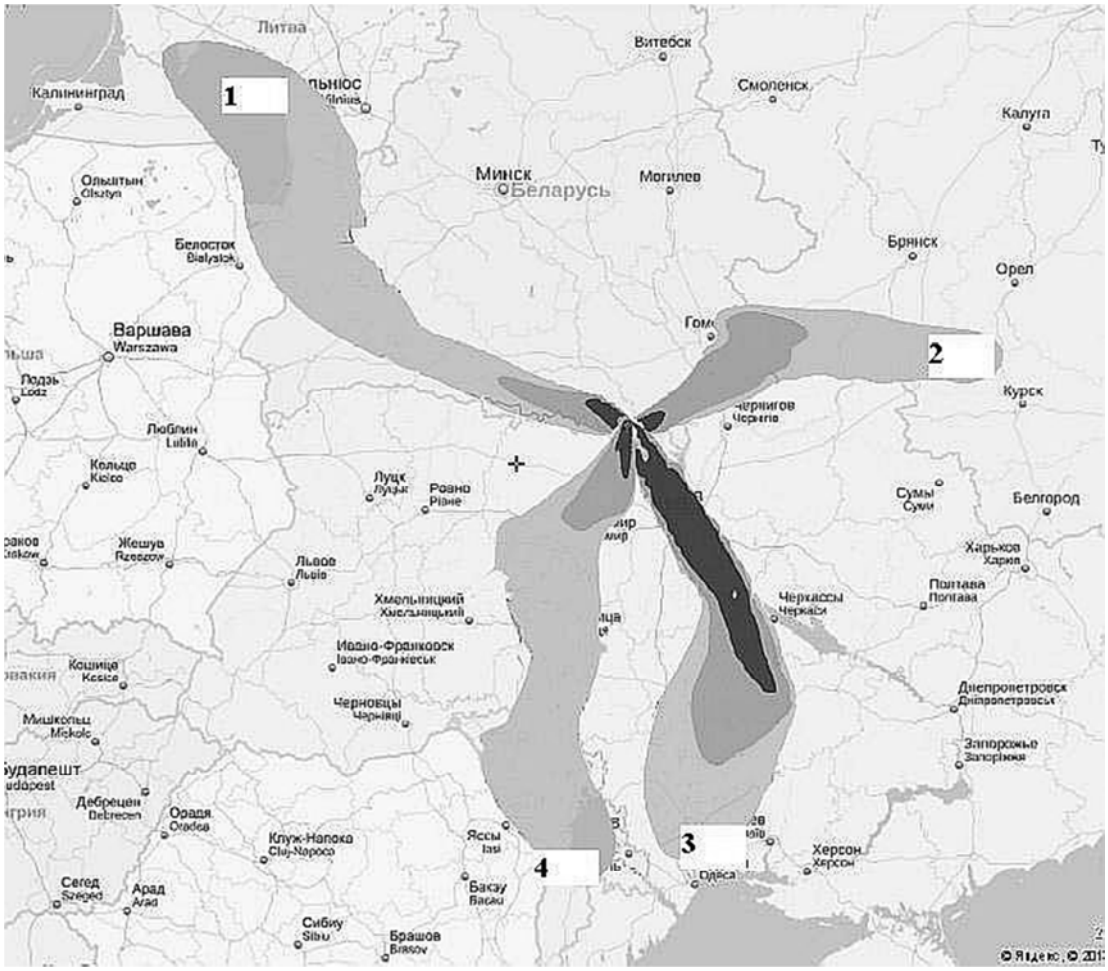

Figure 1. *Chernobyl radioactive emission formation in the first days after the accident in accordance with the weather conditions: 1 - 26 to 28 April; 2 - 29 to 30 April; 3 - 1 to 3 May; 4 - 5 to 6 May.*

The basic emissions from the fourth nuclear unit of the ChNPP lasted for ten days, and their compositions included radioactive gases, condensed aerosols and a great number of fuel particles. The distribution of emissions over time is shown in Figure 2.

During the first ten days after the accident the weather conditions often changed, resulting in considerable changes in the discharge dispersion parameters. Large particles, represented mainly by fuel particles, fell out within a radius of 100 km around the reactor, smaller particles were scattered by the wind for long distances.

