Plutonium isotopes ^{238}Pu, $^{239+240}$Pu, ^{241}Pu and ^{240}Pu/^{239}Pu atomic ratios in the southern Baltic Sea ecosystem*

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Abstract

The paper summarizes the results of plutonium findings in atmospheric fallout samples and marine samples from the southern Baltic Sea during our research in 1986–2007. The activities of ^{238}Pu and $^{239+240}$Pu isotopes were measured with an alpha spectrometer. The activities of ^{241}Pu were calculated indirectly by ^{241}Am activity measurements 16–18 years after the Chernobyl accident. The ^{240}Pu/^{239}Pu atomic ratios were measured using accelerator mass spectrometry (AMS).

The ^{241}Pu activities indicate that the main impact of the Chernobyl accident was on the plutonium concentration in the components of the Baltic Sea ecosystem examined in this work. The highest ^{241}Pu/$^{239+240}$Pu activity ratio was found in sea water (140 ± 33). The AMS measurements of atmospheric fallout samples collected during 1986 showed a significant increase in the ^{240}Pu/^{239}Pu atomic ratio from 0.29 ± 0.04 in March 1986 to 0.47 ± 0.02 in April 1986.

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1. Introduction

The Baltic Sea is a small shelf sea, very sensitive to human impact. The main source of plutonium in the Baltic Sea, before the Chernobyl accident, was global fallout from nuclear weapons testing. At present the contribution from this source is smaller (only 1 GBq per year) (Skwarzec 1992). Other sources of plutonium, e.g. releases from the spent fuel facilities at Sellafield (UK) and Cap de la Hague (France), are less important. Since 26 April 1986 there has been a new source of plutonium – the Chernobyl accident – which should be taken into consideration in environmental risk assessments (Skwarzec 1995). The atmospheric fallout from the Chernobyl accident seems to be more important and it caused a plutonium emission estimated at 30 TBq of ^{238}Pu and 61.5 TBq of ^{239}Pu and ^{240}Pu (IAEA 1986). Moreover, nearly 250 rivers flow into the Baltic Sea carrying some 500 km^3 of fresh water per year containing 2 GBq of $^{239+240}$Pu (Mikulski 1982, Bergström & Carlsson 1994, Skwarzec 1995). Our experiments on plutonium sources in the Gulf of Gdańsk and the Gdańsk Basin (Figure 1) show that the main sources of these radionuclides are rivers (mainly the Vistula and the Neman), which supply these regions with 78% of its total Pu content (Skwarzec et al. 2003).

Marine plants and animals accumulate radionuclides from the aquatic environment. For this reason, it is very important to observe the impact of radionuclides on living organisms and their possible transfer to the human body. In the marine ecosystem the highest plutonium concentrations have been found in sediments, but the complex biogeochemical cycle of this element causes it to be present in all compartments of the environment. The results of our studies indicate an increase in plutonium bioavailability in the marine food chain of the southern Baltic Sea as the effect of its desorption

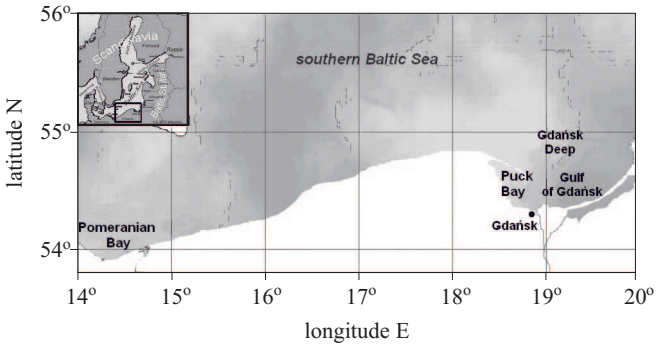

Figure 1. The southern Baltic Sea

from sediments and accumulation in benthic organisms (Skwarzec 1995, 1997a, 1997b, Skwarzec et al. 2001, Strumińska & Skwarzec 2004, 2006).

