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Plutonium isotopes ^{238}Pu, $^{239+240}$Pu and ^{241}Pu in the Baltic Sea ecosystem

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Abstract In the paper there are presented the results of plutonium ^{238}Pu, $^{239+240}$Pu and ^{241}Pu determination in different samples of the Baltic Sea environment. ^{238}Pu and $^{239+240}$Pu isotopes were measured in alpha spectrometer and ^{241}Pu results were calculated indirectly by activity measurements of ^{241}Am isotope increase from β-emitting ^{241}Pu in 16–18 years after Chernobyl accident. The results indicate that the Baltic organisms accumulate plutonium from environment and the bioconcentration factors (BCF) range from 100 for fish to 27,000 for priapulida (benthic organisms) and only 0.1% of total plutonium is deposited in living organisms. Almost whole plutonium is deposited in Baltic sediments (about 99%). The results of ^{241}Pu determination indicate that after Chernobyl accident indicate on its high concentration in analyzed components of the Baltic Sea ecosystem and the highest value of the activity ratios ^{241}Pu/$^{239+240}$Pu were found in seawater (140).

Key words plutonium • ^{238}Pu • $^{239+240}$Pu • ^{241}Pu • Baltic Sea • Chernobyl accident

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Introduction

Wet and dry atmospheric fallout from nuclear weapon tests is one of the most important sources of plutonium in the Baltic Sea. The other sources: plutonium releases from spent fuel facilities in Sellafield (UK) and Cap de la Hague (France) are less important. Since 26 April 1986 it has been a new source of plutonium – Chernobyl plutonium, which should be taken under note in estimation of its radiological effects on the environment [2].

Experiments on plutonium sources in Gdańsk Bay and Gdańsk Basin indicated the main source of these radionuclides is rivers (mainly Vistula and Nemen) inflows which enrich these regions in 78% of its total content [6].

Marine plants and animals are capable to accumulate radionuclides from environment. That is why it is very important to recognize the impact of radionuclides on living organisms and possible transfer to human body through feeding way. In marine ecosystem the highest plutonium concentrations were found in sediments, but complex biogeochemical cycle of this element causes it is present in all environment compartments. The results of experiments indicate increasing of plutonium bioavailability in marine food chain of the southern Baltic Sea as a effect of its desorption from sediments and accumu-lation in benthic organisms [2–5].

This work presents results of determination of the most important α-emitting plutonium isotopes (^{238}Pu and $^{239+240}$Pu) in Baltic seawater, organisms and sediments samples and less known in the Baltic Sea environment β-emitting ^{241}Pu, the result of Chernobyl accident. β-emitting ^{241}Pu is less important according to its radiotoxicity than α-emitting plutonium isotopes (^{238}Pu, ^{239}Pu, ^{240}Pu), but is quite essential because of the highest contribution in the whole plutonium fallout and quite short lifetime (14.9 years) decaying to very highly radiotoxic, long-living, α-emitting americium ^{241}Am ($t_{1/2}$ = 432.7 years) [1].

Materials and methods

Plutonium ^{238}Pu and $^{239+240}Pu$ concentrations were measured using alpha spectrometer. All alpha spectrometric sources were prepared using electrolysis on a stainless steel disc after purifying on anion exchange resin with ^{242}Pu as a yield tracer [2, 3].

The determination of ^{241}Pu in the samples was done indirectly by measurements of ^{241}Am increase from β-emitting ^{241}Pu. The obtained alpha plutonium spectra were compared with the same spectra obtained 16–18 years earlier [2]. A comparison of the obtained spectra allowed the estimation of the ^{241}Pu content based on the increase of the 5.49 MeV peak of ^{241}Am, taking into account the ^{238}Pu present in the samples from the Chernobyl accident. The calculation of the ^{241}Pu activity was based on the following formula from:

$$A_{Pu_0} = 31.3074 \frac{A_{^{241}Pu} \cdot e^{+\lambda_{Am} \cdot t}}{\left(1 - e^{-\lambda_{Pu} \cdot t}\right)} \quad (1)$$

where A_{Pu_0} – ^{241}Pu activity at the time of sampling; 31.3074 – constant value ($\lambda_{Pu}/\lambda_{Am}$); $A_{^{241}Am}$ – ^{241}Am activity increase measured after 16–18 years; λ_{Pu} – decay constant (0.050217 year^{-1}); λ_{Am} – decay constant (0.001604 year^{-1}); t – time from sampling to measurement of ^{241}Am (16–18 years).

