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POLONIUM, URANIUM AND PLUTONIUM RADIONUCLIDES IN AQAUTIC ENVIRONMENT OF POLAND AND SOUTHERN BALTIC

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Abstract

In the paper were presented the results of study for determination of natural (polonium ^{210}Po, uranium ^{234}U and ^{238}U) and artificial (plutonium ^{238}Pu, $^{239+240}$Pu and ^{241}Pu) alpha radionuclides in aquatic environment of Poland and southern Baltic Sea as well as the recognition of their accumulation in marine trophic chain. The obtained results indicated that Vistula and Odra as well as Rega, Parsęta and Słupia are important sources of analyzed radionuclides in southern Baltic Sea. Total annual runoff of polonium, uranium and plutonium from Vistula, Odra and Pomeranian rivers to the Baltic Sea was calculated as about 95 GBq of ^{210}Po, 750 GBq of $^{234+238}$U and 160 MBq of $^{238+239+240}$Pu.

Investigation on the polonium ^{210}Po, uranium ^{234}U and ^{238}U, as well as and plutonium ^{238}Pu, $^{239+240}$Pu and ^{241}Pu. concentration in Baltic biota revealed that these radionuclides, especially polonium and plutonium, are strongly accumulated by some species. The results indicate that the Baltic organisms accumulate polonium and plutonium from environment and the bioconcentration factors (BCF) range from 25 to 27 000. 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 via intersticial water to bottom water. The values of ^{234}U/^{238}U activity ratio in the sediments indicated that the possible reduction process of U(VI) to U(IV) and the removing of autogenic uranium from seawater to sediments in the Gdańsk Deep and Bornholm Deep constitutes a small part only.

Key words: ^{210}Po, ^{234}U, ^{238}U, $^{239+240}$Pu, ^{241}Pu radionuclides, Vistula, Odra, Pomeranian rivers, inflow, accumulation, Poland, Southern Baltic Sea

INTRODUCTION

Amongst radionuclides, alpha emitters play a significant role in state radiological effects connected with accumulation of these radionuclides in organisms because, there are a very strong radiotoxic elements (Skwarzec 1995). In particularly natural polonium ^{210}Po, uranium ^{234}U and ^{238}U as well as artificial plutonium $^{239+240}$Pu are very important for radiological protection. Radionuclides existing in the aquatic environment, both natural and artificial, are accumulated in plants and animals and transfered through the trophic chain.

Polonium ^{210}Po is radionuclide, which belongs to a natural uranium series starting from ^{238}U. The main natural source of polonium in environment is the radioactive decay of uranium ^{238}U, radium ^{226}Ra and radon ^{222}Rn. The total amount of airborne ^{210}Po depends directly on the amount of ^{210}Pb supplied and formed in the atmosphere (Jaworowski 1969, Jaworowski and Kownacka 1976, Skwarzec 1995). Additional amounts of ^{210}Po in the atmosphere are emitted directly from the ocean surface and Earth surface, as a result of forest fires (Moore et al. 1976) and volcanic eruption (Danielsen 1981, Lambert et al. 1979). Polonium isotopes belong to the most radiotoxic nuclide to human being (McDonald et al. 1986). Natural concentration of polonium in environment can be enhanced due to human activity (industry, fossil fuel combustion, phosphate fertilizers in agriculture, domestic and industrial sewage), (Daish et al. 2005). ^{210}Po concentration in the atmosphere ranges from 13 to 240 $\mu Bq \cdot m^{-3}$ and its deposition varies between 10 and 500 $Bq \cdot m^{-2} \cdot year^{-1}$ (Carvalho 1997, Skwarzec 2002). The main area where polonium is retained is aquatic environment, where ^{210}Po is accumulated by marine plants and animals. In aquatic environment polonium exists both in dissolved forms and connected with suspended matter. In the estuary residence time of dissolved ^{210}Po was estimated to be 30 days. Remobilization from sediment of polonium in marine environment is faster in oxygen-free conditions (Carvalho 1997). Constant deposition of ^{210}Po on large impoundments could lead to higher radioactivity buildup than in running waters carrying radioactivity away (Shaheed et al. 1997).