This work presents our results of the determination of the most important α-emitting plutonium isotopes (^{238}Pu and $^{239+240}$Pu) as well as the β^--emitting ^{241}Pu in Baltic sea water, organisms and sediment samples. ^{241}Pu is less important in terms of its radiotoxicity than the α-emitting plutonium radionuclides (^{238}Pu, ^{239}Pu, ^{240}Pu), but is quite significant because of its greatest contribution to the whole plutonium fallout and quite short half-life (14.35 years), decaying to very highly radiotoxic, long-living, α-emitting americium ^{241}Am ($T_{1/2}$ = 432.7 years) (Mussalo et al. 1980). We also give the results of ^{240}Pu/^{239}Pu isotope ratios in some samples. Determination of plutonium ^{240}Pu/^{239}Pu atomic ratios in the environment allows the contamination sources to be identified. As the decay energies of ^{239}Pu and ^{240}Pu are quite similar, it is quite difficult to separate these isotopes using an alpha spectrometer. But with accelerator mass spectrometry (AMS), a very useful technique for low-level mass measurements, this separation is possible (McAninch et al. 2000).

2. Material and methods

Plutonium ^{238}Pu and $^{239+240}$Pu concentrations in Baltic samples taken from 1998 to 2002 were measured using an alpha spectrometer (Canberra-Packard, USA). All alpha spectrometric sources were prepared using electrolysis on a stainless steel disc after mineralization, separation and purification on an anion exchange resin with ^{242}Pu as yield tracer (Skwarzec 1995, 1997a).

The ^{241}Pu isotope was measured in the same samples taken from 1986 to 1988 as part of some earlier research (Skwarzec 1995, 1997b). The alpha plutonium isotope spectra acquired were compared with the same spectra obtained 16–18 years earlier, and the activities on the sampling date were calculated (Skwarzec 1995, 1997b, Strumińska & Skwarzec 2006). The determination of ^{241}Pu in the samples was done indirectly by measuring the increment in ^{241}Am from the decay of β^--emitting ^{241}Pu. The ^{241}Pu activity was calculated from the following formula:

$$A_{Pu_0} = 30.1141 \times \frac{A_{^{241}Am}\, e^{+\lambda_{Am}\, t}}{(1 - e^{-\lambda_{Pu}\, t})}, \qquad (1)$$

where

A_{Pu_0} – ^{241}Pu activity at the time of sampling;
30.1141 – a constant value ($\lambda_{Pu}/\lambda_{Am}$);
$A_{^{241}Am}$ – ^{241}Am activity increment measured after 16-18 years;
λ_{Pu} – 0.050217 year^{-1};

λ_{Am} – 0.001604 year $^{-1}$;
t – time from sampling to ^{241}Am measurement (16–18 years).

The ^{240}Pu/^{239}Pu atomic ratios were measured in 2007 using accelerator mass spectrometry (AMS). This modern analytical technique allows the radioactivity of long-living radionuclides to be measured in environmental samples (Fifield et al. 1996, McAninch et al. 2000, Fifield et al. 2004, Wacker et al. 2005, Chamizo et al. 2006). New generation AMS enables the heaviest elements (actinides) to be detected and the isotopic ratios between them to be estimated. In the case of plutonium it is possible to measure femtograms (10^{-15} g) of this element and calculate the isotopic ratios of ^{238}Pu/^{240}Pu and ^{240}Pu/^{239}Pu. All sources for AMS measurements were prepared as Al/Cu cathode targets after preparing plutonium with Fe/Al. The targets were measured using accelerator mass spectrometry (AMS) (ETHZ/PSI, Zürich, Switzerland) equipped with a 3x3 mm^2 silicon detector (Kirchner & Noack 1988, Fifield et al. 1996, McAninch et al. 2000, Hrnecek et al. 2005, Chamizo et al. 2006).

