THE RADIOACTIVE CONTAMINATION TERRITORY OF UKRAINE BY Pu AND ^{241}Am RADIONUCLIDES DUE TO THE CHERNOBYL ACCIDENT

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(Received March 22, 2017)

The decay of β-emitting ^{241}Pu gives rise to long-lived α-radionuclides ^{241}Am and ^{237}Np, which are highly radiotoxic. The estimations of the ^{241}Pu, ^{241}Am and ^{237}Np content of the Chernobyl NPP accidental release have shown that by the present time nearly 90% of the ^{241}Pu isotope had already disintegrated to turn into ^{241}Am. This generates a need for developing the methods of analytical control and environmental monitoring of ^{241}Pu and ^{241}Am content. The present paper is concerned with estimation of radioactive contamination of the Ukrainian territory by ^{241}Pu and ^{241}Am isotopes as a result of the Chernobyl NPP accident.

PACS: 23.60.+e, 28.41.Kw, 28.41.Te

1. INTRODUCTION

The Chernobyl NPP accident has changed the radiological environment in the sizable areas of many European countries. Thus, for the first ten days, zones of higher radioactive contamination levels were formed. The existence of radioactive areas at distances over 50 km from the Chernobyl NPP is specified by a number of factors such as the emission of radioactive masses into the atmosphere to a height of up to 2000 m and more; falls of rains over the contaminated areas; the presence of complex landscapes which caused changes in the direction and height of the air-mass motion. The radionuclide release height has determined the global pollution pattern, while the rains and landscapes have specified spotty contamination of the areas [1].

Over the last years, large amounts of data on the processes of radionuclide migration in the environment have been amassed. Maps have been drawn to indicate the areas of Ukraine, which are contaminated with most human life-threatening radionuclides of the accident origin. That has become the base point of planning the actions for population health protection and rehabilitation of the contaminated areas. Over a period of 30 years after the accident it still remains currently central to estimate the transuranium elements (TUE) pollution of the Ukrainian area, to analyze the behavior of those elements and to determine the risk of TUE entry into the human body.

The Chernobyl radionuclide release into the environment has resulted in the contamination of a considerable part of the Ukrainian area with α-emitting radionuclides ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{241}Am having the half-lives $T_{1/2}$ = 87.7, 24065, 6537, 14.4 and 432.8 years, respectively. In view of an appreciable energy of the emitted α-particles, long effective half-lives, a high chemical toxicity, these radionuclides are qualified as the most dangerous when entered into the human organism.

By the present time, the primary forms of the TUE fallout have substantially transformed as a consequence of fuel particles damage. For this reason, the information on the current status and migration of the Pu and ^{241}Am isotopes is required. The goal of the present work has been to investigate the process of Pu and ^{241}Am isotope spreading over the Ukrainian area as a result of the Chernobyl accident.

2. THE OBJECT OF STUDY AND THE INPUT DATA

The work is concerned with the research information on the Chernobyl NPP accident and contamination of the Ukrainian area with Pu and ^{241}Am isotopes. The dynamics of radioactive contamination formation in the regions affected by the Chernobyl accident is commonly divided into four periods [2]:

- the 1st period (April-July 1986) – the radiological environment was mainly determined by the short-lived radionuclides: $^{131,133,135}I$, ^{140}La, ^{140}Ba, ^{99}Mo, ^{132}Te, ^{239}Np having the half-lives up to 1 year;

- the 2nd period (1986-1987) – along with the long-lived radionuclides, detectable amounts of the radionuclides ^{144}Ce, ^{106}Ru, ^{134}Cs, ^{242}Cm were registered;

- the 3rd period (after 1988) – the radiological environment was mainly specified by the radionuclides ^{137}Cs, ^{90}Sr, and in the exclusion zone – by the $^{238,239,240,241}Pu$, too;

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- the 4-th period – soil contamination will be governed by ^{241}Am and the radionuclides indicated for the 3-rd period.

The ^{241}Pu isotope is produced in nuclear reactors at neutron capture of ^{240}Pu. Unlike most of the Pu isotopes being accumulated in the irradiated fuel, the isotope ^{241}Pu is characterized by β-radiation. It has the highest specific activity as compared with other isotopes of transuranium element generated in the nuclear fuel [3]. For example, the total activity of ^{241}Pu in the Chernobyl NPP accidental release amounted to $5.52\,PBq$, that being 84% of the total reactor core-accumulated activity of the remaining TUE (Table 1). It should be noted that the radionuclide release to the atmosphere under the accident was assumed to be the same, viz., 3% [4].