The releases are characterized by the presence of radionuclide-containing substances with a wide range of forms and compositions such as gas, aerosol, vapor-aerosol mixtures; fuel particles; mineral particles-carriers of radionuclides condensed on them; agglomerates of different mineral forms; organic compounds. The contamination scale of Ukraine territory by ^{90}Sr, ^{241}Am and by isotopes of Pu, as opposed to ^{137}Cs contamination, is substantially smaller. The mentioned radionuclides belong to the group of hard volatile substances, and their basic amount came to the atmosphere mostly in the first phase of the accident after multiple core explosions on 26.04.1986. In the next days, their discharge in the flow of the vapor-aerosol-gas mixture was due to graphite burning, and then, as the core temperature went up to 2000°C and above, was also due to the increased fuel dispersion, the formation of more easily volatile polyelement compounds, the absorption on mineral particles [6].

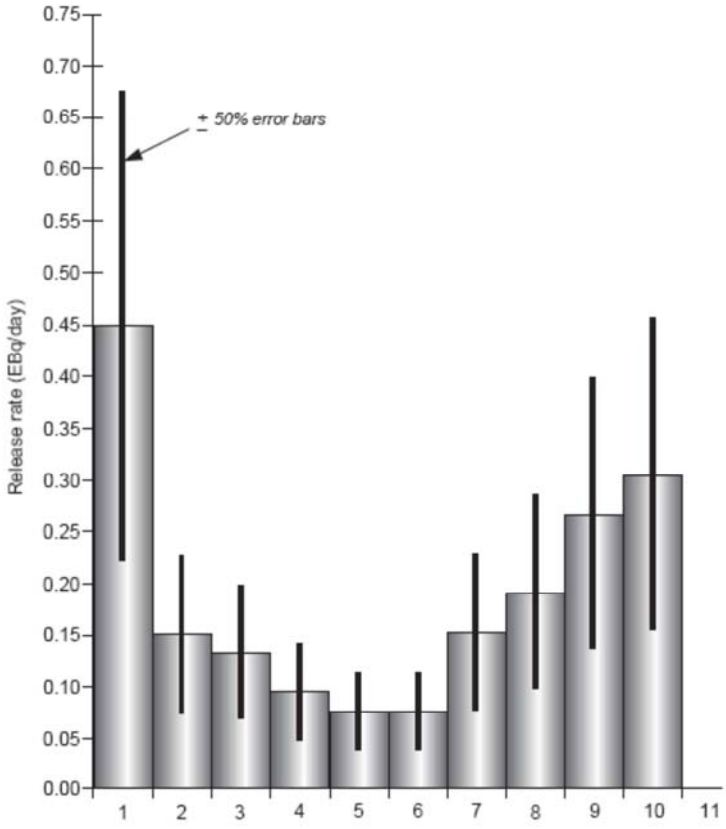

Figure 2. *The rate of radioactivity discharge into the air during the first 10 days after the ChNPP accident [7].*

In the first months after the ChNPP accident, the main works reduced to measurements of gamma-fields and determination of radionuclide content (mostly, γ-emitting nuclides) in individual samples of soil. The Pu content in the samples was calculated on the basis of the $^{239+240}$Pu /^{144}Ce activity ratio equal to $6.4 \cdot 10^{-4}$ as of 26 April, 1986. Later on, the data were obtained on the Pu and ^{241}Am content in the 30 km zone of the ChNPP. Those data were used to construct the maps of area contamination with principal long-lived radionuclides [8, 9].

Since the entry of TUE into the environment after the ChNPP accident (as a result of β-decay of ^{241}Pu), the ^{241}Am content in different ecosystems still continues to increase. A further α-decay of ^{241}Am will result in the production of α-emitting ^{237}Np ($T_{1/2}$ =$2.1 \cdot 10^{6}$ years), which will play an essential role in the radiation environment formation in the TUE-contaminated zone. In comparison with transuranium elements such as Pu, Am and Cm, ^{237}Np is more mobile. The lifetime cancerogenic risk factors for ^{237}Np are found to be: $1.5 \cdot 10^{-8}$ pCi^{-1} with inhalation intake; and $5.8 \cdot 10^{-11}$ pCi^{-1} with food intake [10].