3. Results and discussion

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

The biggest source of $^{239+240}$Pu inflow into the Gulf of Gdańsk and the Gdańsk Basin is river water (particularly that from the Vistula and the Neman) (Skwarzec et al. 2003). According to our results these rivers supply 78% of the total plutonium present in these regions (Skwarzec et al. 2003). The total amount of $^{239+240}$Pu deposited in the Gulf of Gdańsk and the Gdańsk Basin is 1.18 TBq and 3.76 TBq respectively, and almost all (ca 99%) of this plutonium is deposited in the sediments. Because the Gulf of Gdańsk and the Gdańsk Basin respectively constitute only 1.2% and 4.4% of the total area of the Baltic Sea and 1.3% and 5.7% of its volume, the results indicate that the sediments of both regions contain considerable amounts of plutonium (Skwarzec et al. 2003). The sea water from the Gulf of Gdańsk (including suspended particulate matter – SPM) contains about 2.33 GBq (0.2% of the total amount), and that from the Gdańsk Basin 9.92 GBq (0.3% of total amount). In both cases 56% of $^{239+240}$Pu is associated with SPM. The Gulf of Gdańsk, and to a lesser extent the Gdańsk Basin, are under the predominant influence of water from the Vistula. This river supplies ca 32.1 km^3 of water to the Gulf of Gdańsk each year, which constitutes 7% of the total river inflow to the Baltic Sea and 10% of the water volume in the Gulf of Gdańsk (Mikulski 1970). The similar distribution of plutonium between sea water and SPM may be due to the conditions specific to the Baltic Sea. The mechanisms prevailing in

the Baltic Sea differ from those of other aquatic systems. Moreover, the Vistula mouth is an area where the riverine sediments are substantially modified by currents and waves. At the river mouth there is no shallow fraction of the mud and loam that makes up about 60% of the material transported with Vistula waters. Intensive hydrodynamic and chemical processes give rise to highly differentiated and unstable river-marine muddy-loam sediments with sandy interlayers (Uścinowicz & Zachowicz 1994). The result is that plutonium in Vistula water is transported to and deposited in deeper parts of the Gulf of Gdańsk and the Gdańsk Deep, in accordance with the water circulation obtaining in a region where river and sea water mix (Bojanowski 1981).

The plutonium concentration in southern Baltic sea water changes horizontally from east to west, and is the highest in the Pomeranian Bay (small plutonium-rich inflows from the North Sea). The concentration of $^{239+240}$Pu in sea water from the Pomeranian Bay was estimated at 150 ± 4 mBq m^{-3},

Table 1. $^{239+240}$Pu concentrations in water and organisms in the southern Baltic Sea ecosystem, calculated for 95% confidence intervals (2σ). (Skwarzec 1995, Skwarzec et al. 2001, Strumińska & Skwarzec 2004)

Ecosystem component	Average $^{239+240}$Pu concentration [μBq g^{-1} dw $\pm$ SD]	BCF	Reference
Sea water			
Pomeranian Bay	$150 \pm 4^{*}$	-	Strumińska & Skwarzec 2004
Gulf of Gdańsk	$5.2 \pm 0.8^{*}$	-	Strumińska & Skwarzec 2004
Phytoplankton	2.1 ± 0.3	405	Strumińska & Skwarzec 2004
Zooplankton	0.8 ± 0.1	150	Strumińska & Skwarzec 2004
Phytobenthos	12 ± 2	3 400	Skwarzec 1995
Zoobenthos			
Polychaeta	19 ± 2	5 400	Skwarzec 1995
Priapulida	96 ± 7	27 000	Skwarzec 1995
Fish			
sprat	$0.33 \pm 0.04^{**}$	100	Skwarzec et al. 2001
cod	$2.35 \pm 0.11^{**}$	650	Skwarzec et al. 2001
herring	$2.22 \pm 0.11^{**}$	600	Skwarzec et al. 2001
N. melanostomus	$6.50 \pm 0.64^{**}$	1 900	Skwarzec et al. 2001

* – mBq m^{-3}, ** – μBq g^{-1} wet weight.

whereas in the Gulf of Gdańsk it was only 5.2 ± 0.8 mBq m^{-3} (Table 1) (Strumińska & Skwarzec 2004).

