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ScienceDirect

Nuclear Energy and Technology 3 (2017) 285–290

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Plutonium in locations of local sources and its involvement in global circulation

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Available online 15 November 2017

Abstract

This paper seeks to compare the volumetric activities of the $^{239+240}$Pu and ^{238}Pu isotopes in the surface layer of the atmosphere in the locations of different local sources of radioactive contamination, to characterize these sources as applied to the ratio of the ^{238}Pu and $^{239+240}$Pu isotopes, and to estimate the global industry-caused background of plutonium isotopes in the surface atmosphere. Under investigation are an NPP location (the city of Kursk), a territory contaminated as the result of the accident at the Chernobyl NPP (the city of Bryansk), the location of a radiochemical site for the radioactive material reprocessing (PA Mayak in the town of Novogorny, Chelyabinsk Oblast), and the city of Obninsk, the location of nuclear research reactors. The dynamics of the volumetric activity in the locations of interest in 1992–2015 has been reviewed, and the most contaminated areas and the areas with the smallest content of Pu isotopes in the atmosphere's surface layer have been identified. Causes have been revealed for variations in the volumetric activity levels by years and seasonally. The sources of radioactive contamination under consideration have been characterized in terms of the ^{238}Pu–$^{239+240}$Pu ratio, and the possibility for this ratio to be used to identify the release sources has been evaluated.

A much smaller degree of the plutonium isotope involvement in global circulation has been shown based on results of a dedicated study into the volumetric activity of plutonium isotopes at observation points in the Russian subpolar and polar areas, the most distant ones from local sources of the atmospheric Pu release.

Throughout the considered period, the volumetric activity of plutonium isotopes in all of the locations of interest was not exceeding the permissible volumetric activity in the inhaled air for the population, as specified in Radiation Safety Regulations NRB-99/2009, which is equal to 2.5×10^{-3} Bq/m^3.

Keywords: Monitoring; Volumetric activity; Plutonium; Isotope; Atmosphere surface layer; NPP; Radioactive contamination; Radiochemical site.

Introduction

Plutonium in the environment is considered as a man-made element. Natural ^{239}Pu forms in negligible quantities in the process of Uranium fission in Uranium ore at a level of $5 \cdot 10^{-6}$ g of plutonium per 1 ton of Uranium. In all other cases, it has an industry-related origin [1–3].

The requirement for monitoring the plutonium content in the environment is dictated by the fact that it defines to a great extent the long-term consequences of radioactive contamination [4].

Most of the plutonium was released into the environment as a result of nuclear explosions in the atmosphere which were stopped in 1980 (the last nuclear explosion in the atmosphere was conducted in China on 18 October 1980). The total plutonium activity in nature is estimated at 14,600 TBq of which 13,000 TBq is the contribution from the nuclear weapon

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Peer-review under responsibility of National Research Nuclear University MEPhI (Moscow Engineering Physics Institute).

Russian text published: Izvestiya vuzov. Yadernaya Energetika (ISSN 0204–3327), 2017, n.2, pp. 145–156.

https://doi.org/10.1016/j.nucet.2017.11.002

testing. The rest is also basically of an industry-related origin [1].

As compared to other radionuclides, plutonium isotopes are removed from the atmospheric air fairly quickly. The concentration of $^{239+240}$Pu in the air decreases by half for 1.2 years (the "half-cleaning period"), as the ^{137}Cs concentration decreases by half for three years on the average [5].

The maximum annual $^{239+240}$Pu fallout was observed in 1963 after large-scale nuclear tests in 1961–1962 [6]. Following the end of the nuclear tests in the atmosphere, plutonium, for several more years, continued to fall out from the stratospheric reservoir it had filled up in the course of the tests. By the spring of 1984, most of the stratospheric plutonium had fallen out onto the earth surface and its contribution has decreased since 1985, at least, to the level of the resuspension from the US nuclear test sites [7]. It is exactly the wind resuspension and natural fires that are considered the major processes contributing to the plutonium entry into the surface layer of the atmosphere in the period of atmospheric nuclear tests [8,9].

Since 1984, the volumetric activity of plutonium in the surface layer of the atmosphere is defined by the release from local sources of contamination and by resuspension from the surface in the territories contaminated in the course of ground-based nuclear tests and as a result of nuclear accidents [7]. Based on a generalization of the extensive material obtained after 1984 by the observation stations of the US Radiation Monitoring Network, as well as in European countries and Japan [7,10–12], it was found that the normal value of the global $^{239+240}$Pu background in the surface atmosphere was in the limits of $10{\cdot}10^{-9}$ to $100{\cdot}10^{-9}$ Bq/m^3 [10,11].

