

Radionuclides of iron (^{55}Fe), nickel (^{63}Ni), polonium (^{210}Po), uranium (^{234}U , ^{235}U , ^{238}U) and plutonium (^{238}Pu , $^{239+240}\text{Pu}$, ^{241}Pu) in Poland and Baltic Sea environment

Bogdan Skwarzec,
Dagmara I. Strumińska,
Alicja Boryło

Abstract The paper presents the results of determination of natural [polonium (^{210}Po), uranium (^{234}U , ^{235}U , ^{238}U)] and artificial radionuclides [iron (^{55}Fe), nickel (^{63}Ni), and plutonium (^{238}Pu , $^{239+240}\text{Pu}$, ^{241}Pu)] in Poland and the southern Baltic Sea ecosystem as well as the recognition of their accumulation process in the trophic chain. All presented results is a summary of long lasting studies realized by the present authors. Results of this study indicated that the principal sources of ^{55}Fe and ^{63}Ni in the Baltic Sea environment is the release from Swedish nuclear power plants. Investigation on the polonium ^{210}Po and plutonium ^{238}Pu , $^{239+240}\text{Pu}$ and ^{241}Pu concentration in Baltic and land biota revealed that these radionuclides are strongly accumulated by some species. Moreover, it was found that fish, mushrooms, cigarettes constitutes important sources of ^{210}Po for people. The Baltic Sea algae, benthic animals and fish concentrate uranium only to a small degree. In Baltic sediments, the concentration of uranium increases with core depth and it is connected with the diffusion of ^{234}U , ^{235}U and ^{238}U from sediments. The values of $^{234}\text{U}/^{238}\text{U}$ activity ratio in the sediments indicated that the reduction process of U(VI) to U(IV) and the removing of autogenic uranium from seawater to sediments in the Gdańsk Deep constitutes a small part only.

Key words radionuclides • iron • nickel • polonium • uranium • plutonium • Baltic Sea

Introduction

Radionuclides existing in the environment, both natural and artificial, are accumulated in plants and animals and transferred through the trophic chain. That is the way which more and more wakes up to the interest of long-term impact on living organisms and the possibility of their transfer through feed way to human body. Basing on their high radiotoxicity to living organisms the alpha emitting radionuclides (^{210}Po , ^{234}U , ^{235}U , ^{238}U , ^{238}Pu , $^{239+240}\text{Pu}$) and beta emitting radionuclides (^{55}Fe , ^{63}Ni , ^{241}Pu) were chosen for the experiments [20].

In a nuclear power reactor, a stainless construction material contains iron and nickel which are neutron activated and give rise to the radioactive nuclides ^{55}Fe and ^{63}Ni . The ^{55}Fe isotope (half-life time 2.686 y), which is a beta (electron capture) emitter and the ^{63}Ni isotope (half-life time 100.1 y) which is also a beta particle emitter, are fairly important in radiological protection [2, 7].

Polonium and uranium belong to the natural uranium decay series starting from ^{238}U , but ^{210}Po fate depends on the preceding members of this series, especially on ^{226}Ra and ^{210}Pb [18]. The total amount of airborne ^{210}Po depends directly on the amount of ^{210}Pb supplied and formed in the atmosphere [8, 9]. Additional amounts of ^{210}Po in the atmosphere are emitted directly from

B. Skwarzec[✉], D. I. Strumińska, A. Boryło
Chair of Analytical Chemistry,
Faculty of Chemistry,
University of Gdańsk,
18/19 Sobieskiego Str., 80-952 Gdańsk, Poland,
Tel.: +48 58 345 03 38, Fax: +48 58 345 04 72,
E-mail: bosk@chem.univ.gda.pl

Received: 10 October 2005

Accepted: 15 December 2005

the surface ocean and Earth surface, as a result of forest fires [15] and volcanic eruptions [4, 11]. Polonium from the air is precipitated within dry atmospheric fallout, rain and oceans [17]. Polonium isotopes belong to the most radiotoxic nuclide to human beings [14]. The main sources of ^{210}Po in the human body are food and cigarette smoke [20].

Uranium occurs naturally in the Earth's crust and is present in much higher concentrations, along with thorium and rare-earth elements, in areas where monazite sand occurs [18]. Naturally occurring uranium contains three alpha emitting radionuclides: ^{238}U , ^{235}U and ^{234}U . The source of uranium in the marine environment is the atmospheric precipitation of terrigenous (rock) materials, as well as rivers [10]. In ocean, seawater and river water in oxidation conditions, natural uranium exists predominantly in the form of uranyl carbonate anion $[\text{UO}_2(\text{CO}_3)_2]^{2-}$ and $[\text{UO}_2(\text{CO}_3)_3]^{4-}$ [12]. In estuarine zones, where rivers and seawater mix, uranium generally behaves conservatively, that is, its concentration is varying linearly with salinity [13, 18].