Table 1. *Content of main long-lived radionuclides in the damaged reactor core [5]*

Radionuclide	Weight, kg	Activity, PBq	Activity TUE, %
^{90}Pr	44	228	-
^{134}Cs	3.2	153	-
^{137}Cs	81	260	-
^{238}Pu	1.5	0.94	0.43
^{239}Pu	413	0.95	0.43
^{240}Pu	176	1.5	0.683
^{241}Pu	49	184	84.126
^{242}Pu	14.2	0.0021	0.001
^{241}Am	1.1	0.14	0.064
^{243}Pu	0.73	0.0054	0.003
^{242}Cm	0.26	31	14.18
^{244}Pu	0.06	0.18	0.083

The radioactive-decay scheme of ^{241}Pu has the form:

$$^{241}Pu(T_{1/2} = 14.4\,years;\ \beta) \rightarrow$$
$$^{241}Am(T_{1/2} = 4.32 \cdot 10^2\,years;\ \alpha) \rightarrow$$
$$^{237}Np(T_{1/2} = 2.14 \cdot 10^6\,years;\ \alpha).$$

After entry of TUE into the environment, the β-decay of ^{241}Pu contributes to an increase of ^{241}Am content in various ecosystem objects. A further α-decay of ^{241}Am results in the production of α-emitting ^{237}Np, which will play an essential part in the formation of the radiological environment in the TUE contamination area. In this case, ^{241}Am and ^{237}Np act as alpha emitters and have a higher radiotoxicity than ^{241}Pu has. Fig.1 shows the total amount of different long-lived radionuclides in the environment as a function of the time since the accident.

The maximum ^{241}Am content of the environment will be reached in 2059 (i.e. within 73 years after the accident) and it will be 40 times higher than its accidental entry into the atmosphere in 1986 (Fig.2). The increase in the migration capability of ^{241}Am results from the damage of fuel particles. Unlike ^{241}Pu, the ^{241}Am compounds are more readily soluble, and thus, are more capable of migrating. Therefore, the now comparatively safe areas may become of serious hazard to habitation of people.

Plutonium migrates in the form of soluble organic complexes, nitrates, etc., which mostly (99%) settle in both the bottom deposits and the surface soil. The greatest concentration of Pu has been found to fall on short-grown plants, grass, mushrooms, mosses, lichens. Pu is considered to be one of the most hazardous substances. On entering into the biological environment, Pu gets involved in different biochemical cycles.

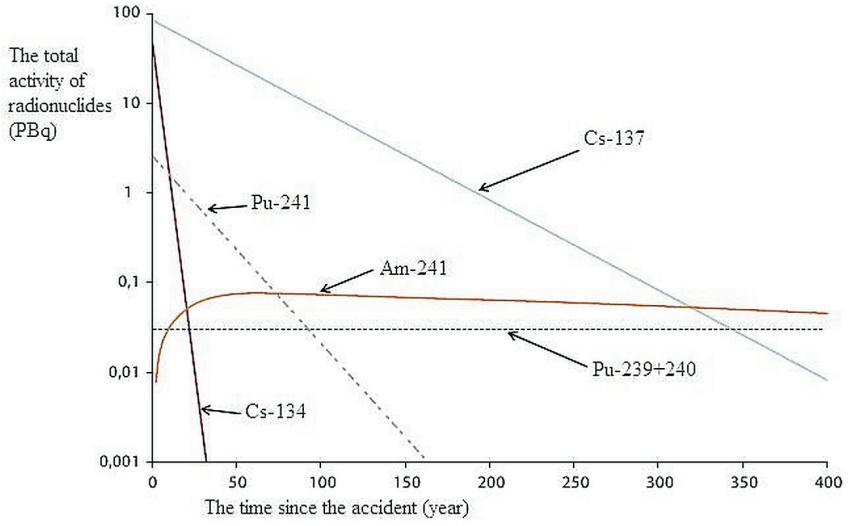

Fig.1. *Total amount of different long-lived radionuclides in the environment as a function of time since the accident [6]*

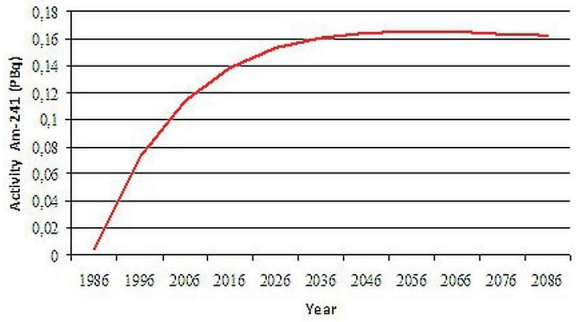

Fig.2. *Temporal dynamics of ^{241}Am activity build-up in the Ukrainian area after the Chernobyl NPP accident*

