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Applications of ^{137}Cs and Pu isotopes in tracer studies

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This study was performed to assess activity concentrations of radionuclides and characteristic Pu activity and atom ratios in the atmosphere, soil, suspended particulate matter and bottom sediments with the aim of their possible application to trace the pollutants in the environment. Analyses of airborne radioactive aerosols were carried out in daily samples collected in Vilnius as well as in 10-day samples at the background station in Preila (at the Baltic Sea shore, Lithuania). ^{137}Cs , ^{241}Am and Pu isotopes were also analyzed in soil, seawater, bottom sediments (BS) and suspended particulate matter (SPM) samples collected in the Baltic Sea and the Curonian Lagoon during 1997–2013.

The observed $^{238}\text{Pu}/^{239,240}\text{Pu}$ activity ratios ranged from 0.44 to 0.50, while the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio varied after the Chernobyl accident. The $^{240}\text{Pu}/^{239}\text{Pu}$ ratio in aerosol in 1995 – 1999 varied from 0.14 to 0.40 whereas in Preila the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio varied from 0.135 to 0.247. The exponential decrease in the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio from 0.30 to 0.19 observed from 1995 to 2003 was explained by a decrease in the amount of the Chernobyl derived plutonium in the environment. The analyses of frequency count of the Pu atom ratio ($N=74$) indicated that Pu originated from at least two different sources. Activity $^{238}\text{Pu}/^{239,240}\text{Pu}$ and atom $^{240}\text{Pu}/^{239}\text{Pu}$ ratios indicated a different contribution of the Chernobyl-originated Pu to the suspended particulate matter (SPM) and bottom sediments. The largest amount of the Chernobyl-derived Pu was found in the smallest suspended matter particles of 0.2 – 1 μm in size collected in the Klaipeda Strait in 2011 – 2012. The decrease of characteristic activity $^{238}\text{Pu}/^{239,240}\text{Pu}$ and atom $^{240}\text{Pu}/^{239}\text{Pu}$ ratios towards the global fallout ones in surface soil and the corresponding increase of plutonium ratios in the suspended particulate matter and bottom sediments have indicated that the Chernobyl-derived Pu, primarily deposited on the soil surface, was washed out and transported to the Baltic Sea.

The obtained results indicated that although the main source of Pu in the Baltic Sea environment was considered to originate from the global fallout after atmospheric nuclear weapons tests, the recently observed inflow of the Chernobyl-derived Pu to the Baltic Sea is linked to the contamination of soil surface after the accident. This secondary source of Pu was attributed to the dissolution of particles deposited on the soil surface and transport of Pu isotopes by water flow. Contrary to Pu isotopes data on activity concentrations of ^{137}Cs in the bottom sediments indicated its redistribution in the Curonian Lagoon – the Baltic Sea system that points to certain limitations in the application of ^{137}Cs in the tracer studies.

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