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 (Skwarzec 1995). The average concentration of uranium in the Earth's crust is $4 \cdot 10^{-4}$% of weight. Naturally occurring uranium contains three alpha emitting radionuclides: ^{238}U (99.2745%), ^{235}U (0.7200%) and ^{234}U (0.0055%). Isotope ^{238}U is the parent nuclide of the radioactive decay uranium-radium series. In the rocks, ^{234}U and ^{238}U isotopes are in generally in radioactive equilibrium, but in natural waters (rivers, seawaters) activity of ^{234}U is slightly higher in comparison to activity of ^{238}U (activity ratio $^{234}U/^{238}U$ is about 1.2-1.4) (Skwarzec et al. 2004). The uranium from rocks, soils and sediments is rinsing by water and next is inflowing to rivers. The particles containing uranium are also precipitated from atmosphere as a result of rocks erosion and resuspension of soil (Ku et al. 1977, Skwarzec 1995). In seawater and river water, in oxidation conditions, natural uranium exists predominantly in the dissolved form of uranyl carbonate anion $[UO_2(CO_3)_2]^{2-}$ and $[UO_2(CO_3)_3]^{4-}$ (Langmuir 1978). Uranium concentration in natural river varies linearly with salinity (Skwarzec 1995).

Plutonium radionuclides (^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Pu) belong to the group of caused by human activity, artificial radionuclides. These nuclides are important from the radiological point of view due to their high radiotoxicity, long physical half-life, high chemical reactivity and long residence in biological system (Coughtrey et al. 1984). The principal source of plutonium radionuclides in the environment is atmospheric fallout from nuclear weapon tests (Hardy et al. 1973, Skwarzec 1995). Wet and dry atmospheric fallout from nuclear weapon tests is one of the most important sources of plutonium in Poland and 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 another source of plutonium isotopes, the Charnobyl-originated radioactive debris had to be taken into account (Aarkrog 1988).

Vistula and Odra Rivers and its basin

Over 95% of the water supply in Poland originates from atmospheric precipitation. The total annual runoff from Poland to the Baltic Sea is approximately 60,000 m^3 per year, of which 50% are discharges by the Vistula River and about 34% of which are discharged by the Odra River (Makinia et al. 1996). The area of the Vistula watershed is 194,424 km^2 whereof 168,699 km^2 covers Polish territory and represents 86.6% of Vistula catchment area (Ochrona środowiska... 2005). The area of the Odra River watershed is 118,861 km^2, whereof 106,050 km^2 belongs to the Polish territory and represents 89.2% of the Odra catchments area. The summary areas of Vistula and Odra catchments content more than 80% of Polish territory global area (Ochrona środowiska... 2005). Annually Vistula and Odra rivers transport about 2.1 mln m^3 water with various fluvial material, where 1.8 mln ton is salt from coal mines of the Upper Silesian Coal Factory. This salt contains considerable amount of natural radionuclides from uranium decay series (^{238}U, ^{234}U, ^{226}Ra, ^{210}Pb and ^{210}Po), (Flues et al. 2002). Moreover, phosphate fertilizers have large influence on polonium and uranium concentrations (Boryło et al. 2009). In the area of Poland (31.2 mln ha), the cultivated land area is about 16.6 mln ha, where about 18.7 $kg \cdot ha^{-1}$ (about $3.1 \cdot 10^5$ tons) of phosphate fertilizers was used by farmers (Ochrona środowiska... 2005, Użytkowanie gruntów... 2005).

Southern Baltic Sea

The Baltic Sea is a young postglacial inland sea, with its drainage basin over four times bigger than its sea area. The drainage basin – densely inhabited and urbanized – is used mainly for agricultural and industrial purposes. The Baltic Sea is connected to the North Sea (Atlantic Ocean) via the Kattegat and narrow inlets of the Belt Sea and Sund – the transition zone. The Southern Baltic is a part of Baltic Proper. Several regions are distinguished based on the bottom topography: Arkona Basin, Pomeranian Bay, Bornholm Basin, Słupsk Narrow and Gdańsk Bay (Szefer 2002). Atmospheric precipitation and inflow rivers waters are the global sources of chemical substances entering the Baltic Sea (Skwarzec 2002).

