

Cs, Am and Pu isotopes in the atmosphere: source terms and tracer studies

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Anthropogenic radionuclides introduced into the atmosphere from various sources have been applied as tools for investigation of contaminant behavior and transport phenomena in the atmosphere. Recent developments in analytical techniques such as gamma-spectrometry and mass spectrometry of long-lived radionuclides have enabled to study atmospheric processes and transport of Saharan, North America and China desert dust. Specifically Pu isotopes have been representing a new tool for investigation of atmospheric processes in Europe, North America and in Asia.

¹³⁷Cs, ²⁴¹Am and Pu isotopes in aerosol samples collected at Preila background and Vilnius sampling stations during 1993–2011 were analyzed with special emphasis on better understanding of different sources with characteristic signatures observed at a given site. ^{239,240}Pu monthly activity concentrations in Vilnius ranged from 0.9 to 300 nBq/m³ (mean value of 13.4 nBq/m³) and well correlated with amount of total suspended particles, mineral dust and Al concentrations, however, no correlation with PM₁₀ was found. The highest ^{239,240}Pu activity within the 1995–2011 record with the ²⁴⁰Pu/²³⁹Pu atom ratio close to the Chernobyl value was found in 1995, and it was attributed to the transport of “hot” particles from the areas contaminated after the Chernobyl accident. The ¹³⁷Cs/^{239,240}Pu ratios varied from 80 to 2500.

The results from long-term measurements of ²⁴⁰Pu/²³⁹Pu atom ratios showed a bimodal frequency distribution with median values of 0.195 and 0.253, indicating two main sources contributing to the Pu activities at the Vilnius sampling station (Lujanienė et al., 2012 a). The first mode with the median value of 0.195 (from 0.155 to 0.215) corresponds to the global fallout ratio of ~0.17–0.19 characteristic for the Northern Hemisphere (Kelley et al., 1999). The second mode showed the median value of 0.253 (from 0.225 to 0.285) which could be attributed to the nuclear reactor Pu with characteristic ²⁴⁰Pu/²³⁹Pu atom ratio of 0.23–0.67 (Warneke et al., 2002).

Three-day backward trajectories of air-masses for 4 selected sectors for three arrival heights: 20, 500 and 1000 m AGL were modeled to assess the transport of aerosol particles to the Vilnius sampling station in 1997–2001 and 2005–2006. A weak correlation between the ¹³⁷Cs activity concentration in daily samples and height ($r = 0.28$ (20 m), 0.32 (500 m), and 0.31 (1000 m) in 1997–2001, and for 2005–2006 ($r = 0.41$ (20 m), 0.49 (500 m), and 0.49 (1000 m)) was found for the Chernobyl sector, while for other sectors no correlation was observed (Lujanienė et al., 2012 b).



Figure 1. An example of air-mass backward trajectories ending at Vilnius sampling site.

The ²⁴⁰Pu/²³⁹Pu ratio in monthly samples and corresponding trajectories ($n = 1069$) were analyzed. A weak negative correlation (-0.37) of the ²⁴⁰Pu/²³⁹Pu atom ratio with transport direction was found for the Siberian region. By contrast to the North–East sector (Figure 1), the ²⁴⁰Pu/²³⁹Pu ratio showed a weak positive correlation of 0.46 (significance level of correlation $p > 0.99$) with the number of trajectories arriving from the Arctic and the Baltic region. This implies that particles with higher ²⁴⁰Pu/²³⁹Pu ratios (up to 0.295) tend to be transported from the North–West while no correlation with the Chernobyl region was observed. Measurements carried out during the Fukushima accident showed a negligible impact of this source with Pu activities by four orders of magnitude lower as compared to the Chernobyl accident (Lujanienė et al., 2012 a, b). In spite of the low detected activities, these observations are important for tracer studies for the assessment of transport of various pollutants, for investigation of atmospheric circulation and validation of existing models for future use.

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