Plutonium may get into the human body as a result of inhalation, during taking food, or through skin integuments. Considering that Pu tends to accumulate in the top soil, it is obvious that a considerable amount of deposited Pu comes back to the atmosphere together with a dust. Getting into the lungs, Pu partially settles on the pulmonary surface, and partly, it goes to the blood. Then, moving, plutonium gets into the bone marrow and lymph nodes. The Pu clearance rate is such that 50 years after getting into the human body 80% of the adopted amount remains in the organism. If plutonium resides in the bone marrow, its biologic half-life makes between 80 and 100 years; so, the content of Pu remains practically unchanged. The biologic half-life of Pu in the liver reaches 40 years [7].

The Am may get into to the human organism through meals, water or inhaled air. The portion of Am that enters into the blood settles approximately in equal parts in the liver and in the skeleton, where it stays for long periods of time, the biologic half-life being about 20 and 50 years, respectively. The Environmental Protection Agency (EPA) has established the maximum contaminant level (MCL) for all α-active particles (except Ra and U) in the drinking water to be $15\,pCi/l$. This value also holds for ^{241}Am. The life cancerogenic risk factors have been calculated for nearly all radionuclides, including ^{241}Pu and ^{241}Am (Table 2).

Table 2. *Radiological risk factors*

Isotope	Life cancerogenic risk	
	Inhalation (pCi^{-1})	Food intake (pCi^{-1})
^{241}Am	$2.4 \cdot 10^{-8}$	$9.5 \cdot 10^{-11}$
^{241}Pu	$2.8 \cdot 10^{-10}$	$1.9 \cdot 10^{-12}$

Typically, food intake is the most common type of geting into to the human organism. However, the risk factor for this type of contaminant entry is much lower than that at inhalation [8].

Of grave concern for the human health are the diseases caused by the ionizing radiation of Pu and Am isotopes. Taking into account a long half-life of ^{241}Am, they pose a hazard to health for many generations of Ukrainians.

Thus, ^{241}Am is considered as one of the most important technogenic isotopes. The ^{241}Am decay products would exert a significant influence on the environmental contamination over a long many of years. Therefore, the currently central problem is to improve the efficiency of the analytical methods of determining ^{241}Am in different media.

3. THE DETECTION METHODS OF CONTAMINATION SOURCE

Nowadays, the receptor modeling (RM) technique described in refs. [9-11] appears to be the most efficient mathematical apparatus for determining the characteristics and localization of the contamination source. The technique includes two types of software products, namely, the PMF program intended for localizing the contaminant sources, and the HYSPLIT program [12]. Relying on the meteorological data, the last program makes it possible to reconstruct the trajectories of radionuclide propagation in the atmosphere and to obtain the true radionuclide distribution pattern for the area under study.

4. THE RESEARCH RESULTS

As regards the Chernobyl NPP accident, the main release of Pu and ^{241}Am isotopes took place on 26-27 April, 1986. This is confirmed by radiological environment monitoring after the accident. The radionuclides under discussion belong to the semi-volatile group, and their basic amount had appeared in the atmosphere mainly during the first phase of the accident, after a series of core explosions on 26.04.86. In the next days, the release of the mentioned radionuclides in the mixed stream (vapor-aerosol-gas flow) was caused by graphite burning, fuel dispersal increase after rise in the core temperature up to 2000°C and more, and also by formation of volatile compounds [1].

The processing of the data on Pu isotope release from the damaged reactor (see Table 1) has resulted in drawing the maps of ^{241}Pu concentration in air and Pu fallouts on the soil in the days of the main release. Fig.3 shows the map of ^{241}Pu concentration in air on 27 April, 1986. The Chernobyl emission plume was directed north-westwardly, and for that reason it was the areas of Ukraine and cross-border regions that were exposed to pollution.