Living organisms in the Gulf of Gdańsk and in the Gdańsk Basin contain 3.81 MBq and 7.45 MBq $^{239+240}$Pu respectively. Of these amounts in the Gulf of Gdańsk, 82.1% is deposited in the zoobenthos, 13.6% in the phytobenthos, 1.6% in the phytoplankton, 1.5% in the zooplankton and 1.2% in fish. In the Gdańsk Basin, 83.2% of plutonium is deposited in the zoobenthos, 7.5% in the phytobenthos, 3.6% in the phytoplankton, 3.2% in the zooplankton and 2.5% in fish (Figures 2 and 3) (Skwarzec et al. 2003). Our studies show that organisms from the open Baltic Sea do not accumulate plutonium to the same levels. $^{239+240}$Pu concentrations in marine animals from the southern Baltic Sea ranged from 0.33 ± 0.04 μBq g^{-1} ww (wet weight) in fish (sprat *Sprattus sprattus*) to 96 ± 7 μBq g^{-1} dw (dry weight) in the priapulid worm *Halicryptus spinulosus*, the respective bioconcentration factors (BCF) being from 100 to 27 000 (Bergström & Carlsson 1994, Skwarzec 1995, Skwarzec et al. 2001). The mean $^{239+240}$Pu concentration in phytoplankton was 2.1 ± 0.3 μBq g^{-1} dw and the bioconcentration factor (BCF) of plutonium in these organisms

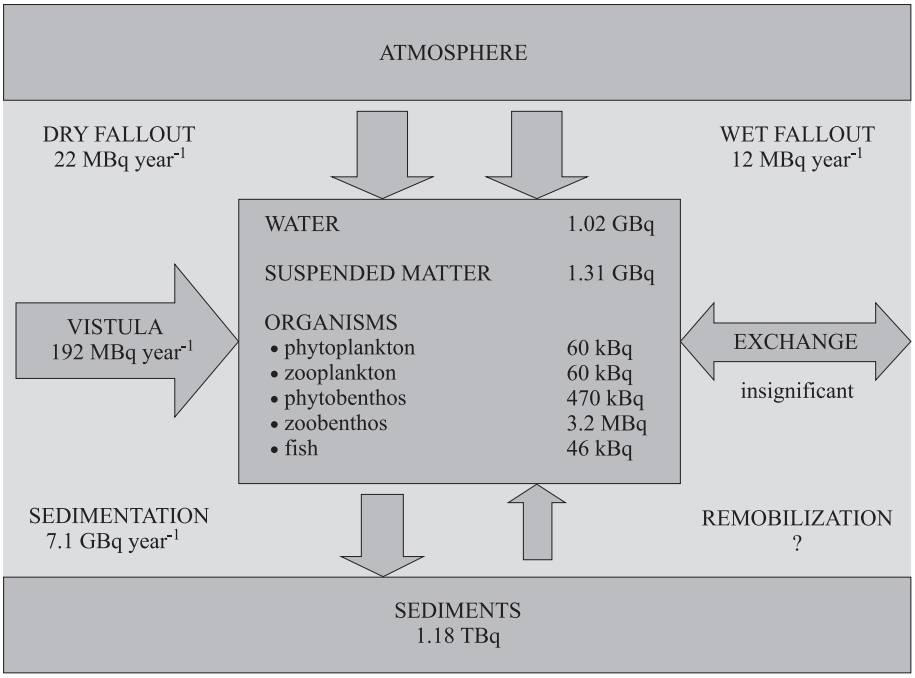

Figure 2. The scheme of plutonium inventories in the Gulf of Gdańsk (Skwarzec et al. 2003)