Table 1 lists the data on the amount of Pu isotopes and ^{241}Am in the fuel of the 4th nuclear power unit [11] and in the accidental discharge. Beyond the ChNPP industrial site, the average emission of all radionuclides went to make up about 3.5% of the total amount of fission products being, which was in the reactor of the 4th nuclear unit during its operation by theory (this is except the nuclear fuel) [12].

The maximum ^{241}Am content in the environment will be reached by 2059 (i.e., in 73 years after the accident). It will be 40 times higher than its entry into the atmosphere during the accident in 1986 (Figure 3) [13].

The migration capacity increase of the Chernobyl ^{241}Am is determined by the destruction of fuel particles and the transition of radionuclides from inert to bioavailable forms. In contrast to ^{241}Pu, the ^{241}Am compounds have a higher solubility, and hence, a higher migration capacity [14]. For this reason, the now relatively safe territories may become dangerous to habitation of people.

Table 1. *Estimates of Pu isotopes and ^{241}Am emission during the Chernobyl NPP accident.*

Radionuclide	Decay period, $T_{1/2}$	Mass, kg	Total activity, PBq	Total emission during the accident	
				In percentage of total activity	Activity, PBq
^{238}Pu	87,7 years	2	1.3	3.5	0.0455
^{239}Pu	24065 years	412	0.92	3.5	0.0322
^{240}Pu	6537 years	185	1.5	3.5	0.0525
^{241}Pu	14.4 years	48	180	3.5	6.3
^{242}Pu	379000 years	20	0.0029	3.5	0.0001
^{241}Am	432.8 years	12	0.16	3.5	0.0056

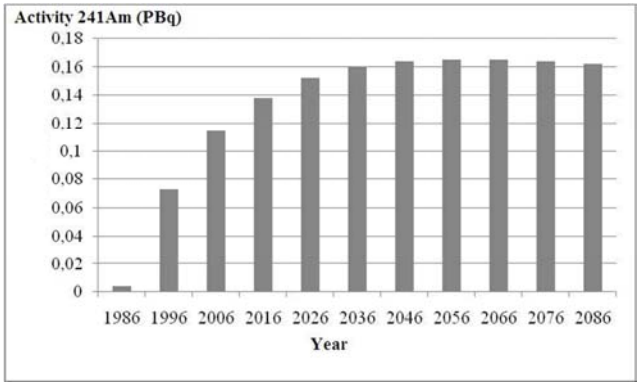

Figure 3. *Temporal dynamics of ^{241}Am activity accumulation inside Ukraine after the ChNPP accident.*

In the central and southern areas of the Kiev region, the contribution of the Chernobyl fallouts, as estimated from the ^{238}Pu/$^{239 + 240}$Pu and ^{241}Am/$^{239 + 240}$Pu ratios, varies between 75% and 85 %. The studies of the radionuclide composition and $^{239+240}$Pu, ^{241}Am contamination levels in certain regions of Ukraine have demonstrated that the western, southern and eastern regions of Ukraine are contaminated mostly with ^{238}Pu, $^{239+240}$Pu and ^{241}Am radionuclides, this being the result of global fallouts after nuclear tests in the fifties-sixties of the 20th century. The contribution of the Chernobyl fallouts, as estimated from the ^{238}Pu/$^{239 + 240}$Pu and ^{241}Am/$^{239 + 240}$Pu ratios, does not exceed 10 to 30%. The soil contamination with $^{239+240}$Pu in the majority of the Ukrainian regions is estimated to range from 30 to 500 Bq/m^2, this being in agreement with the data, obtained for some European countries [4].

Thus, the analysis of Pu isotopes and ^{241}Am propagation inside Ukraine will enable one to estimate the supply of the radionuclides in the ecosystems, and to predict their redistribution in the soil-cover complex.

3. Methods of Contamination Source Identification

The constructive approach to the problem of contamination source identification is based on the mathematical modeling in combination with experimental studies.