Plutonium radionuclides (^{238}Pu , ^{239}Pu , ^{240}Pu and ^{241}Pu) belongs to the group of man-made artificial radionuclides. These nuclides are important from the radioecological point of view due to their high radiotoxicity, long physical half-life, high chemical reactivity and long residence in biological system [3]. The principal source of plutonium radionuclides in the Baltic Sea is atmospheric fallout from nuclear weapon tests [5, 18]. Since 26 April 1986 another source of plutonium isotopes, the Chernobyl-originated radioactive debris, had to be taken into account [1].

Materials and methods

Samples of natural waters (seawater, river and consumption water), biota (Baltic and land), sediments and soil were collected in the years 1980–2003 in various regions of Poland and southern Baltic Sea.

Radioanalytical procedures for the determination of ^{55}Fe and ^{63}Ni in analyzed samples (water, sediments, soil and biological material) are described by Skwarzec [23]. The procedure is based on the coprecipitation of ^{55}Fe with iron hydroxide in natural and reactor water, ashing and mineralization of sediments, soil and biota samples, and sequential separation and purification of the iron and nickel on anion exchange resin. The

separated elements are electroplated onto copper discs and the activity of ^{55}Fe and ^{63}Ni was measured by beta spectrometry using an anti-coincidence Geiger-Müller gas flow counter (four-channel system from the Risø National Laboratory, Denmark) [6, 23]. The details of radiochemical methods and measurements of the described radionuclides are well known to the authors and were published in papers [7, 18, 19, 23].

Polonium, uranium and plutonium in natural waters were coprecipitated with manganese dioxide. The biological, sediment and soil materials were mineralized using concentrated nitric, hydrochloric and hydrofluoric acids. Polonium, uranium and plutonium were separated using an anion-exchange resin [18]. The activity of ^{210}Po , ^{234}U , ^{235}U , ^{238}U , ^{238}Pu , $^{239+240}\text{Pu}$ was measured using alpha spectrometry (S470 Alpha Analyst, Canberra-Packard). The determination of ^{241}Pu in the analyzed samples were done indirectly by alpha measurements of ^{241}Am ingrowth from the β -emitting ^{241}Pu .

The accuracy and precision of radiochemical methods were satisfactory, i.e. less than 10%, as estimated by the analysis of IAEA reference materials. The chemical yield varied from 60% for plutonium radionuclides to 95% for ^{210}Po . The counting efficiency was around 25% for ^{55}Fe , 30% for the alpha emitters (^{210}Po , ^{234}U , ^{235}U , ^{238}U , ^{238}Pu , $^{239+240}\text{Pu}$) and 40% for ^{63}Ni giving the least detectable activity of 1–2 mBq at 3000 min counting time for ^{55}Fe and ^{63}Ni , 0.33 mBq at 1500 min for ^{210}Po and uranium isotopes, and 0.12 mBq at 75,000 min for plutonium radionuclides [19, 23].

Results and discussion

Iron ^{55}Fe and nickel ^{63}Ni

The activity concentrations of ^{55}Fe and ^{63}Ni are presented in Table 1. The analysis of seawater samples showed that the concentration of ^{55}Fe and ^{63}Ni in the surface Baltic water are less than 2–5 mBq·dm $^{-3}$. However, the concentration of ^{55}Fe in the reactor water samples from the Barseback nuclear power plant (Sweden) fall within the range 0.15–2185 mBq·dm $^{-3}$. Baltic organisms, thanks to their ability to concentrate ^{55}Fe and ^{63}Ni , are often used as bioindicators of environmental radioactive pollution. The brown algae *Fucus vesiculosus* have been found to preferentially accumulate ^{55}Fe and ^{63}Ni [6, 7]. The Baltic fish accumulate ^{63}Ni , but, in contrary to stable

Table 1. The activity concentrations of ^{55}Fe and ^{63}Ni in the Baltic Sea

Sample	Activity concentration	
	^{55}Fe	^{63}Ni
Seawater	<2 mBq·dm $^{-3}$	<5 mBq·dm $^{-3}$
Reactor water (Barseback)	0.15–2185 Bq·dm $^{-3}$	–
<i>Fucus vesiculosus</i>	0.13–3.5 Bq·kg $^{-1}$ d.w.	0.1–100 Bq·kg $^{-1}$ d.w.
<i>Cladonia alpestris</i>	–	0.1–0.5 Bq·kg $^{-1}$ d.w.
<i>Mytilus trossulus</i>	–	10–20 mBq·kg $^{-1}$ d.w.
Fish	<3 mBq·kg $^{-1}$ w.w.	3–340 mBq·kg $^{-1}$ w.w.
Sediments	<1 mBq·kg $^{-1}$ d.w.	0.1–3.8 mBq·kg $^{-1}$ d.w.