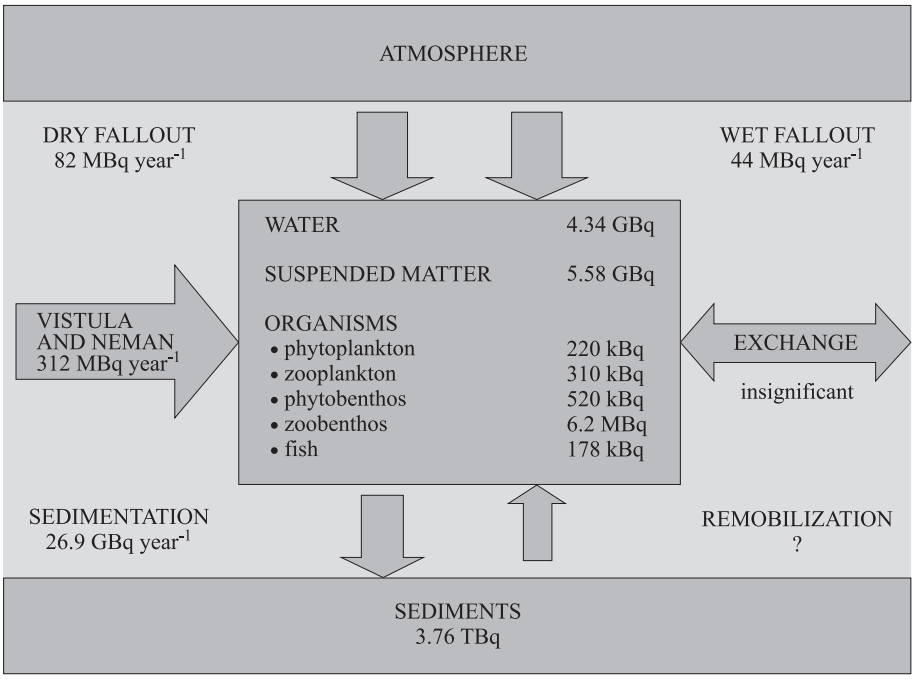

Figure 3. The scheme of plutonium inventories in the Gdańsk Basin (Skwarzec et al. 2003)

was estimated at 405. The corresponding values for zooplankton were 0.8 ± 0.1 μBq g^{-1} dw for $^{239+240}$Pu and 150 (BCF) (Table 1) (Strumińska & Skwarzec 2004). The results of studies show significant differences in plutonium concentrations between the fish species examined. The highest whole body $^{239+240}$Pu activities were found in the benthic *Neogobius melanostomus* (6.50 ± 0.64 μBq g^{-1} ww) and the lowest in pelagic sprat (0.33 ± 0.04 μBq g^{-1} ww) (Skwarzec et al. 2001). Plutonium is non-uniformly distributed between the organs and tissues of these fish. Most of the $^{239+240}$Pu in fish was located in their internal organs (mainly the alimentary tract), and the least in the muscles. The contribution of Chernobyl-derived plutonium in Baltic fish was from 20% in *Neogobius melanostomus* to 70% in herring (Skwarzec et al. 2001).

3.2. Plutonium ^{241}Pu

The ^{241}Pu isotope was measured in the same samples taken from 1986 to 1988, which were examined during earlier research (Skwarzec 1995, 1997b).

Before the Chernobyl accident the $^{241}Pu/^{239+240}Pu$ activity ratio in airborne dust over Gdynia ranged from 33 to 39. In Tsukuba (Japan), by contrast, it was 14.5, mostly as a result of Chinese nuclear tests (Hirose 1995).