The mathematical model approach consists in constructing the models of occurrent physical processes with the use of mathematical formalism. The most effective forecast model of emergency situation presents a set of equations that take into account the physical processes taking place at a the accident object and in the environment. In meteorological forecasting, this is the set of equations for atmospheric hydrothermodynamics. The initial data for constructing the models are the dynamic and energy characteristics of the release, as well as the initial spatial distribution of pollutants and meteorological parameters.

For solving the diffusion problems there are two basic approaches: the gradient transfer theory and the statistical theory. The theory of gradient transfer from a continuous point source in atmosphere represents the Eulerian model, which considers the properties of the fluid/gas motion in relation to the fixed-in-space system of coordinates. As for the statistical theory considering the motion of separate particles, it turns up to be the Lagrangian representation [15].

To determine the contamination source characteristics, we have used here the Receptor Modeling technique described in refs. [16-18]. For computing the trajectories of radionuclide propagation in atmosphere, and for preparing the radionuclide fallout maps, the HYSPLIT program was used [19]. The program permits modeling of the process of aerial-contamination cloud formation and propagation from the given source, and unites the two approaches, i.e., Eulerian and Lagrangian.

4. Research Results

The soil contamination with TUE radionuclides in Ukraine has stemmed mainly from two sources: i) global fallouts, ii) ChNPP accident. In event of the Chernobyl NPP accident, the main release of Pu isotopes and ^{241}Am fell on the first two days, in the first stage of emission, after multiple core explosions on 26. 04. 1986. In the second stage, the intensity of emission decreased owing to the measures undertaken to stop graphite burning. At that period the radionuclide

composition in the emission was also close to that in the fuel. At that stage, finely dispersed fuel was brought out from the reactor by a hot stream of air and graphite burning products [20].

The processing of data on the Pu isotopes discharge from the emergency reactor (Table 1) has enabled us to construct the maps of ^{241}Pu concentration in air and fallout on the soil at the time of the main bang pip, i.e., on 26-29 April, 1986. The total radioactivity of ^{241}Pu release (3.5% of the reactor loading) was determined to be 6.3 PBq (see Table 1), the mass was 1.68 kg.

Figure 4 shows the map of contamination with ^{241}Pu on 26-28 April, 1986. The plume in those days was of north-west trending, therefore the neighboring areas of Belarus and Poland were also exposed to contamination with ^{241}Pu.

As a result of the south-wester on 29-30 April, 1986, the plume changed its direction to north-east (Figure 1). Figure 5 gives the map of ^{241}Pu contamination area for the time period from 28.04.1986 to 29.04.1986. As a result of change in the direction of wind, the plume started to shift in the line of Kaluga-Tula-Orel (Russia). So, the main bang pip of ^{241}Pu took place on 26-27 of April, in the early stage of the accident. As a result of the plume movement, the ^{241}Pu fallout on the Ukrainian territory took place directly in the 30 km zone around the reactor, and also, in the north-west, north and north-east directions away from the ChNPP.