Table 2. The mean values of polonium activity concentrations in seawater, drinking water and beer in Poland

Water	^{210}Po activity concentration [mBq·dm ⁻³]
Southern Baltic Sea	0.59
Drinking water (Gdańsk)	0.48
Bottled mineral drinking water	1.28
Beer	4.63

nickel, the radioactive nickel is not uniformly distributed in the fish organism [22]. More than 95% of the total ^{63}Ni content in cod and herring is located in the fillet with skin and scales. Analyzing low concentration of ^{63}Ni in the alimentary track, a low activity of ^{63}Ni in soft tissues of cod and herring can be foreseen. This means that a passive adsorption onto the surface of skin and scale with mucus is responsible for the uptake of ^{63}Ni by Baltic fish [22].

Polonium ^{210}Po

The analysis of seawater samples revealed that the mean concentration of ^{210}Po in Baltic water was 0.59 mBq·dm⁻³, 80% of which was in soluble forms. There were significant differences in the concentrations of dissolved polonium in the samples analyzed. Earlier studies revealed that the concentration of ^{210}Po in suspended matter was 74 mBq·kg⁻¹ d.w. [19]. Radiochemical analysis of drinking water (Gdańsk agglomeration and mineral bottled water) as well as beers in Poland indicate low activity levels of polonium. They are safe for people from the radiological point of view (Table 2) [25–27].

Taking into account all the analyzed radionuclides it can be said that ^{210}Po is accumulated in land and Baltic organisms to the highest degree. Mean values of the bioconcentration factor (BCF) of ^{210}Po in the Baltic organisms calculated fell within the range 1.5×10^3 – 3.2×10^4 (Table 3). In the case of planktonic organisms, the BCF values increased in the sequence: phytoplankton < macrozooplankton < mesozooplankton, while in the zoobenthos they increased in the following order: Polychaeta < Priapulida < Malacostraca < Bivalves (soft tissues) [19].

The data on *Saduria entomon* Crustaceans and *Mya arenaria* Bivalves indicated that ^{210}Po is not uniformly distributed in their bodies. Polonium content in the internal organs of these animals decreased in the sequence: hepatopancreas > alimentary track > gills > muscles. It was additionally demonstrated that in the case of fish, the organs directly connected with digestion (intestine, liver, spleen, pyloric caeca) contain much more ^{210}Po as compared to muscles tissue. On the basis of the data on polonium content in fish, it has been established that this nuclide is absorbed by fish through their food. Moreover, ^{210}Po and ^{210}Pb (precursor ^{210}Po) in Baltic organisms do not exist in radioactive equilibrium and higher disequilibrium ($^{210}\text{Po} \geq ^{210}\text{Pb} \geq 1$) is observed in animals [19].

Table 3. ^{210}Po activity concentration and bioconcentration factor (BCF) in Baltic and land organisms in Poland

Organisms	^{210}Po concentration [Bq·kg ⁻¹ d.w.]	BCF × 10 ³
Baltic organisms		
Phytoplankton	41	4.2
Phytoplankton	9	1.8
Zooplankton	126	32
Zoobenthos		
Polychaeta	65	17
Priapulida	53	7.5
Crustaceans	60	25
Bivalves	29	23
– soft tissues	143	37
– shell	8.5	12
Fish		
alimentary track	420	70
muscles	4.5	1.5
Land organisms		
Mushrooms	14	
<i>Boletus edulis</i>	62	
Tobacco	27.5	

More than 30% of ^{210}Po consumed by a statistical Polish inhabitant is the uptake from fish and the annual radioactive dose caused by this fact is 43 μSv [20]. Among analyzed tobacco leaves about 66% of polonium is located in the oldest, over-ground part. The concentration of ^{210}Po in these leaves reaches a value of 136.3 Bq·kg⁻¹ d.w. The observed differences in the polonium distribution in the analyzed leaves indicate that ^{210}Po is generally taken up by tobacco directly from the dry or wet deposition of the radiation fall-out onto the plant. Polonium is highly accumulated in tobacco leaves what means a high risk for cigarette smokers as well. Mean value of polonium activity concentration in tobacco is 27.5 Bq·kg⁻¹ d.w. [30]. Polonium is also consumed with mushrooms, vegetables and fruits and the annual effective dose for mushrooms consumers in Poland is about 37 μSv , and for vegetables and fruits consumers is much lower and is equal to about 8 μSv [16, 24]. The highest value, 62 Bq·kg⁻¹ d.w. of polonium activity in Polish mushrooms, was found in *Boletus edulis*, while the mean value of ^{210}Po activity concentration in Polish mushrooms is 14 Bq·kg⁻¹ d.w. (Table 3). Our results indicate that King Bolate *Boletus edulis* is a good bioindicator for ^{210}Po radioactivity in the land environment [24].