The aim of this work was determination of natural (polonium ^{210}Po, uranium ^{234}U and ^{238}U) and artificial (plutonium ^{238}Pu, $^{239+240}Pu$ and ^{241}Pu) alpha radionuclides in aquatic environment of Poland and Southern Baltic as well as the recognition of their accumulation in marine trophic chain. Moreover on the basis of obtained results should be calculated seasonal and annul runoff of polonium, uranium and plutonium from Vistula, Odra and Pomeranian rivers to the Southern Baltic.

MATERIALS AND METHODS

Different environmental samples, like seawater and river water, suspension, sediments and marine organisms were taken between 1980 and 2005 from Polish rivers (Vistula and Odra as well as Rega, Pasłęka and Słupia Rivers catchments) and the Southern Baltic Sea areas. The places where the samples of surface river water where taken are presented in Figure 1. In laboratory, the analyzed samples were prepared immediately after their delivery for radiochemical procedure for determination of polonium ^{210}Po, uranium ^{234}U and ^{238}U and plutonium $^{239+240}$Pu. Before radiochemical analysis, to each sample about 50 mBq of ^{209}Po, 100 mBq of ^{232}U and 5 mBq of ^{242}Pu were added as a yield tracers. Polonium, uranium and plutonium in natural water (river and sea water samples) were coprecipitated with manganese dioxide. The biological and sediment materials were mineralized using concentrated nitric and hydrochloric acids. Polonium was electrodeposited on silver discs but uranium and plutonium, after separation on Dowex anion-exchange resin, were electrolyzed on stainless steel discs (Skwarzec 1997a, 2009). The activity of ^{210}Po, ^{234}U, ^{238}U and $^{239+240}$Pu was measured using alpha spectrometry equipped with semiconductor silicon detectors and 300 mm^2 active surface barrier (Canberra-Packard, USA). The determination of ^{241}Pu (emitter β^-) in the analyzed samples was done indirectly by alpha measurements. The plutonium sources were remeasured after 16-18 years and determination of ^{241}Pu in the samples was done by measurements of ^{241}Am ingrowth from β-emitting ^{241}Pu. A comparison of the obtained spectra allowed us to make the 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 Chernobyl accident. The calculation of the ^{241}Pu activity was based on the following formula from:

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

where:

A_{Pu0}	–	^{241}Pu activity in the time of sampling
31.3074	–	constant value ($\lambda_{Pu}/\lambda_{Am}$)
$A_{^{241}Am}$	–	^{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)

The least detectable activity (LDA) was 0.3 mBq for 2 days counting time for polonium and uranium activity, and 0.1 mBq for 10 days counting time for plutonium activity. Polonium and uranium samples were measured for 1-5 days, but plutonium preparates were measured between 10-30 days. The accuracy and precision of the radiochemical methods were estimated by contribution in international laboratory exercises and using International Atomic Energy Agency reference materials and estimated at less than 10%. The polonium and uranium yield in analyzed sam-