Results and discussion

Plutonium ^{238}Pu and $^{239+240}Pu$

The plutonium concentration in southern Baltic changes horizontally from east to west, and the highest is in Pomeranian Bay, where richer in plutonium water inflows. The concentration of $^{239+240}Pu$ in seawater from Pomeranian Bay ranged 150 mBq·dm^{-3} and Gdańsk Bay it ranged only 5.2 μBq·m^{-3} (Table 1).

The obtained results indicate the Baltic organisms do not accumulate them on the same levels. $^{239+240}Pu$ concentration in marine animals from the southern Baltic Sea ranged from 0.33 mBq·kg^{-1} w.w. (fish) to 96 mBq·kg^{-1} w.w. (priapulida), and the respective bioconcentration factors (BCF) were 100 to 27,000. $^{239+240}Pu$ concentration in phytoplankton is 2.1 mBq·kg^{-1} w.w. The bioconcentration factor (BCF) of plutonium was estimated at 405. For zooplankton respective values were estimated at 0.8 mBq·kg^{-1} w.w. for $^{239+240}Pu$ and 150 for BCF (Table 1).

These data for Baltic fish show significant differences in plutonium concentrations between all the species examined. The highest values of $^{239+240}Pu$ activities for whole body were found in *Neogobius melanostomus* (6.50 mBq·kg^{-1} w.w.) and the lowest in sprat (0.33 mBq·kg^{-1} w.w.). The plutonium is non-uniformly distributed between the organs and tissues of the fish. Most of the $^{239+240}Pu$ in analyzed fish was located in internal organs (mainly alimentary track), the lowest values was found in muscles. Participation of evaluated Chernobyl-derived plutonium in Baltic fish reached from 20% for *Neogobius melanostomus* to 70% for herring [5].

The main source of $^{239+240}Pu$ inflow in Gdańsk Bay and Gdańsk Basin are rivers' water, which enrich these regions with 78% of total plutonium (Figs. 1 and 2). The total $^{239+240}Pu$ amount deposited in Gdańsk Bay and Gdańsk Basin is 1.18 TBq and 3.76 TBq, respectively. Almost whole plutonium is deposited in Baltic sediments. Because the area of Gdańsk Bay and Gdańsk Basin constitute only 1.2% and 4.4% of the Baltic Sea area and the capacity 1.3% and 5.7%, respectively, the results indicate the sediments of both regions are highly enriched in plutonium. In seawater of Gdańsk Bay (with suspended matter) there is about 2.33 GBq (0.2% of

Table 1. The concentrations of plutonium $^{239+240}Pu$ in water and organisms in southern Baltic ecosystem

Ecosystem compound	$^{239+240}Pu$ concentration [mBq·kg^{-1} w.w.]	BCF
Seawater		
Pomeranian Bay	150 ± 4*	–
Gdańsk Bay	5.2 ± 0.8*	–
Phytoplankton	2.1 ± 0.3	405
Zooplankton	0.8 ± 0.1	150
Phytobenthos	12 ± 2	3400
Zoobenthos		
Polychaeta	19 ± 2	5400
Priapulida	96 ± 7	27,000
Fish		
sprat	0.33 ± 0.04	100
cod	2.35 ± 0.11	650
herring	2.22 ± 0.11	600
N. melanostomus	6.50 ± 0.64	1900

