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Plutonium isotopes in the atmosphere of Central Europe: Isotopic composition and time evolution vs. circulation factors

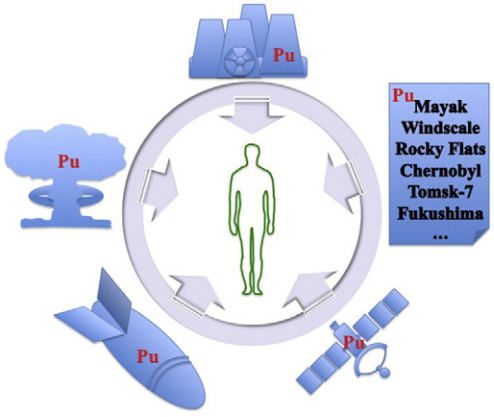
Renata Kierepko [a,*], Jerzy W. Mietelski [a], Zbigniew Ustrnul [b,c], Robert Anczkiewicz [d], Herbert Wershofen [e], Zoltan Holgye [f], Jacek Kapała [g], Krzysztof Isajenko [h]

[a] Institute of Nuclear Physics, Polish Academy of Sciences, Krakow, Poland
[b] Jagiellonian University, Krakow, Poland
[c] Institute of Meteorology and Water Management, National Research Institute, Krakow, Poland
[d] Institute of Geological Sciences, Polish Academy of Sciences, Krakow, Poland
[e] Physikalisch-Technische Bundesanstalt, Braunschweig, Germany
[f] National Radiation Protection Institute, Prague, Czech Republic
[g] Medical University of Bialystok, Poland
[h] Central Laboratory for Radiological Protection, Warsaw, Poland

HIGHLIGHTS

- Evidence of Pu isotopes in the lower part of the troposphere of Central Europe
- The effective annual doses associated with Pu inhalation
- New approach to the problem of solving mixed Pu origins in one sample (3SM)
- Relationship between Pu isotopes activity concentration and circulation factors

GRAPHICAL ABSTRACT



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ABSTRACT

This paper reports evidence of Pu isotopes in the lower part of the troposphere of Central Europe. The data were obtained based on atmospheric aerosol fraction samples collected from four places in three countries (participating in the informal European network known as the Ring of Five (Ro5)) forming a cell with a surface area of about 200,000 km^2. We compared our original data sets from Krakow (Poland, 1990–2007) and Bialystok (Poland, 1991–2007) with the results from two other locations, Prague (Czech Republic; 1997–2004) and Braunschweig (Germany; 1990–2003) to find time evolution of the Pu isotopes. The levels of the activity concentration for ^{238}Pu and for $^{(239+240)}$Pu were estimated to be a few and some tens of nBq m^{-3}, respectively. However, we also noted some results were much higher (even about 70 times higher) than the average concentration of ^{238}Pu in the atmosphere. The achieved complex data sets were used to test a new approach to the problem of solving mixing isotopic traces from various sources (here up to three) in one sample. Results of our model, supported by

* Corresponding author.
E-mail address: Renata.Kierepko@ifj.edu.pl (R. Kierepko).

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Atmospheric circulation parameters
Plutonium effective dose

mesoscale atmospheric circulation parameters, suggest that Pu from nuclear weapon accidents or tests and nuclear burnt-up fuel are present in the air.

1. Introduction

On July 16, 1945 the first nuclear weapon test was conducted in New Mexico (USA). Since then, development of nuclear technologies for weapons and peaceful uses has been continuous, and in tandem, the number of nuclear events (intentional or accidental) has been rising. Fission products released into the atmosphere have caused widespread distribution of artificial radionuclides worldwide. Most countries have developed monitoring methodologies of airborne nuclides, in particular, gamma radionuclides. The determination of alpha and beta emitters, however, requires more sophisticated research methods.

One of the main issues in our research was to determine Pu isotope composition (^{238}Pu, $T_{1/2} = 87.8$ years; ^{239}Pu, $T_{1/2} = 24110$ years; ^{240}Pu, $T_{1/2} = 6563$ years) in the ground level air (in the lower part of the troposphere) that could have originated from the following sources.