Poland is one of the numerous countries with a very high tobacco consumption. The amount of smokers was calculated to be 10 million people who smoke about 90×10^9 cigarettes per year. The 14 samples of the most often bought cigarette brands which consist of over 70% of total sale in Poland were taken for the analysis of ^{210}Po (Table 4). ^{210}Po content in Polish cigarettes varies to a large extent. The highest contents of polonium were found in the Popularne, Mewa Menthol and Sobieski King-Size, 24.1, 21.5 and 20.1 mBq of ^{210}Po in one cigarette, respectively. The experiments indicate the

Table 4. ^{210}Po content in smoke from 1 cigarette bought in Poland

Cigarettes	^{210}Po content [mBq]
Domestic	
Popularne	22.9
Mewa Menthol	18.3
Golden American	15.1
Marlboro	12.5
Extra Mocne	12.3
Caro Lights	11.5
Radomskie	10.4
Klubowe	8.9
Sobieski Lights	7.0
Sobieski King-Size	6.0
Mocne	5.9
Mars King-Size	3.6
Mars Lights	3.3
Caro	2.7
External	
LM Light	7.2
Mild Seven Light	5.5
LM	4.6
Salem	3.0
Davidoff	2.0

Polish tobacco used to the cigarette production contains much more ^{210}Po comparing with the tobacco used in other countries. Similar ^{210}Po activities were noticed only in cigarettes produced in Turkey and India [28, 31].

Considering that both radionuclides (^{210}Po and ^{210}Pb) exist in cigarette smoke in radioactive equilibrium, it can be calculated that on smoking 2 packs of cigarettes per day, the smoker receives an annual radioactive dose of over 100 μSv . Such a dose is similar to that received in the first two years after the Chernobyl accident for inhabitants of north-eastern Poland as a result of radio-caesium absorption (^{134}Cs and ^{137}Cs) which were equal to about 128 μSv [31]. The radiation dose originating from inhalation of cigarette smoke is much higher than the effective dose from the intake of ^{210}Po and ^{210}Pb with food and drinking water in Poland [16, 20]. Smoking cigarette resulting in the inhalation of ^{210}Po and ^{210}Pb , dioxins, nicotine and polycyclic aromatic hydrocarbons (PAH) is probably the reason of the high incidence of the lung cancers, esophagus and larynx, and others diseases of the respiratory system in the Polish population [33].

Uranium ^{238}U and ^{234}U

The uranium concentrations range from 0.68 to 0.85 $\mu\text{g}\cdot\text{dm}^{-3}$ in the surface samples, but from 0.64 to 1.28 $\mu\text{g}\cdot\text{dm}^{-3}$ in waters of the Vistula river. The mean

Table 5. The mean values of uranium activity concentrations in seawater, drinking water and beer in Poland

Water	^{238}U activity concentration [mBq·dm ⁻³]	$^{234}\text{U}/^{238}\text{U}$ activity ratio
Southern Baltic Sea	8.40	1.18
Drinking water (Gdańsk)	2.80	1.04
Bottled mineral drinking water	0.80	1.00
Beer	4.94	0.72

$^{234}\text{U}/^{238}\text{U}$ activity ratio in Baltic seawater was found to be 1.17, while in Vistula river water it was 1.31. The uranium concentration in seawater increased, but the values of $^{234}\text{U}/^{238}\text{U}$ activity ratio decreased with salinity [19]. Radiochemical analysis of drinking water (Gdańsk agglomeration and mineral bottled water) and beers in Poland indicate low activity values of uranium and they are safe from the radiological point of view (Table 5) [25–27].

Uranium, in contrary to polonium, is not significantly accumulated by Baltic organisms. Marine algae and benthic animals accumulated uranium isotopes to a small extent only. The values of BCF ranged from 0.4 to 82. (Table 6). The mean uranium concentration in phytoplankton was about twice as high as in zooplankton. Only small differences were observed between the uranium levels in benthic organisms. The higher values were found in the Bivalves, the lower in Crustaceans. The fish investigated showed that uranium is not uniformly distributed in their bodies, its concentration increasing in the sequence muscle < skeleton < viscera [19].