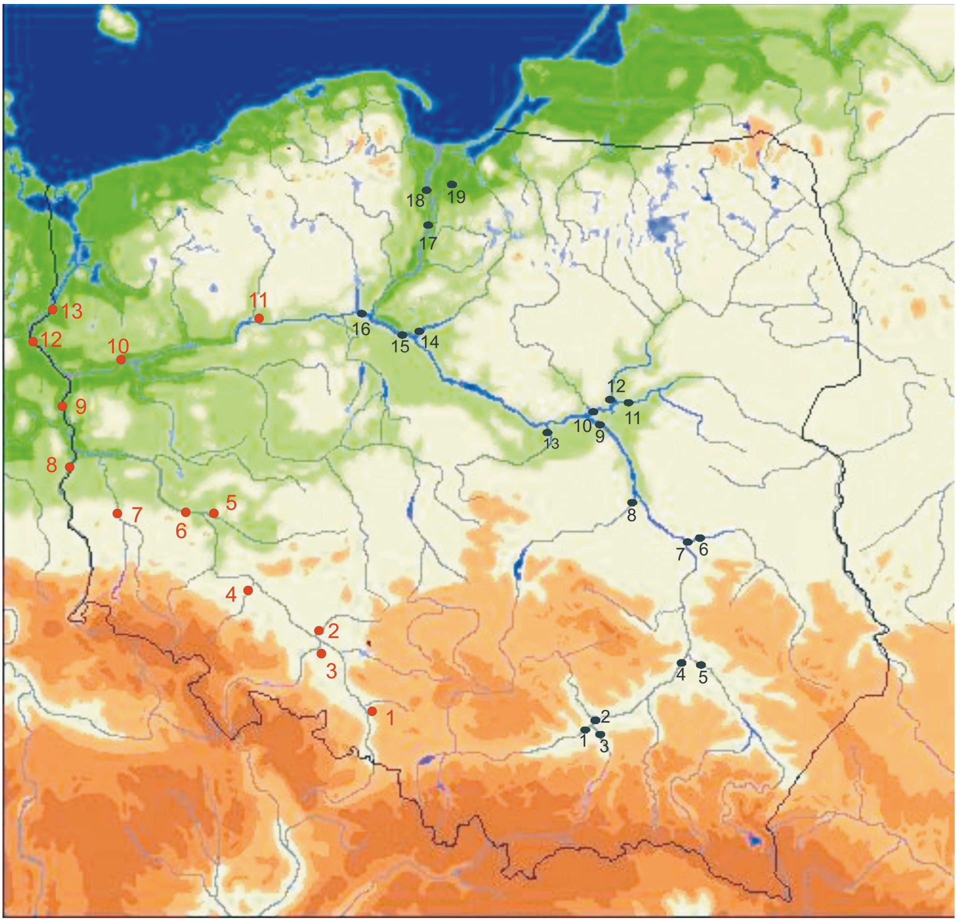

Fig. 1. Place of taking water samples
Vistula drainage: 1. Kraków, 2. Nida, 3. Dunajec, 4. Sandomierz, 5. San, 6. Wieprz, 7. Dęblin, 8. Pilica, 9. Warszawa, 10. Bug with Narew, 11. Bug, 12. Narew, 13. Bzura, 14. Drwęca, 15. Toruń, 16. Brda, 17. Grudziądz, 18. Leniwka-Kiezmark, 19. Nogat-Malbork
Odra drainage: 1. Chałupki, 2. Mała Panew, 2. Nysa Kłodzka, 4. Bystrzyca, 5. Barycz, 6. Głogów, 7. Bóbr, 8. Nysa Łużycka, 9. Słubice, 10. Warta, 11. Noteć, 12. Gozdowie, 13. Widuchowa
Pomerania rivers: 1. Rega, 2. Parsęta, 3. Słupia

ples ranged from 70% to 96%, but for plutonium was between 40-65%. (Skwarzec 1997a, 2009). The results of ^{210}Po, ^{238}U and $^{239+240}$Pu concentrations are given with standard deviation (SD) calculated for 95% confidence internals.