Wind resuspension is most effective in dry regions with an unstable vegetation cover. So, in Astana, situated 500 km west of the Semipalatinsk test range, the Soviet Union's long-term nuclear weapon atmospheric test site, the activity of ^{238}Pu and $^{239+240}$Pu in the surface air was recorded in 2000 and 2001 at the level of $9{\cdot}10^{-9}$ and $29{\cdot}10^{-9}$ Bq/m^3 respectively. In Kurchatov, a city in the immediate vicinity of the Semipalatinsk test site's contaminated territories, the average activities of these radionuclides are much higher: $34{\cdot}10^{-9}$ and $100{\cdot}10^{-9}$ Bq/m^3 respectively [13].

The studies conducted in Spain based on the material from 2001–2002 showed that the activity of $^{239+240}$Pu in the surface air varied in a range of (1–20)·10–9 Bq/m^3 with the maximums recorded in the summer period. Further studies on the composition of dust in aerosol samples made it possible to suggest that the summertime maximums had been caused by the transport of dust from the Sahara [14,15].

In Russia, local sources of the plutonium entering the environment include radiochemical sites, nuclear power plants, nonproduction nuclear facilities for various applications, and the territories contaminated by nuclear accidents [16]. Considered hereinafter are the levels and the peculiarities of the surface air pollution with ^{238}Pu and $^{239+240}$Pu in such locations as the PA Mayak radiochemical site in Chelyabinsk Oblast, nonproduction nuclear reactor sites at the Institute for Physics and Power Engineering (IPPE) and a branch of the

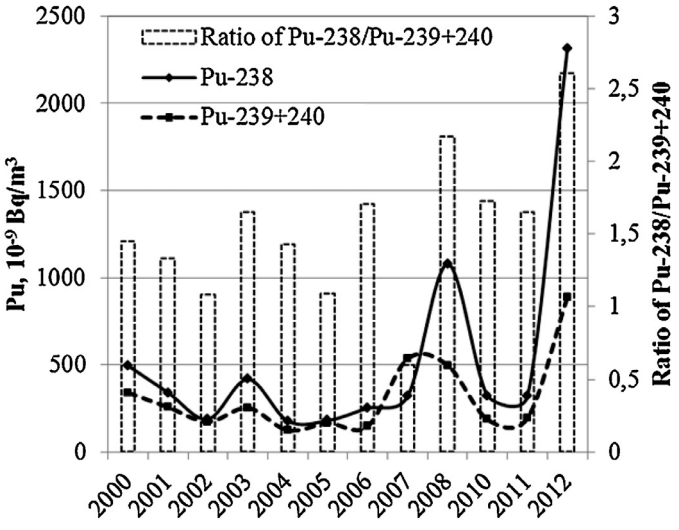

Fig. 1. Average annual volumetric activity of ^{238}Pu and $^{239+240}$Pu in the atmospheric air in Novogorny (PA Mayak) and the ratio of ^{238}Pu/$^{239+240}$Pu in 2000–2012.

L.Ya. Karpov Institute for Physical Chemistry (IPC) in Obninsk, Kursk NPP, and the cities of Kursk and Bryansk being under the influence of wind resuspension from the Chernobyl-contaminated territories.

The error of the ^{238}Pu determination is 15–45% and that of the $^{239+240}$Pu determination is 20–32%. Each local source of plutonium isotopes has its own ratio of ^{238}Pu/$^{239+240}$Pu which makes it possible to identify the contamination source [17]. Therefore, this ratio has been calculated and shown for all local sources of plutonium along with their absolute activity values.

Radiochemical sites

At the present time, radiochemical sites have become the major source of the plutonium entering the environment. The oldest of these is PA Mayak in Chelyabinsk Oblast with a history of activities for over 60 years. The annual average values of the volumetric activity of plutonium isotopes in the air (town of Novogorny) vary in a broad range: $190{\cdot}10^{-9}$ to $2300{\cdot}10^{-9}$ Bq/m^3 for ^{238}Pu, and $120{\cdot}10^{-9}$ to $890{\cdot}10^{-9}$ Bq/m^3 for $^{239+240}$Pu. The ratio of the average annual volumetric activity of ^{238}Pu and $^{239+240}$Pu in the atmospheric air in the period under consideration is from 0.6 in 2007 to 2.6 in 2012 (Fig. 1).

High monthly average volumetric activity values for $^{239+240}$Pu in the surface air during the year are randomly distributed (Fig. 2). The highest values were recorded in 2000 (June through July), in 2008 (February), and in 2012 (April through May), while there were no major deviations from the annual average level in 2004.

Nonproduction nuclear reactors

Much lower levels of the volumetric activity of plutonium isotopes (by two or three orders of magnitude) are observed