The average of $^{234}\text{U}/^{238}\text{U}$ activity ratio in Baltic suspended matter is about 1.0, i.e. similarly to that in sediments. This indicates that a terrigenous material is the main source of uranium in the Baltic sediments. As the $^{234}\text{U}/^{238}\text{U}$ activity ratio in Baltic organisms (1.12–1.15) range is similar to that in seawater (1.17), it can be inferred that the dissolved forms of this element in seawater are a source of uranium in Baltic biota (Fig. 1).

The concentration of uranium in sediments from the southern Baltic increases with depth core, probably indicating the diffusion from sediment to water through interstitial water, where uranium concentration is much higher than in bottom water. The values of $^{234}\text{U}/^{238}\text{U}$ activity ratio in sediments from reduction area of the southern Baltic (Gdańsk Deep and Bornholm Deep) indicate that the reduction process of U(VI) to U(IV) and the removal of autogenic uranium from seawater to sediments constitute only a small part in the Gdańsk Deep [21]. The most important process in geochemical uranium migration in the southern Baltic Sea ecosystem is the diffusion from sediments to bottom water of the terrigenous material and river (Vistula) suspended matter [20].

Plutonium ^{238}Pu , $^{239+240}\text{Pu}$ and ^{241}Pu

The average level of $^{239+240}\text{Pu}$ in Baltic seawater was found to be 4.8 $\mu\text{Bq}\cdot\text{dm}^{-3}$ (Table 7), near 60% of which

Table 6. The average values of ^{238}U concentrations, bioconcentration factor (BCF) and $^{234}\text{U}/^{238}\text{U}$ activity ratio in Baltic organisms

Baltic organisms	^{238}U concentration [Bq·kg ⁻¹ d.w.]	BCF	$^{234}\text{U}/^{238}\text{U}$ activity ratio
Phytoplankton	5.5	45	1.15
Phytobenthos	3.5	48	1.14
Zooplankton	1.3	30	1.15
Zoobenthos			
Crustaceans	1.5	51	1.15
Bivalves	0.9	55	1.15
– soft tissues	4.5	82	
– shell	0.3	30	
Fish	0.05	1.3	1.12
alimentary track	0.64	7.6	
muscles	0.0112	0.4	

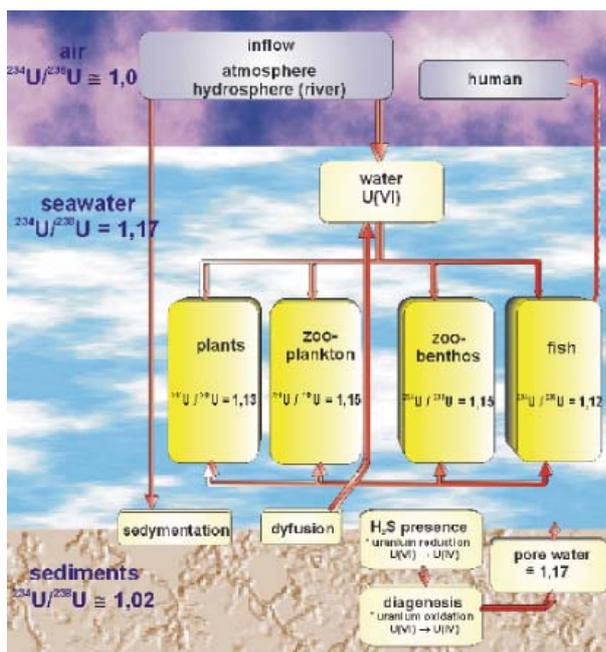
constituted filterable forms ($\leq 0.45 \mu\text{m}$). The concentration of plutonium in the suspended matter was $1.5 \text{ Bq}\cdot\text{kg}^{-1} \text{ d.w.}$, and the distribution coefficient (DC) of plutonium was of the order of 4×10^5 [19].

Plutonium is considerably accumulated in Baltic organisms and BCF values range from 25 to 27×10^3 . The Baltic plants and benthic animals concentrate plutonium isotopes to various extents (Table 7). The $^{239+240}\text{Pu}$ concentrations in the Baltic organisms range from $14 \text{ mBq}\cdot\text{kg}^{-1} \text{ d.w.}$ (fish) to $957 \text{ mBq}\cdot\text{kg}^{-1} \text{ d.w.}$ (Priapulida). In the seaweeds, the highest plutonium concentrations were found in *Pylaiella littoralis* collected in Puck Bay in 1987. In the zoobenthos, plutonium concentrations were higher in Priapulida and Polychaeta and lower in Entomostraca, Malacostraca and Bivalves (soft tissues) [19].