RESULTS AND DISCUSSION

The Vistula River catchment area

The concentration values of ^{210}Po, ^{238}U and $^{239+240}$Pu in water samples from the Vistula River and its tributaries are presented in Table 1. The obtained data indicated that we observed difference of analyzed radionuclides in dependence from seasons and place of taking samples. Generally, the polonium and uranium concentrations are higher in spring in comparison to winter. The highest value of ^{210}Po concentration in the Vistula River was measured in water sample from Dęblin (5.98±0.03 Bq·m^{-3}), but the lowest from Warszawa (1.15±0.05 Bq·m^{-3}). Among the Vistula tributaries maximum value of ^{210}Po concentration was observed in Wieprz (9.80±0.02 Bq·m^{-3}), but minimum in Nida (0.59±0.02 Bq·m^{-3}), (Skwarzec and Jahnz 2007). In case of uranium, the highest value of ^{228}U concentration in the Vistula River was measured in water sample from Malbork (12.45±0.21 Bq·m^{-3}), but the lowest from Dęblin (1.55±0.07 Bq·m^{-3}). Amongst the Vistula tributaries the highest value of ^{238}U concentration was observed in Bug (20.45±0.75 Bq·m^{-3}), but the lowest in Brda (1.18±0.04 Bq·m^{-3}). In the analyzed Vistula River water samples was observed radioactive disequilibrium between concentration of uranium isotopes and the value of ^{234}U/^{238}U activity ratio ranged between 1.00 (in Bug with Narew) and 1.74 (in Vistula), (Jahnz 2007). In case of plutonium the highest value of $^{239+240}$Pu concentration in the Vistula River was measured in Dęblin (8.74±1.19 mBq·m^{-3}), but the lowest in Kraków (1.03±0.22 mBq·m^{-3}). Amongst the Vistula River tributaries, Drwęca was characterized by maximum value of plutonium concentration (29.35±4.08 mBq·m^{-3}), but in Wieprz the $^{239+240}$Pu concentration was minimum (1.14±0.38 mBq·m^{-3}). The analysis of results for water samples taken in winter season indicated that the highest $^{239+240}$Pu concentration was found in water from Sandomierz (5.84±0.95 mBq·m^{-3}), but the lowest from Warszawa (1.88±0.30 mBq·m^{-3}) and Kiezmark (1.89±0.21 mBq·m^{-3}). Amongst Vistula tributaries the highest $^{239+240}$Pu concentration was noticed in San (14.80±4.70 mBq·m^{-3}) and Pilica (12.20±1.80 mBq·m^{-3}), but the lowest in Dunajec (2.44±0.51 mBq·m^{-3}). During the spring season the highest plutonium concentration was noticed in Dęblin (8.74±1.19 mBq·m^{-3}) and Malbork (5.30±0.94 mBq·m^{-3}), but the lowest in Warszawa (1.78±0.40 mBq·m^{-3}). $^{239+240}$Pu concentration is highly varied in all analyzed Vistula River tributaries, from 3.21±0.61 mBq·m^{-3} in Bug with Narew to 12.07±4.56 mBq·m^{-3} in Brda. The summer season was characterized by the lowest plutonium concentrations amongst all analyzed quarters. In this season the highest $^{239+240}$Pu concentration was found in Dęblin (3.22±0.44 mBq·m^{-3}), but the lowest in water from Kraków (1.03±0.22 mBq·m^{-3}). Amongst Vistula tributaries the highest plutonium concentration was noticed in Pilica (6.56±1.37 mBq·m^{-3}) and Brda (6.30±2.57 mBq·m^{-3}), but the lowest was found in Wieprz (1.14±0.38 mBq·m^{-3}). Autumn season was very rich in rains what causes higher plutonium concentrations in analyzed water samples (Tab. 1). Along the Vistula River the highest plutonium concentration was noticed in Warszawa and Malbork (7.73±0.91 and 7.12±0.70 mBq·m^{-3} respec-

Table 1

Average concentration of ^{210}Po, ^{238}U and $^{239+240}Pu$ in the Vistula River waters and its tributaries

