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# Transport of $^{137}Cs$, $^{241}Am$ and Pu isotopes in the Curonian Lagoon and the Baltic Sea

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ABSTRACT

Activities of $^{137}Cs$, $^{241}Am$ and $^{239,240}Pu$ were analyzed with special emphasis on better understanding of radionuclide transport from land via the Neman River estuaries to the Baltic Sea and behavior in the marine environment. Although activity concentrations of $^{137}Cs$ in water samples collected the Baltic Sea were almost 100 times higher as compared to the Curonian Lagoon, its activities in the bottom sediments were found to be comparable. Activity $^{238}Pu/^{239,240}Pu$ and atom $^{240}Pu/^{239}Pu$ 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 $^{238}Pu/^{239,240}Pu$ and atom $^{240}Pu/^{239}Pu$ ratios towards the global fallout ones in surface soil and the corresponding increase of plutonium (Pu) 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. Behavior of $^{241}Am$ was found to be similar to that of Pu isotopes.


## 1. Introduction

The anthropogenic radionuclides were mainly introduced into the Baltic Sea due to the atmospheric deposition after the nuclear weapons tests in the 1950's and the 1960's and during the Chernobyl nuclear power plant (NPP) accident in 1986. Other sources such as discharges from NPPs and the fuel reprocessing have injected relatively smaller amounts of the radionuclides into the Baltic Sea environment (Livingston and Povinec, 2000, 2002).

The total amount of the Chernobyl-derived $^{137}Cs$ in the Baltic Sea was estimated at about 4700 TBq, while that of the global fallout from nuclear weapons tests was found to be 900 TBq (HELCOM, 1995). The radioactive contamination of the Baltic Sea was mainly studied in relation to the high $^{137}Cs$ activity concentrations persisting for a long time in water after the Chernobyl NPP accident (La Rosa et al., 2001; Knapinska-Skiba et al., 2001; Livingston and Povinec, 2002; Zalewska and Lipska, 2006; Ilus, 2007). The average activity concentration of $^{137}Cs$ in the surface water of the Baltic Sea was estimated to be about 60 Bq/m$^3$, while the worldwide average concentration due to the global fallout is about 2 Bq/m$^3$ (Livingston and Povinec, 2000). The historical record of the contamination of the Baltic Sea by anthropogenic radionuclides can be found in the IAEA GLOMARD/MARIS database (Povinec et al., 2004; http://maris.iaea.org). The average activity concentration of $^{239,240}Pu$ in the Baltic Sea water for the year 2000 was reported to be ~1.5 μBq/L. However, large areas of the Earth's surface contaminated by radioactive substances after the Chernobyl accident still remain as potential sources of radionuclides (De Cort et al., 1998). The estimated fluxes of $^{137}Cs$ into Gdansk Basin in 1993–1994 comprised 0.02 TBq/year from the atmospheric fallout, and 0.1 TBq/year from the River Vistula (Knapinska-Skiba et al., 2001).

In the Chernobyl plume, a wide spectrum of radionuclides including "hot particles" containing alpha-emitters was found. Although fuel particles containing Pu isotopes were mainly deposited in the Ukraine and Belarus, they were detected in many European countries including Lithuania (Lujanienė et al., 2009b). It has been shown that, unlike the deposition of transuranium elements from the weapons tests in the 1950's and the 1960's, the $^{239,240}Pu$ fallout from the Chernobyl accident was typically very unevenly distributed (Paatero et al., 2002). Plutonium activity and atom ratios were used to distinguish between the Chernobyl and