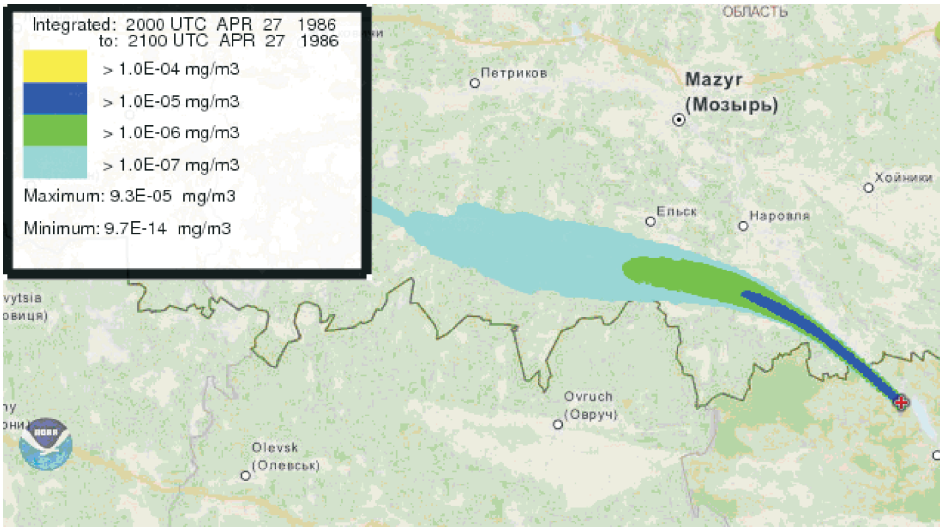

Fig.3. *^{241}Pu concentration in air on 27 April, 1986*

Fig.4 gives the map of soil contamination with ^{241}Pu fallout lasting from 27.04.1986 up to 30.04.1986 (84-hour duration). The total radioactivity of ^{241}Pu emission (3% of the reactor loading) has been estimated to be $5.52\,PBq$, the mass being $1.47\,kg$. The bulk of this isotope is concentrated in the $30-km$ zone of the Chernobyl NPP accident, also embracing partially the areas of Byelorussia, Poland and Russia. So, the ^{241}Am activity gradually grows due to the decay of ^{241}Pu. The extent of ^{241}Am propagation in the territory of Ukraine is comparable with that of Pu isotopes.

Fig.4. *^{241}Pu fallouts on the soil from 27.04.1986 to 30.04.1986*

5. CONCLUSIONS

Contamination of the Ukrainian area with ^{241}Pu and ^{241}Am radionuclides after the Chernobyl NPP accident has been estimated. The application of the RM technique for determining the contamination source has resulted in drawing the maps of ^{241}Pu isotope concentration in air and soil in the territory of Ukraine in 1986. The obtained information gives an insight into the accumulation, transformation and migration dynamics of ^{241}Pu and ^{241}Am isotopes, as also enables one to investigate the effect of ionizing radiation on the environment. The given data point to the necessity of further research on the buildup of ^{241}Am in the contaminated ecosystems.

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РАДИОАКТИВНОЕ ЗАРАЖЕНИЕ ТЕРРИТОРИИ УКРАИНЫ РАДИОНУКЛИДАМИ Pu И ^{241}Am В РЕЗУЛЬТАТЕ АВАРИИ НА ЧАЭС

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При распаде β-излучающего ^{241}Pu образуются долгоживущие α-радионуклиды ^{241}Am и ^{237}Np, которые обладают высокой радиотоксичностью. Расчеты содержания ^{241}Pu, ^{241}Am и ^{237}Np в аварийном выбросе ЧАЭС показали, что к настоящему времени около 90 % изотопа ^{241}Pu уже распалось и превратилось в ^{241}Am. Это определяет необходимость развития методов аналитического контроля и мониторинга содержания изотопов ^{241}Pu и ^{241}Am в объектах окружающей среды. В представленной работе произведена оценка радиоактивного загрязнения территории Украины изотопами ^{241}Pu и ^{241}Am в результате аварии на ЧАЭС.

РАДІОАКТИВНЕ ЗАРАЖЕННЯ ТЕРИТОРІЇ УКРАЇНИ РАДІОНУКЛІДАМИ Pu І ^{241}Am ВНАСЛІДОК АВАРІЇ НА ЧАЕС

М. Ф. Кожевнікова, В. В. Левенець, І. Л. Ролік, А. О. Щур

При розпаді ^{241}Pu, який випромінює β-частинки, утворюються довгоіснуючі α-радіонукліди ^{241}Am і ^{237}Np, що мають високу радіотоксичність. Розрахунки вмісту ^{241}Pu, ^{241}Am і ^{237}Np в аварійному викиді ЧАЕС показали, що до теперішнього часу близько 90 % ізотопу ^{241}Pu вже розпалося і перетворилося в ^{241}Am. Це визначає необхідність розвитку методів аналітичного контролю і моніторингу вмісту ізотопів ^{241}Pu і ^{241}Am в об'єктах навколишнього середовища. У представленій роботі зроблена оцінка радіоактивного забруднення території України ізотопами ^{241}Pu і ^{241}Am в результаті аварії на ЧАЕС.