Sampling location	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn
	^{210}Po±SD [Bq·m^{-3}]				^{238}U ± SD [Bq·m^{-3}]				$^{239+240}Pu$ ± SD [mBq·m^{-3}]			
the Vistula River												
Kraków	1.28 ± 0.03	3.03 ± 0.03	1.44 ± 0.05	2.26 ± 0.03	7.03 ± 0.12	4.08 ± 0.11	3.56 ± 0.19	6.34 ± 0.47	3.07 ± 0.49	2.68 ± 0.54	1.03 ± 0.22	2.13 ± 0.39
Sandomierz	1.33 ± 0.03	2.28 ± 0.04	1.68 ± 0.05	2.48 ± 0.03	1.91 ± 0.06	4.70 ± 0.12	4.15 ± 0.09	10.77 ± 0.40	5.84 ± 0.95	4.74 ± 0.51	1.92 ± 0.26	1.64 ± 0.34
Deblin	1.22 ± 0.03	5.98 ± 0.03	2.37 ± 0.03	3.28 ± 0.02	1.55 ± 0.07	5.26 ± 0.17	5.53 ± 0.15	9.08 ± 0.15	4.37 ± 0.93	8.74 ± 1.19	3.22 ± 0.44	3.46 ± 0.44
Warszawa	1.15 ± 0.05	3.49 ± 0.04	2.68 ± 0.05	3.93 ± 0.03	5.66 ± 0.08	6.73 ± 0.14	5.76 ± 0.23	8.64 ± 0.54	1.88 ± 0.21	1.78 ± 0.40	3.10 ± 0.47	7.73 ± 0.91
Torun	2.72 ± 0.04	2.22 ± 0.05	1.67 ± 0.06	1.89 ± 0.05	7.34 ± 0.16	6.32 ± 0.15	5.31 ± 0.16	6.52 ± 0.43	4.39 ± 0.42	2.49 ± 0.32	2.75 ± 0.92	3.96 ± 0.36
Grudziadz	1.98 ± 0.06	3.29 ± 0.03	2.60 ± 0.04	1.51 ± 0.04	8.93 ± 0.29	6.69 ± 0.15	4.24 ± 0.14	9.88 ± 047	2.37 ± 0.14	3.10 ± 0.34	1.11 ± 0.19	2.29 ± 0.94
Malbork (Nogat)	2.17 ± 0.04	3.21 ± 0.03	3.18 ± 0.04	4.13 ± 0.03	12.45 ± 0.21	10.77 ± 0.17	7.60 ± 0.21	15.12 ± 0.23	2.06 ± 0.22	5.30 ± 0.94	2.78 ± 0.30	7.12 ± 0.70
Kiezmark (Leniwka)	2.36 ± 0.05	3.49 ± 0.03	1.59 ± 0.06	1.88 ± 0.04	9.47 ± 0.18	8.63 ± 0.27	6.36 ± 0.15	9.26 ± 0.20	1.89 ± 0.30	4.70 ± 1.02	1.25 ± 0.26	4.47 ± 0.35
Vistula tributaries												
Nida	0.59 ± 0.02	5.30 ± 0.03	3.63 ± 0.05	1.89 ± 0.04	4.01 ± 0.05	10.08 ± 0.27	7.05 ± 0.30	9.12 ± 0.32	3.26 ± 1.15	8.11 ± 3.31	1.88 ± 0.52	6.21 ± 3.58
Dunajec	1.88 ± 0.04	4.36 ± 0.04	1.07 ± 0.08	1.65 ± 0.03	5.58 ± 0.15	3.96 ± 0.09	3.01 ± 0.13	5.94 ± 0.19	2.44 ± 0.51	4.43 ± 0.61	1.54 ± 0.45	4.83 ± 1.83
San	0.95 ± 0.04	3.43 ± 0.04	1.36 ± 0.06	4.23 ± 0.02	6.61 ± 0.20	7.10 ± 0.33	4.51 ± 0.05	7.27 ± 0.13	14.8 ± 4.70	11.38 ± 2.48	4.57 ± 2.28	4.57 ± 0.57
Wieprz	5.46 ± 0.07	9.80 ± 0.02	0.49 ± 0.09	3.87 ± 0.04	3.66 ± 0.11	6.90 ± 0.23	3.46 ± 0.17	9.82 ± 0.23	8.69 ± 1.23	8.40 ± 1.68	1.14 ± 0.38	6.83 ± 1.25
Pilica	1.96 ± 0.06	3.63 ± 0.03	4.02 ± 0.02	7.65 ± 0.03	2.01 ± 0.10	5.12 ± 0.24	6.10 ± 0.12	11.95 ± 0.21	12.20 ± 1.80	6.25 ± 1.43	6.56 ± 1.37	8.74 ± 2.34
Narew (Pultusk)	1.98 ± 0.07	1.20 ± 0.12	2.25 ± 0.04	8.32 ± 0.02	7.67 ± 0.57	11.35 ± 0.39	4.21 ± 0.04	9.90 ± 0.22	3.94 ± 0.82	5.89 ± 1.08	3.37 ± 0.22	14.48 ± 1.26
Bug	1.47 ± 0.06	3.11 ± 0.01	4.64 ± 0.02	5.39 ± 0.04	5.69 ± 0.24	20.45 ± 0.75	8.68 ±0.54	6.41 ± 0.48	4.94 ± 1.03	5.32 ± 0.77	5.73 ± 0.92	9.63 ± 2.33
Bug with Narew (NDM)	3.12 ± 0.08	4.76 ± 0.03	1.92 ± 0.05	7.13 ± 0.02	6.48 ± 0.07	15.90 ± 0.35	5.42 ± 0.09	6.07 ± 0.36	3.16 ± 0.29	3.21 ± 0.61	2.46 ± 0.54	3.80 ± 1.01
Drweca	1.86 ± 0.05	1.45 ± 0.04	0.81 ± 0.04	4.48 ± 0.02	1.37 ± 0.04	10.11 ± 0.34	5.28 ± 0.11	11.12 ± 0.17	3.48 ± 0.68	6.00 ± 1.31	2.50 ± 0.67	29.35 ± 4.08
Brda	2.18 ± 0.06	1.77 ± 0.05	2.47 ± 0.05	2.90 ± 0.01	1.18 ± 0.04	3.58 ± 0.08	3.34 ± 0.06	7.22 ± 0.23	5.90 ± 0.88	12.07 ± 4.56	6.30 ± 2.57	7.68 ± 1.86
Bzura	4.11 ± 0.03	5.78 ± 0.04	5.30 ± 0.02	8.93 ± 0.03	9.36 ± 0.68	13.23 ± 0.43	6.63 ± 0.12	10.13 ± 0.23	3.40 ± 0.55	5.23 ± 2.13	6.08 ± 1.43	8.54± 1.74