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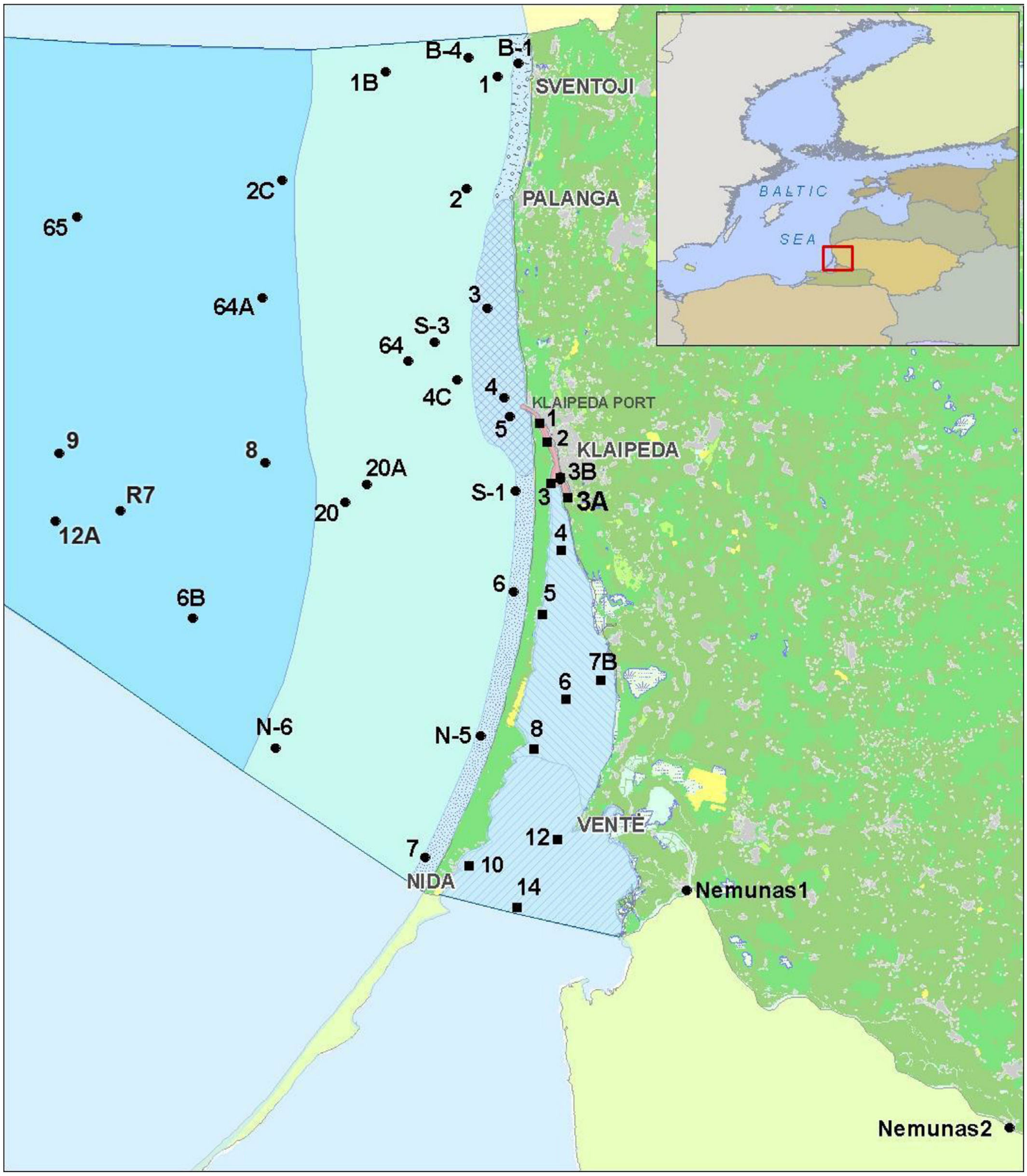


**Fig. 1.** Sampling locations in the Baltic Sea.

stratospheric fallout. The activity $^{238}Pu/^{239,240}Pu$ and $^{240}Pu/^{239}Pu$ atom ratios were about 0.45 and 0.40 in the Chernobyl fallout and 0.03 and 0.18 in the stratospheric fallout, respectively (UNSCEAR, 1982; Livingston and Povinec, 2002).

Following the Chernobyl accident, the activity concentrations of $^{239,240}Pu$ in Lithuanian soil ranged from 0.05 to 1.30 Bq/kg dry weight and the $^{238}Pu/^{239,240}Pu$ activity ratio was reported to be 0.3–0.45 (Druteikienė, 1999). Slightly higher $^{239,240}Pu$ activities were found in Polish soil while the $^{238}Pu/^{239,240}Pu$ activity ratios (0.04–0.46) were close to those observed in Lithuania. According to the mass spectrometric data (Povinec, 2005), the contribution of the Chernobyl-derived Pu to the total activities in soil was (40–62) % and (58–96) % in southern and in northeastern regions of Poland, respectively (Ketterer et al., 2004). It was assumed that the different source term was responsible for the faster migration of the Chernobyl-derived Pu as compared to the global fallout (Matisoff et al., 2011). The higher mobility of the Chernobyl-derived Pu can be explained by data obtained inside the restricted Chernobyl zone where dissolution of the released fuel particles was found and the distribution of Pu isotopes over the surface soil layer with a low infiltration capacity was attributed to Pu affinity for soil organics, most probably for humic substances (Matsunaga and Nagao, 2009). It has been reported that organics can facilitate Pu migration through the formation of Pu complexes with dissolved organic matter (DOM) in neutral to alkaline waters (Zhao et al., 2011). Thus, the dissolution of deposited fuel particles and transport of Pu isotopes to water bodies are possible. Estimations made by Polish researchers indicated that the inflow of nearly 250 rivers into the Baltic Sea can contribute 500 km$^3$ fresh water and 2 GBq $^{239,240}Pu$ per year (Skwarzec et al., 2003). The Pu input via rivers to Gdansk Basin of the Baltic Sea was found to be 312 MBq/year whereas wet + dry depositions amounted to only 126 Bq/year.

The level of present activities in the Baltic Sea is rather low. However, the investigation of the migration behavior of radionuclides is important for future estimations and from the point of view of their possible application in tracer studies of various