The plutonium released into the environment at the moment of the reactor explosion was in the form of insoluble PuO_2, and almost the whole amount fell as dry atmospheric fallout or with wet precipitation (Skwarzec 1995). Analysis of air filter samples (airborne dust samples in Gdynia were collected monthly in 1986) indicated that the ^{241}Pu concentration rose abruptly by a factor of ca 3500 after the Chernobyl accident, slowly diminishing thereafter (Table 2). The average $^{241}Pu/^{239+240}Pu$ activity ratios measured in April and May were similar to those calculated in Vienna at the beginning of May 1986, when they ranged from 0.33 to 0.76 with a most reliable value of 0.47. Also ^{241}Pu activities were very high – with a maximum value of 6090 μBq m^{-3} (Irlweck & Wicke 1997). The $^{241}Pu/^{239+240}Pu$ activity ratio in the vicinity of the damaged nuclear power plant ranged from 54.7 to 73.1 with a mean of 60.2, which corresponds fairly well with our data in April (54 ± 0.8). This data also corresponds with the $^{241}Pu/^{239+240}Pu$ activity ratio measured in Salzburg (51.2) (Irlweck & Wicke 1997). Vukanac et al. (2006) gave much higher ^{241}Pu concentrations in surface air over Belgrade (Serbia and Montenegro) (to 7800 μBq m^{-3}) in comparison to Gdynia, and the average activity ratio $^{241}Pu/^{239+240}Pu$ due to the Chernobyl accident at the Belgrade site was approximately 100 at the collection time. By November 1986 the ^{241}Pu

Table 2. ^{241}Pu concentration, $^{241}Pu/^{239+240}Pu$ activity ratio and $^{240}Pu/^{239}Pu$ atomic ratio in airborne dust collected over Gdynia (Poland) in 1986, calculated for 95% confidence intervals (2σ)

Sample	^{241}Pu concentration* [μBq g^{-1} dw $\pm$ SD]	$^{241}Pu/^{239+240}Pu$* activity ratio	$^{240}Pu/^{239}Pu$ atomic ratio
January	0.162 ± 0.034	39 ± 10	0.36 ± 0.06
February	1.008 ± 0.143	31 ± 5.3	0.23 ± 0.03
March	0.860 ± 0.160	33 ± 7.9	0.29 ± 0.04
April	3504 ± 32.9	54 ± 0.8	0.47 ± 0.02
May	31.9 ± 4.1	48 ± 10	0.38 ± 0.02
June	2.45 ± 1.00	40 ± 23	0.41 ± 0.06
July	2.34 ± 0.49	40 ± 13	0.49 ± 0.10
September	2.85 ± 0.86	69 ± 30	0.47 ± 0.07
November	1.04 ± 0.35	57 ± 26	0.44 ± 0.15
December	0.99 ± 0.28	38 ± 14	0.37 ± 0.10

*Strumińska & Skwarzec 2006.

concentration in Gdynia had fallen back to the levels recorded before the Chernobyl accident (February 1986) (Table 2).

Results obtained for sea water indicated that the ^{241}Pu concentration in samples from May 1987 was 0.17 ± 0.02 Bq m^{-3} for the Gulf of Gdańsk and 0.09 ± 0.02 Bq m^{-3} for the Gdańsk Deep (Table 3).

Table 3. ^{241}Pu concentration and ^{241}Pu/$^{239+240}$Pu activity ratios in sea water and sediments from the southern Baltic Sea collected in May 1987, calculated for 95% confidence intervals (2σ). (Strumińska & Skwarzec 2006)

Sample	Average ^{241}Pu concentration [μBq g^{-1} dw $\pm$ SD]	^{241}Pu/$^{239+240}$Pu activity ratio
Sea water		
Gulf of Gdańsk	$0.17 \pm 0.02^*$	74 ± 17
Gdańsk Deep	$0.09 \pm 0.02^*$	89 ± 20
Sediments		
Gulf of Gdańsk	0.86 ± 0.14	7.0 ± 1.2
Gdańsk Deep	13.7 ± 2.4	37 ± 8
Inner Puck Bay	2.82 ± 0.26	6.9 ± 0.7
Outer Puck Bay	1.46 ± 0.12	6.4 ± 0.6

* – Bq m^{-3}.