The analysis of $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratios in Baltic flora and fauna show increasing with time Chernobyl plutonium participation, especially in fish. It indicates on plutonium remobilization from sediments to bottom water via benthic organisms and the effect of late inflow

Table 7. The average values of $^{239+240}\text{Pu}$ concentrations and bioconcentration factor (BCF) in Baltic seawater and organisms

Organisms	$^{239+240}\text{Pu}$ concentration [Bq·kg ⁻¹ d.w.]	BCF $\times 10^3$
Baltic seawater	$4.8 \mu\text{Bq}\cdot\text{dm}^{-3}$	
Baltic organisms		
Phytoplankton	0.006	0.6
Phytobenthos	0.118	3.5
Zooplankton	0.004	0.025
Zoobenthos		
Polychaeta	0.169	5.5
Priapulida	0.957	27.0
Crustaceans	0.046	2.7
Bivalves (soft tissues)	0.073	3.2
Fish	0.014	0.9
alimentary track	0.100	2.9
muscles	0.0039	0.25

**Fig. 1.** $^{234}\text{U}/^{238}\text{U}$ activity ratio in Baltic Sea ecosystem.

of plutonium from the Baltic catchment area. It is especially important in the Gdańsk Bay because, as it is shown, this basin is more contaminated with plutonium (Table 8). The sediments of the Gdańsk Bay contain 6.4% of $^{239+240}\text{Pu}$ deposited in sediments of the whole Baltic Sea but the area of Gdańsk Bay constitutes only 1.2% [29].

The main source of $^{239+240}\text{Pu}$ inflow in Gdańsk Bay is river water (Vistula river), which enriches these regions with 78% of total plutonium. The total $^{239+240}\text{Pu}$ amount deposited in the Gdańsk Bay is 1.18 TBq (Fig. 2). Almost the whole plutonium is deposited in sediments. In seawater of the Gdańsk Bay (including suspended matter) there is about 2.33 GBq (0.2% of total amount) and 56% of $^{239+240}\text{Pu}$ is associated with suspended matter. Organisms in the Gdańsk Bay contain 3.81 MBq and from this value 82.1% is deposited in zoobenthos, 13.6% in phytobenthos, 1.6% in phytoplankton, 1.5% in zooplankton and 1.2% in fish [29].

Estimate the impact of Chernobyl accident on Poland territory also ^{241}Pu was determined to. The plutonium sources were remeasured after 16–18 years and the determination of ^{241}Pu in the samples was done

Table 8. $^{239+240}\text{Pu}$ plutonium inventories in the ecosystem of the Gdańsk Bay and the Baltic Sea

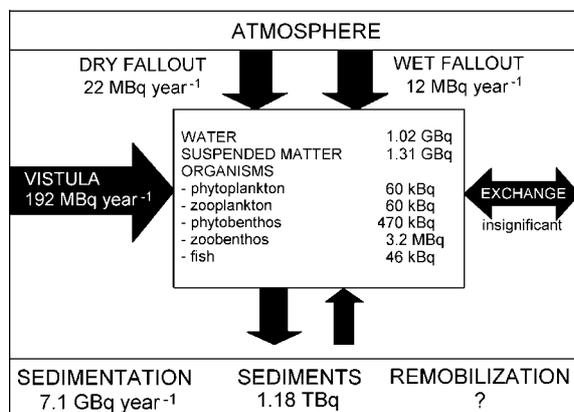
Compartment	Baltic Sea	Gdańsk Bay	
		total	[%]
Sediments [TBq]	15.2–24.2	1.19	6.4
Water [GBq]	200	1.0	0.5
Water and suspended matter [GBq]		2.3	1.2
Organisms [MBq]	a few GBq	3.81	~0.1
Area [km ²]	415,266	4940	1.2
Capacity [km ³]	21,721	291.2	1.3

indirectly by measurements of ^{241}Am ingrowth from β -emitting ^{241}Pu . A comparison of the obtained spectra allowed us to make an estimation of the ^{241}Pu content based on the ingrowth 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:

$$A_{\text{Pu}_0} = 31.3074 \cdot \frac{A_{^{241}\text{Am}} \cdot e^{+\lambda_{\text{Am}} \cdot t}}{(1 - e^{-\lambda_{\text{Pu}} \cdot t})}$$

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

Just after the Chernobyl accident the ^{241}Pu activity concentration in air had increased from a value near 1 mBq·g⁻¹ d.w. to 3643 mBq·g⁻¹ d.w. and then slowly decreased. Its concentration in seawater from the Gdańsk Bay was 0.23 mBq·dm⁻³ and $^{241}\text{Pu}/^{239+240}\text{Pu}$ activity ratio was the same as in the moment of accident and was equal to 140. Plutonium is strongly accumulated in Baltic organisms. Among all analyzed organisms higher ^{241}Pu activity concentrations were found in brown algae *Pylaiella littoralis* (1.01 mBq·g⁻¹ d.w.), Priapulida *Halicryptus sinulosus* (9.20 mBq·g⁻¹ d.w.) and Polychaeta *Antinöella sarsi* (7.71 mBq·g⁻¹ d.w.). The $^{241}\text{Pu}/^{239+240}\text{Pu}$ activity ratio indicates that in a few years after the Chernobyl accident it is too early to estimate the impact of this accident on the Baltic Sea environment [32].