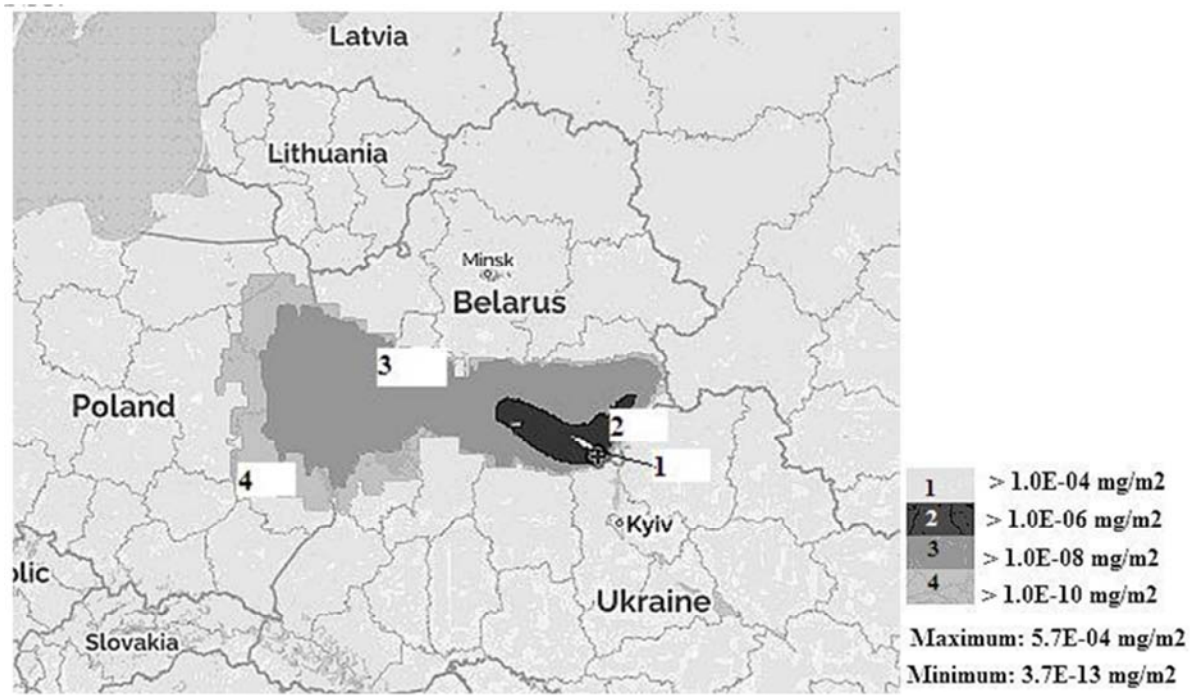

Figure 4. *^{241}Pu contamination from 26.04.1986 to 27.04.1986.*

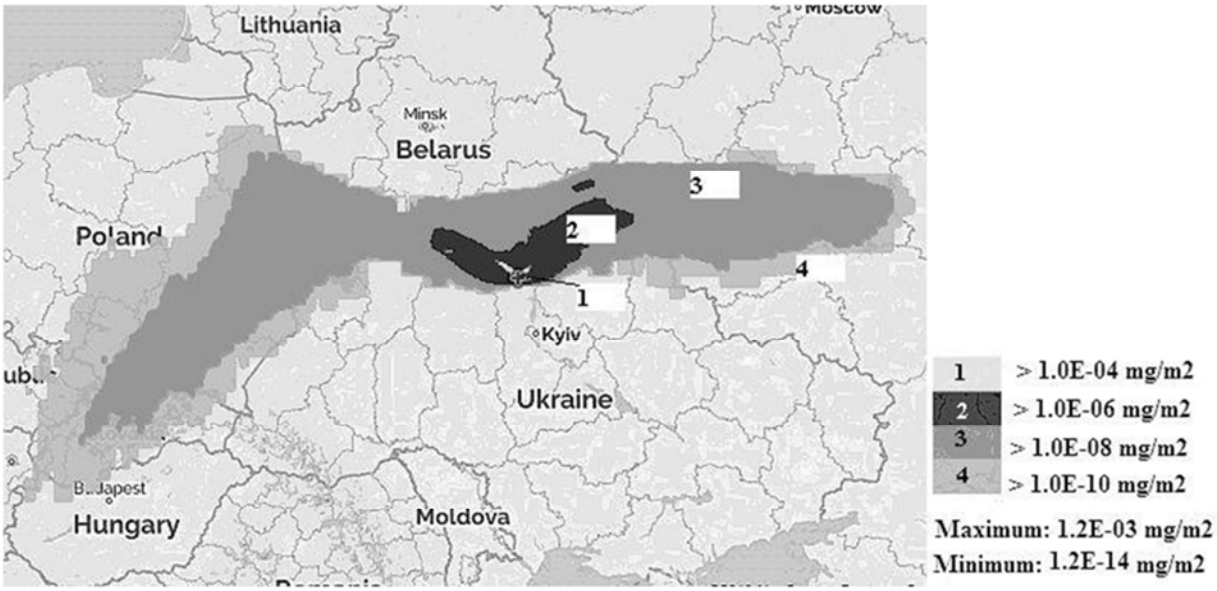

Figure 5. *^{241}Pu contamination from 28.04.1986 to 29.04.1986.*

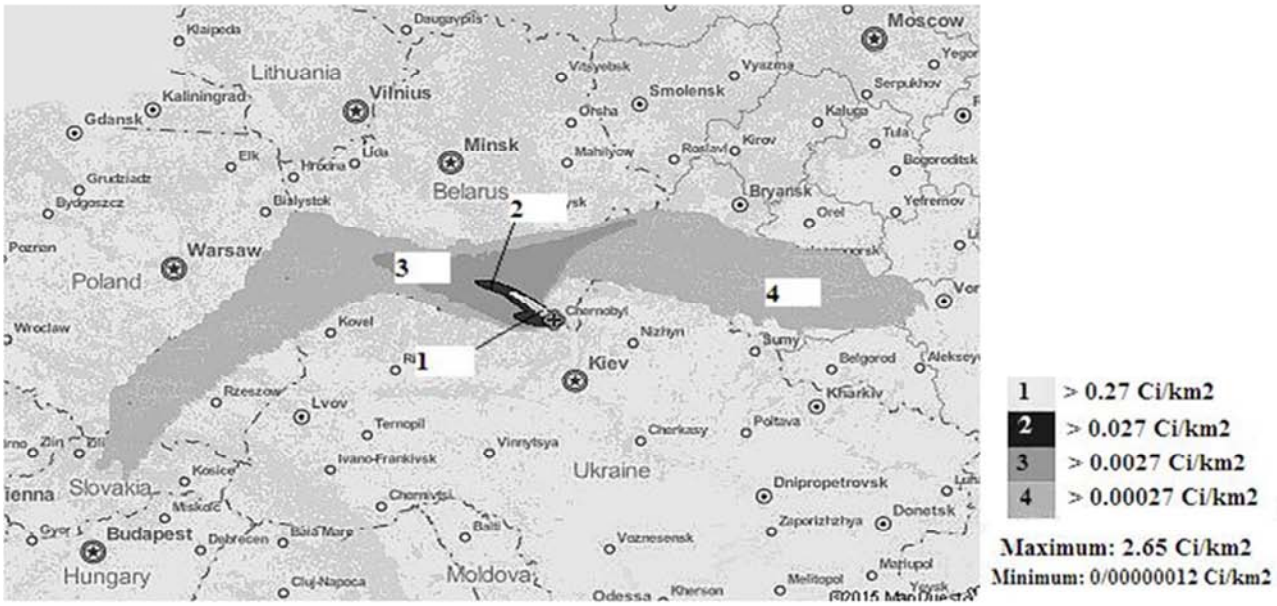

Figure 6. *Forecast map of ^{241}Am distribution at the time of maximum activity in 2059, Ci/km^2.*

Figure 6 gives the forecast model of ^{241}Am distribution at the time of maximum activity of the isotope in 2059, which will be approximately 0.191 PBq. The basic amount of the isotope is predicted to be concentrated in the 30 km zone around the ChNPP accident, and will be also found is cross-border regions of Belarus, Russia, Poland, Slovakia.

In consequence of the present studies we have constructed the maps of ^{241}Pu concentrations in air and soil in the territory of Ukraine in 1986. The forecast model has been constructed for ^{241}Am distribution at the time of the maximum isotope activity in 2059.

5. Conclusions

The radionuclide contamination with Pu isotopes and ^{241}Am inside Ukraine has resulted from global fallouts that stemmed from nuclear tests and Chernobyl NPP accident.

The ChNPP accident has led to nonuniform contamination with transuranium radionuclides, including Pu isotopes and ^{241}Am. The accumulation of ^{241}Am due to ^{241}Pu decay, the destruction of fuel particles, and the increased bioavailability of inert forms of radionuclides, all these factors give rise to migration of the mentioned radionuclides in the "soil-plant-human" chain.

The given results point to the necessity of further investigation of the data on the ^{241}Am accumulation and its effect on different ecosystems. This will permit the improvement of forecast models in emergency planning for protection of population health and rehabilitation of affected territories.

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