Almost all the ^{241}Pu, like the plutonium alpha emitters, is deposited in sediments. The sedimentation of suspended matter in the southern Baltic Sea depends on the depth, redox conditions and its chemical composition (Skwarzec 1995, 1997b). The different activities of plutonium isotopes in the sediments of the investigated areas in the southern Baltic Sea depend on their chemical composition. The sandy sediments from the Gulf of Gdańsk and the outer Puck Bay contain less plutonium than the muddy sediments from the Gdańsk Deep and the inner Puck Bay (Skwarzec 1992, 1995, 1997b). The ^{241}Pu activity concentration was the highest in the muddy sediments of the Gdańsk Deep (13.7 ± 2.4 mBq g^{-1} dw (dry weight)) and the ^{241}Pu/$^{239+240}$Pu activity ratio was estimated at 37 ± 8. The lowest ^{241}Pu activity concentration was found in the southern Gulf of Gdańsk (0.86 ± 0.14 mBq g^{-1} dw). The ^{241}Pu/$^{239+240}$Pu activity ratios ranged from 37 ± 8 in the Gdańsk Deep to 6.4 ± 0.6 in the outer Puck Bay. Such low activity ratios in the sediments indicate the low impact of the Chernobyl accident in the following years or a delayed inflow effect (Table 3).

^{241}Pu activity concentrations in seaweed from the Puck Bay, collected in 1986–87, ranged from 0.37 ± 0.06 mBq g^{-1} dw in the macrophyte *Potamogeton pectinalis* to 0.97 ± 0.11 mBq g^{-1} dw in the brown alga *Pilayella litoralis* (Table 4). A large amount of ^{241}Pu was also found in the

green alga *Cladophora rupestris* 0.28 ± 0.05 mBq g^{-1} dw. The morphology of *C. rupestris* facilitates the contamination of the plant through the deposition of organic matter, mud and small benthic organisms in comparison to other seaweeds (Skwarzec 1995). The values of $^{241}Pu/^{239+240}Pu$ activity ratios in seaweeds ranged from 5.0 ± 1.0 in *Cladophora rupestris* to 32 ± 5 in *Potamogeton pectinalis* (Table 4). This indicated that in 1987 there were no detectable amounts of ^{241}Pu from the Chernobyl accident in the Puck Bay phytobenthos.

Table 4. ^{241}Pu concentration and $^{241}Pu/^{239+240}Pu$ activity ratios in phyto- and zoobenthos from Puck Bay collected in 1986 and 1987, calculated for 95% confidence intervals (2σ). (Strumińska & Skwarzec 2006)

Organism	Average ^{241}Pu concentration [μBq g^{-1} dw $\pm$ SD]	$^{241}Pu/^{239+240}Pu$ activity ratio
Phytobenthos		
Cladophora rupestris	0.28 ± 0.05	5.0 ± 1.0
Ulva lactuca	0.27 ± 0.08	26 ± 8
Elodea canadensis	0.21 ± 0.09	9.0 ± 3.9
Potamogeton pectinalis	0.37 ± 0.06	32 ± 5
Pilayella litoralis	0.97 ± 0.11	9.1 ± 1.2
Zostera marina	0.23 ± 0.06	18 ± 5
Zoobenthos		
Antinella sarsi	7.42 ± 0.82	55 ± 6
Balanus improvisus	0.19 ± 0.02	27 ± 3
Cardium glaucum	1.56 ± 0.02	60 ± 18
Gammarus sp.	1.73 ± 0.21	29 ± 4
Halicryptus spinulosus	8.85 ± 1.15	10 ± 1
Mytilus trossulus	0.27 ± 0.03	1.7 ± 0.4
Saduria entomon	0.13 ± 0.06	4.9 ± 2.3

^{241}Pu activity concentrations in the zoobenthos varied from 0.13 ± 0.06 mBq g^{-1} dw in the crustacean *Saduria entomon* and 0.19 ± 0.02 mBq g^{-1} dw in the barnacle *Balanus improvisus* to 7.42 ± 0.82 mBq g^{-1} dw in the polychaete *Antinöella sarsi* and 8.85 ± 1.15 mBq g^{-1} dw in the priapulid worm *Halicryptus spinulosus* (Table 4). The $^{241}Pu/^{239+240}Pu$ activity ratios also varied over a wide range, from 1.7 ± 0.4 in bivalves *Mytilus trossulus* to 60 ± 18 in *Cardium glaucum*. Higher concentrations were found in priapulids and polychaetes, lower ones in crustaceans and bivalves. The zoobenthos contained more plutonium than the phytobenthos. This indicates that plutonium in the zoobenthos is accumulated from sediments and interstitial water, unlike the phytobenthos, where plutonium can be adsorbed from sea water (Skwarzec 1995, 1997b).