* – μBq·dm^{-3}

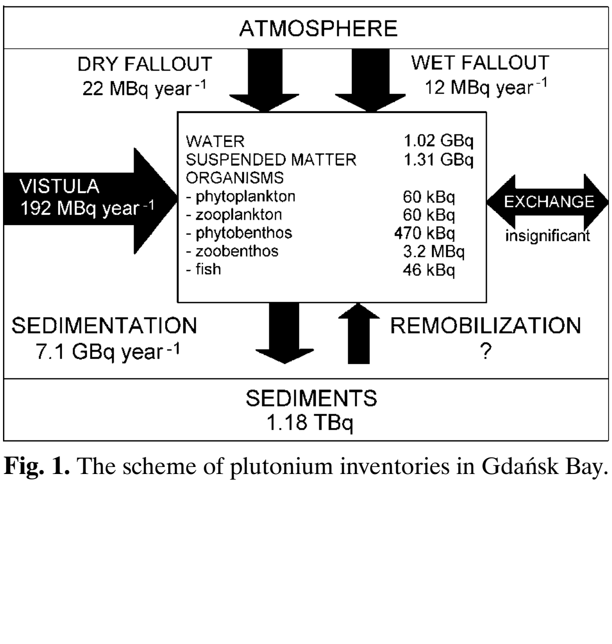

Fig. 1. The scheme of plutonium inventories in Gdańsk Bay.

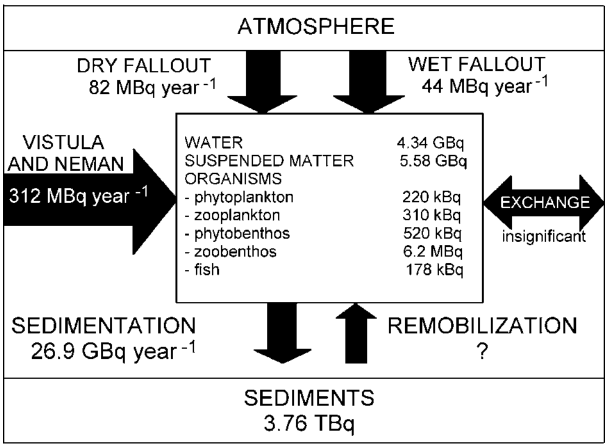

Fig. 2. The scheme of plutonium in inventories in Gdańsk Basin.

total amount) and 9.92 GBq (0.3% of total amount) in Gdańsk Basin. In both cases 56% of $^{239+240}$Pu is associated with suspended matter. Organisms in Gdańsk Bay contain 3.81 MBq and 7.45 MBq $^{239+240}$Pu in Gdańsk Basin. From this value in Gdańsk Bay 82.1% is deposited in zoobenthos, 13.6% in phytobenthos, 1.6% in phytoplankton, 1.5% in zooplankton and 1.2% in fish. In Gdańsk Basin 83.2% of plutonium in organisms is deposited in zoobenthos, 7.5% in phytobenthos, 3.6% in phytoplankton, 3.2% in zooplankton and 2.5% in fish (Figs. 1 and 2).

Plutonium ^{241}Pu

Plutonium released in the moment of reactor explosion existed as insoluble PuO_2, and almost whole amount fell as dry atmospheric fallout or with the wet fallout. Air filter analysis indicates after Chernobyl accident ^{241}Pu concentration increased suddenly over 3500 times and was decreasing slowly. In November 1986 got the level before the accident [2].

Results obtained for seawater indicates that ^{241}Pu concentration in samples from May 1987 was 0.23 Bq·m^{-3} for Gdańsk Bay and 0.11 Bq·m^{-3} for Gdańsk Deep (Table 2).

Almost whole plutonium is deposited in sediments, mostly as insoluble chemical forms (mainly PuO_2). The sedimentation of suspended matter in southern Baltic depends on the depth, redox conditions and its chemical constitution [2, 4]. Different plutonium activities in sediments were observed in different areas of the southern Baltic Sea are involved with their chemical constitution. Sandy sediments from Gdańsk Bay and external Puck Bay are less enriched in plutonium than muddy sediments from internal Gdańsk Deep and Puck Bay. The highest ^{241}Pu activity concentration was found in muddy Gdańsk Deep sediments (14.2 mBq·g^{-1} d.w.) and the value of the ^{241}Pu/$^{239+240}$Pu activity ratio was estimated at 39. The lowest ^{241}Pu activity concentration was found in southern Gdańsk Bay near Vistula River (0.16 mBq·g^{-1} d.w.). The values of the ^{241}Pu/$^{239+240}$Pu activity ratios were from 39 for Gdańsk Deep to 6.6 for external Puck Bay. Such low activity ratios in sediments indicate on low Chernobyl accident impact in the year after (Table 2).