tively), but the lowest in Sandomierz (1.64±0.34 mBq·m^{-3}). $^{239+240}$Pu concentrations are highly varied in all analyzed tributaries and varied from 3.80±1.01 mBq·m^{-3} in Bug with Narew to 29.35±4.08 mBq·m^{-3} in Drwęca.
On the basis of average seasonal polonium, uranium and plutonium concentration in the Vistula River and its tributaries, were calculated average annual values of these radioelements i concentration n analyzed samples and the results were presented in Table 2. The highest value of ^{210}Po, ^{238}U and $^{239+240}$Pu concentrations appear in: Dęblin for polonium (3.21±0.04 Bq·m^{-3}) and also plutonium (4.95±0.75 mBq·m^{-3})

Table 2

Annual concentration of ^{210}Po, ^{238}U and $^{239+240}$Pu in the Vistula River waters and its tributaries

Sampling location	^{210}Po	^{238}U	$^{239+240}$Pu
	Bq·m^{-3}	Bq·m^{-3}	mBq·m^{-3}
the Vistula River			
Kraków	2.00±0.03	5.25±0.22	2.23±0.41
Sandomierz	1.94±0.03	5.38±0.17	3.54±0.52
Dęblin	3.21±0.04	5.36±0.14	4.95±0.75
Warszawa	2.81±0.04	6.70±0.25	3.62±0.55
Toruń	2.21±0.05	6.37±0.23	3.40±0.76
Grudziądz	2.35±0.04	7.44±0.26	2.22±0.40
Malbork (Nogat)	3.17±0.05	11.49±0.21	4.32±0.54
Kiezmark (Leniwka)	2.33±0.04	8.43±0.20	3.08±0.54
Vistula tributaries			
Nida	2.85±0.03	7.57±0.24	4.87±2.78
Dunajec	2.24±0.05	4.62±0.14	3.31±0.85
San	2.49±0.04	6.37±0.18	8.83±2.98
Wieprz	4.91±0.05	6.71±0.19	6.27±1.17
Pilica	4.32±0.03	6.30±0.17	8.44±1.74
Narew (Pułtusk)	3.44±0.04	8.28±0.31	6.92±1.04
Bug	3.65±0.03	10.31±0.19	5.90±1.26
Bug with Narew (DM)	4.23±0.04	9.43±0.18	3.16±0.67
Drwęca	2.15±0.04	6.97±0.17	10.33±1.69
Brda	2.33±0.04	3.83±0.10	7.99±2.47
Bzura	6.03±0.03	9.83±0.37	5.81±1.46

and Malbork for uranium (11.49±0.21 Bq·m^{-3}). The lowest concentration values of analyzed radionuclides were in the Vistula River water samples taken in Sandomierz (1.94±0.03 Bq·m^{-3} for ^{210}Po), in Kraków (5.25±0.22 Bq·m^{-3} for ^{238}U) and in Grudziądz and Kraków (2.22±0.40 and 2.23±0.41 mBq·m^{-3}, respectively, for $^{239+240}$Pu). Amongst Vistula tributaries the highest ^{210}Po, ^{238}U and $^{239+240}$Pu concentration was found in Bzura (6.03±0.03 Bq·m^{-3}), Bug (10.31±0.19 Bq·m^{-3}) and Drwęca (10.33±1.69 mBq·m^{-3}) respectively, the lowest in Drwęca (2.15±0.04 Bq·m^{-3}), Brda (3.83±0.10 Bq·m^{-3}) and Narew with Bug (3.16±0.67 mBq·m^{-3}) respectively.