- Nuclear weapon tests: over 500 atmospheric explosions (CTBTO, 2016) have been carried out in the open atmosphere as a part of over 2000 nuclear weapon tests in the past. Atmospheric explosions were conducted at many locations like Novaya Zemlya, Semipalatinsk, Bikini Atoll, Sahara, New Caledonia, Nevada, and Lob Nor which resulted in global fallout.
- Nuclear accidents: these have occurred at weapons-related sites, Mayak (near Kyshtym, 1957 and Lake Karchay, 1968), Rocky Flats (1969), and Tomsk –7 (1993), and at non-weapons-related sites, Windscale (1957), Chernobyl (1986), and Fukushima (2011). It is still not clear, however, if during the Fukushima accident Pu was released to the environment over distances larger than just within Japan (Schneider et al., 2013).
- Disintegration of satellites; these include SNAP 9A (launched by the USA, 1964) and Cosmos 958 (launched by the former USSR, 1978).
- Crashes of nuclear weapon-equipped military aircraft; these include the crashes at Palomares, Spain (1966) and Thule, Greenland (1968).
- Nuclear industries: these include the production of nuclear weapons at e.g. Mayak (Russia) and Hanford (USA); and non-weapons related industries, including the production of nuclear fuel at e.g.: Marcoule and Romans (France), Lingen (Germany), Ust Kamenogorsk (Kazakhstan), and Richland and Wilmington (USA); and reprocessing of nuclear fuel at locations like Sellafield (UK) or La Hague (France).

Each source of Pu is characterized by its unique isotopic ratios between ^{238}Pu, ^{239}Pu and ^{240}Pu (Table 1). These specific isotopic fingerprints of trace amounts of nuclear materials depend on the composition of the source material and subsequent irradiation history.

At present, the Pu activity in the air is almost completely controlled by resuspension (Lehto et al., 2006) and sea spray (Kierepko et al., 2009). Eroded or remobilized particles of soil or marine aerosols could be transported over long distances with air masses from the deposition places. The study of the determination of Pu in the air is of high interest and of great importance for two reasons. Firstly, Pu isotopes are useful as tracers of environmental processes (e.g.: mixing, re-distribution and transport of atmospheric aerosols (Alvarado et al., 2014; Cambray and Eakins, 1982; Hirose and Povinec, 2015), resuspension (Hirose et al., 2003; Hirose et al., 2004; Hölgye, 2008; Montero and Sanchez, 2001; Wershofen and Arnold, 2005), etc.). Secondly, Pu, which is recognized as one of the most radiotoxic elements, may be directly inhaled with aerosols and accumulated in plants and animals at some rate. Moreover, knowledge of the local background values of Pu concentrations in air including its isotopic composition could be helpful in the case of any future releases of nuclear materials. With today's nuclear renaissance, the risk of nuclear terrorism, and undeclared nuclear activities to be considered, events such as the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident have shown that samples, where the concentration of Pu isotopes is extremely low, in the femtogram range per sample (Zheng et al., 2012), may provide important information in various fields.

Our main purpose in this paper is to present a way to identify Pu sources in the air based on the activity and mass ratios of ^{238}Pu, ^{239}Pu and ^{240}Pu.

2. Material and methods

2.1. Sampling

In order to understand the time evolution of the activity concentration of Pu isotopes in the air and to trace their source based on the Pu isotopic composition, we followed the experience of a German research group from Physikalisch-Technische Bundesanstalt in Braunschweig (Arnold et al., 1999; Wershofen and Arnold, 2005) and used air dust loaded filters as the information base for our study. The aerosol samples were obtained using three high efficiency aerosol sampling stations of the type ASS-500 (Aerosol Sampling Station, 500 m^3/h nominal average weekly flow rate, suitable for continuous operation under different weather conditions). Two of them are part of a Polish state monitoring network of twelve stations located throughout Poland that monitor gamma-ray emitters in ground-level air. The network is coordinated by the Central Laboratory of Radiological Protection in Warsaw and the National Atomic Energy Agency of Poland.

Table 1
The literature values of the activity ratio (A) and the mass ratio (m) of ^{238}Pu, ^{239}Pu and ^{240}Pu for selected types of environmental Pu sources.

Pu source	$A^{238}Pu/A^{(239+240)}Pu$	$m^{240}Pu/m\ ^{239}Pu$	$A^{238}Pu/A^{239}Pu$
Nuclear weapons		0.01–0.07 (Nassef et al., 2008)	
Palomares accident		0.056 (Mitchell et al., 1997)	0.0275 (Mitchell et al., 1997)
Thule accident	0.014 (Dahlgaard et al., 2001)	0.033 (Mitchell et al., 1997) 0.045 (Dahlgaard et al., 2001)	0.0150 (Mitchell et al., 1997)
Global fallout + SNAP 9A (Northern Hemisphere)	0.03 (UNSCEAR, 1982)	0.018 (Nassef et al., 2008)	
Burnt-up fuel	0.18 (Mitchell et al., 1995) −>1.8 (Skipperud et al., 2008)	0.23 (Nassef et al., 2008) 1 (Skipperud et al., 2008)	
Chernobyl accident	0.55 (Mietelski, 2003) 0.6 (Pietruszewski and Bojanowski, 1990)	0.4 (Nassef et al., 2008)	
Sellafield	0.18–0.21 (Mitchell et al., 1995)	0.25 (Kershaw et al., 1995)	
La Hague	0.26–0.34 (Skipperud et al., 2008)	0.36 (Skipperud et al., 2008)	

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