**Fig. 2.** A scheme of plutonium inventories in the Gdańsk Bay.

Conclusions

1. Polonium and plutonium are strongly accumulated by land and Baltic organisms. BCF values for polonium range from 1.5×10^3 to 3.2×10^4 and for plutonium from 25 to 27×10^3 .
2. Polonium and plutonium in land and Baltic organisms are not uniformly distributed:
 - most of the accumulated ^{210}Po and $^{239+240}\text{Pu}$ in the analyzed Baltic invertebrates and fish are distributed in their soft tissues (hepatopancreas, alimentary track),
 - most of ^{210}Po in tobacco plant is located in the oldest leaves.
3. Some organisms are good bioindicators for radioactive contamination of land and the Baltic environment:
 - the brown algae *Pylaiella littoralis* and Priapulida *Halicryptus spinulosus* are very good bioindicators for plutonium contamination of the southern Baltic Sea ecosystem,
 - the mushroom *Boletus edulis* is a very good bioindicator for ^{210}Po in land ecosystems.
4. The impact of Chernobyl plutonium on the Baltic organisms is in 2003 still significant and ranges from 10% to 60%.
5. The sediments of the Gdańsk Bay are relatively enriched in plutonium. The area of the Gdańsk Bay constitutes only 1.2% of the Baltic Sea area, but sediments of this basin contain 6.4% of the total $^{239+240}\text{Pu}$ activity inventory within the Baltic Sea sediments.
6. The radiation dose originating from ^{210}Po and ^{210}Pb inhaled with cigarette smoke is much higher (50–100 μSv) than the effective dose from the intake of these radionuclides with marine food (43 μSv), mushrooms (especially *Boletus edulis* – 37 μSv), vegetables, fruits (8 μSv), as well as drinking water and beers (less than 1 μSv) in Poland.
7. Uranium in comparison to polonium and plutonium is not strongly accumulated by Baltic organisms and BCF values for ^{238}U in biota ranged from 1 to 80.
8. The analysis of $^{234}\text{U}/^{238}\text{U}$ activity ratio indicates the main sources of uranium in Baltic organisms are its dissolved forms in seawater.
9. The most important process in geochemical uranium migration in the southern Baltic ecosystem is the terrigenous material and the river (Vistula river)

suspended matter diffusion from sediment to bottom water through interstitial water.

Acknowledgment The authors would like to thank the State Committee for Scientific Research (KBN) for partial financial support of this work under Grants BW/8000-5-0284-5, BW/8000-5-0309-6, BW/8000-5-0412-6 and DS/8210-4-0086-6.