3.3. ^{240}Pu/^{239}Pu atomic ratios in samples collected in 1986

The plutonium nuclide compositions in the filter samples corresponded to their means of release into the environment. The measurements of atmospheric fallout samples collected monthly throughout 1986 enabled the Chernobyl peak effect on Gdynia (Poland) to be tracked (Table 2). We observed quite high ^{240}Pu/^{239}Pu atomic ratios from January to March 1986 (from 0.24–0.36) compared to the global stratospheric fallout ^{240}Pu/^{239}Pu atomic ratios (0.18) (Buesseler 1997). Reported atomic ratios of ^{240}Pu/^{239}Pu from global stratospheric fallout were 0.176 ± 0.014 and 0.180 ± 0.014, whereas those from close-in tropospheric fallout from the Pacific Proving Grounds (PPG) in the Marshall Islands were 0.33–0.36 (Diamond et al. 1960, Krey et al. 1976, Komura et al. 1984, Buesseler 1997, Kelley et al. 1999). Air filter data obtained from January to March 1986 should be correlated with typical values for samples affected only by global fallout, although other accidents should also be recalled (Kyshtym 1957, Leningrad 1975, Chernobyl-1 1982) (Bojarski et al. 1992, IAEA 1993). One of the reasons for the elevated ^{240}Pu/^{239}Pu atomic ratio over Poland could, for example, have been the Chernobyl accident (reactor No. 1) in September 1982, where part of the core containing radioactive material was released into the industrial zone and city of Pripyat, causing irradiation of the staff involved in the repair of the core as well as irradiation of the plant and the city area (Medvedev 1990). The other, more probable, reason is the impact of aerosols from North Sea inflows. The atomic ratios of ^{240}Pu/^{239}Pu from western European nuclear fuel reprocessing facilities were 0.34 ± 0.03 in the effluent from the Cap la Hague (Oughton et al. 1999).

Our data indicate a considerable increase in the ^{240}Pu/^{239}Pu atomic ratio from 0.29 ± 0.04 in March 1986 to 0.47 ± 0.02 in April 1986 (Table 2). This value corresponds with the ^{240}Pu/^{239}Pu atomic ratios estimated in the reactor core (from 0.39 to 0.563) (Kirchner & Noack 1988, Begichev et al. 1990). Such a high ^{240}Pu/^{239}Pu atomic ratio (0.42) was also recorded a few days after the Chernobyl accident in aerosols over Vilnius (Lithuania) (Lujaniene et al. 2009). A similarly high increase in the ^{240}Pu/^{239}Pu atomic ratio to 0.47 ± 0.07 (a value close to that of the reactor core) was also recorded in September 1986. From November 1986 ^{240}Pu/^{239}Pu atomic ratios decreased, which confirmed that the plutonium concentration in the Baltic Sea had returned to the same level as before the accident (Skwarzec 1995). In December 1986 the ^{240}Pu/^{239}Pu atomic ratio was calculated at 0.37 ± 0.10, which is similar to the value recorded in January 1986 – 0.36 ± 0.04. Slightly smaller, but still similar ^{240}Pu/^{239}Pu atomic ratios were found in forest soil collected from north-eastern Poland a few years later: Szczerba – 0.309 ± 0.011, Plaska – 0.348 ± 0.011, Trzcianka – 0.308 ± 0.024 (Ketterer

et al. 2004). Clearly, we can expect $^{240}Pu/^{239}Pu$ atomic ratios to continue falling. Over Vilnius (Lithuania) this ratio decreased from 0.42 in 1986 to 0.20 in 2002 (Lujaniene et al. 2009).

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