^{241}Pu activity concentration in analyzed seaweed from Puck Bay, collected in 1986–1987, ranged from 0.22 mBq·g^{-1} d.w. for seed plants *Elodea canadensis* and 0.24 mBq·g^{-1} d.w. for *Zostera marina,* to 1.01 mBq·g^{-1} d.w. for brown algae *Pylaiella litoralis* (Table 3). The morphology of *Pylaiella litoralis* makes it easier to pollute the plant by deposition of the organic matter, mud and small benthic organisms in comparison to other seaweeds [2]. The values ^{241}Pu/$^{239+240}$Pu activity ratios were from 5.4 for green algae *Cladophora rupestris* to 35 for seed plant *Potamogeton pectinalis* (Table 3). It indicates that in 1987 there was no plutonium ^{241}Pu from Chernobyl accident in phytobenthos from Puck Bay.

In zoobenthos ^{241}Pu activity concentrations vary from 0.13 mBq·g^{-1} d.w. for crustacean *Saduria entomon* and 0.20 mBq·g^{-1} d.w. for *Balanus improvisus,* to 7.71 mBq·g^{-1} d.w. for polychaeta *Antinöella sarsi* and 9.20 mBq·g^{-1} d.w. for priapulida *Halicryptus spinulosus* (Table 3). The values of the ^{241}Pu/$^{239+240}$Pu activity ratios are varied also, from 1.7 for bivalves *Mytilus trossulus* to 64 for *Cardium glaucum*. Higher concentrations were found in organisms from priapulida and polychaeta and lower in crustaceans and bivalves. Benthic organisms contain more plutonium compared to phytobenthos. This indicates that plutonium in zoobenthos is accumu-

Table 2. ^{241}Pu concentration in seawater and sediments from southern Baltic collected in May 1987

Sample	^{241}Pu concentration [mBq·dm^3 ± SD]	^{241}Pu/$^{239+240}$Pu activity ratio
Seawater		
Gdańsk Bay	0.23 ± 0.03	140
Gdańsk Deep	0.11 ± 0.02	53
Sediments		
Gdańsk Bay	0.90 ± 0.14	7.5
southern Gdańsk Bay	0.16 ± 0.02	16
Gdańsk Deep	14.2 ± 2.5	39
internal Puck Bay	2.93 ± 0.27	7.2
external Puck Bay	1.52 ± 0.12	6.6

Table 3. ^{241}Pu concentration in phyto- and zoobenthos from Puck Bay collected in 1986 and 1987

Organism	^{241}Pu concentration [mBq·g^{-1} d.w. ± SD]	^{241}Pu/$^{239+240}$Pu activity ratio
Phytobenthos		
Cladophora rupestris	0.30 ± 0.05	5.4
Ulva lactuca	0.28 ± 0.08	28
Elodea canadensis	0.22 ± 0.09	9.2
Potamogeton pectinalis	0.39 ± 0.06	35
Pylaiella litoralis	1.01 ± 0.12	9.4
Zostera marina	0.24 ± 0.06	18
Zoobenthos		
Antinöella sarsi	7.71 ± 0.85	59
Balanus improvisus	0.20 ± 0.02	29
Cardium glaucum	1.66 ± 0.02	64
Gammarus sp.	1.80 ± 0.21	30
Halicryptus spinulosus	9.20 ± 1.20	11
Mytilus trossulus	0.28 ± 0.03	1.7
Saduria entomon	0.13 ± 0.06	5.0

lated from sediments and interstitial water inversely to phytobentos, where plutonium can be adsorbed from seawater [2].

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