References

- Aarkrog A (1988) The radiological impact of the Chernobyl debris compared with that from nuclear weapon fallout. *J Environ Radioact* 6:151–162
- Browne E, Firestone EB (1986) Table of radioactive isotopes. John Wiley and Sons, New York
- Coughtrey PJ, Jackson D, Jones CH, Kane P, Thorne MC (1984) Radionuclides distribution and transport in terrestrial and aquatic ecosystem, a critical review of data. AA Balkema, Rotterdam
- Danielsen EF (1981) Trajectories of the Mount St. Helen plume. *Science* 221:891–921
- Hardy EP, Krey PW, Volchok HL (1973) Global inventory and distribution of fallout plutonium. *Nature* 241:444–445
- Holm E, Oregoni B, Vas D, Pettersson H, Rioseco J, Nilsson U (1990) Nickel-63: radiochemical separation and measurement with an ion implanted silicon detector. *J Radioanal Nucl Chem* 138:111–116
- Holm E, Roos P, Skwarzec B (1992) Radiochemical studies of fallout ^{63}Ni . *Appl Radiat Isot* 43:371–376
- Jaworowski Z (1969) Radioactive lead in the environment and in the human body. *At Energ Rev* 7:3–45
- Jaworowski Z, Bilkiewicz J, Kownacka L, Włodek S (1972) Artificial sources of natural radionuclides in environment. In: Proc of the 2nd Int Conf: The natural radiation environment, 5 August 1972, Houston, Texas. CONF-720805-P2. Vol. II, pp 809–818
- Ku TL, Knauss KG, Mathieu GG (1977) Uranium in open ocean: concentration and isotopic composition. *Deep Sea Research* 24:1005–1017
- Lambert G, Buisson A, Sanak J, Ardonin B (1979) Modification of the atmospheric polonium-210 to lead-210 ratio by volcanic emissions. *Journal Geophysics Research* 84:6980–6986
- Langmuir D (1978) Uranium solution-mineral equilibria at low temperature with applications to sedimentary ore deposits. *Geochim Cosmochim Acta* 42:547–569
- Martin JM, Nijampurkar V, Salvadori F (1978) Uranium and thorium isotope behavior in estuarine system. In: Goldberg ED (ed.) *Biochemistry of estuarine sediments*. UNESCO, pp 111–127
- McDonald P, Fowler SW, Heyrand MW, Baxter MS (1986) Polonium-210 in mussels and its implications for environmental alpha-autoradiography. *J Environ Radioact* 3:293–303
- Moore HE, Martell EA, Poet SE (1976) Sources of polonium-210 in the atmosphere. *Environ Sci Technol* 10:586–591
- Pietrzak-Flis Z, Chrzanowska S, Dembińska S (1997) Intake of ^{226}Ra , ^{210}Pb and ^{210}Po with food in Poland. *Sci Total Environ* 203:157–165
- Rangarajan C, Gopalakrishnam S, Eapen CD (1976) Global variation of ^{210}Po in surface air and precipitation. In: Hardy EP (ed.) *Health and safety laboratory environmental quarterly*. HASL-298. US EPA, New York, pp 163–182
- Skwarzec B (1995) Polonium, uranium and plutonium in ecosystem of the southern Baltic Sea. *Rozprawy i Monografie no. 6*. Instytut Oceanologii PAN, Sopot (in Polish)
- Skwarzec B (1997) Polonium, uranium and plutonium in the southern Baltic Sea. *Ambio* 26;2:113–117
- Skwarzec B (2002) Environmental radiochemistry and radiological protection. Wydawnictwo DJ, Gdańsk (in Polish)
- Skwarzec B, Boryło A, Strumińska DI (2004) Activity disequilibrium between ^{234}U and ^{238}U isotopes in southern Baltic. *Water, Air Soil Pollut* 159:165–173
- Skwarzec B, Holm E, Roos P, Pempkowiak J (1994) Nickel-63 in Baltic fish and sediments. *Appl Radiat Isot* 54:609–611
- Skwarzec B, Holm E, Strumińska DI (2001) Radioanalytical determination of ^{55}Fe and ^{63}Ni in the environmental samples. *Chem Anal (Warsaw)* 46:23–30
- Skwarzec B, Jakusik A (2003) ^{210}Po bioaccumulation by mushrooms from Poland. *J Environ Monit* 5:791–794
- Skwarzec B, Strumińska DI, Boryło A (2001) The radionuclides ^{234}U , ^{238}U and ^{210}Po in drinking water in Gdańsk agglomeration (Poland). *J Radioanal Nucl Chem* 250;2:315–318
- Skwarzec B, Strumińska DI, Boryło A (2003) Radionuclides of ^{210}Po , ^{234}U and ^{238}U in drinking bottled water in Poland. *J Radioanal Nucl Chem* 256:361–364
- Skwarzec B, Strumińska DI, Boryło A, Falandysz J (2004) Intake of radionuclides of ^{234}U and ^{238}U with beer in Poland. *J Radioanal Nucl Chem* 261;3:661–663
- Skwarzec B, Strumińska DI, Boryło A, Ulatowski J (2001) Polonium ^{210}Po in cigarettes produced in Poland. *J Environ Sci Health A* 36:23–30
- Skwarzec B, Strumińska DI, Prucnal M (2003) Estimates of $^{239+240}\text{Pu}$ inventories in Gdańsk Bay and Gdańsk Basin. *J Environ Radioact* 70:237–252
- Skwarzec B, Strumińska DI, Ulatowski J, Gołębiowski M (2001) Determination and distribution of polonium ^{210}Po in tobacco plant from Poland. *J Radioanal Nucl Chem* 250;2:319–322
- Skwarzec B, Ulatowski J, Strumińska DI, Boryło A (2001) Inhalation of ^{210}Po and ^{210}Pb from cigarette smoking in Poland. *J Environ Radioact* 57:221–230
- Strumińska DI, Skwarzec B (2006) Plutonium ^{241}Pu concentrations in southern Baltic Sea ecosystem. *J Radioanal Nucl Chem* 268;1:59–63
- Zatoński W (1996) Development of health situation in Poland after 1988 year. Centrum Onkologii, Instytut im. M. Skłodowskiej-Curie, Zakład Epidemiologii i Prewencji Nowotworów, Warsaw (in Polish)