Table 3

Seasonal and annual runoff of ^{210}Po from the Vistula River drainage

Sampling location	Winter	Spring	Summer	Autumn	All year [GBq·year^{-1}]
	[GBq·quarter^{-1}]				
the Vistula River					
Kraków	0.66	1.94	0.22	0.78	3.60
Sandomierz	2.18	10.38	1.66	2.23	16.45
Dęblin	4.48	37.46	4.63	5.94	52.51
Warszawa	4.02	23.08	4.32	10.53	41.95
Toruń	15.08	30.95	7.66	4.61	58.30
Grudziądz	16.96	34.26	6.82	3.88	61.92
Malbork	0.36	0.54	0.54	0.70	2.14
Kiezmark	19.67	40.82	4.31	6.77	71.57
Vistula tributaries					
Nida	0.08	0.87	0.37	0.11	1.43
Dunajec	1.43	9.29	0.41	0.57	11.70
San	0.98	7.09	0.46	2.16	10.69
Wieprz	1.94	3.08	0.07	0.80	5.89
Pilica	0.88	1.88	0.67	2.05	5.48
Narew (Pułtusk)	1.51	1.70	0.64	7.47	11.32
Bug	1.05	7.81	1.32	3.53	13.71
Bug with Narew	7.59	16.12	1.19	9.63	34.53
Drwęca	0.46	0.37	0.10	1.04	1.97
Brda	0.55	0.38	0.29	0.62	1.84
Bzura	0.89	1.28	0.25	0.84	3.26

On the basis of average seasonal and annual flow of the Vistula River and its tributaries were calculated the seasonal and annual runoff of polonium, uranium and plutonium from Vistula drainage – the results are presented in Tables 3, 4 and 5. The values of ^{210}Po seasonal flow, for the Vistula River and their tributaries, were the highest in Vistula delta area (the Leniwka River in Kiezmark – from 4.31 GBq·quarter^{-1} in summer to 40.82 GBq·quarter^{-1} in spring). Polonium flow with the Nogat River (Malbork) ranged from 0.36 GBq·quarter^{-1} in winter to 0.70 GBq·quarter^{-1} in autumn. Annual flow of ^{210}Po in both Vistula delta rivers was 71.57 GBq·year^{-1} in Kiezmark (Leniwka) and 2.14 GBq·year^{-1} in Malbork (Nogat).

Table 4

Seasonal and annual runoff of ^{238}U from the Vistula River drainage

Sampling location	Winter	Spring	Summer	Autumn	All year [GBq·year^{-1}]
	[GBq·quarter^{-1}]				
the Vistula River					
Kraków	3.7	2.6	0.5	0.2	7.0
Sandomierz	3.1	21.4	4.1	9.7	45.3
Dęblin	5.2	32.9	10.8	16.5	65.4
Warszawa	19.9	44.5	9.3	23.1	96.8
Toruń	40.7	61.8	24.4	15.9	142.8
Grudziądz	76.4	69.7	11.1	25.4	182.6
Malbork	2.1	1.8	1.3	2.6	7.8
Kiezmark	78.8	93.3	17.3	33.4	222.8
Vistula tributaries					
Nida	0.5	1.7	0.7	0.5	3.4
Dunajec	4.3	8.4	1.2	2.0	15.9
San	6.8	14.7	1.5	3.7	26.7
Wieprz	1.3	2.2	1.0	2.0	6.5
Pilica	0.9	2.7	1.0	3.9	8.5
Narew (Pułtusk)	5.9	16.1	1.2	8.9	32.1
Bug	10.8	36.5	2.2	1.2	50.7
Bug with Narew	15.8	53.8	3.4	8.2	81.2
Drwęca	0.3	2.6	0.7	2.6	5.9
Brda	0.3	0.8	0.4	1.6	3.1
Bzura	2.0	2.9	0.3	1.0	6.2

These data indicated, that the Vistula drainage is the important source of ^{210}Po in the Baltic Sea and boosts this region annually with 73.71 GBq of ^{210}Po. Amongst Vistula tributaries, the highest ^{210}Po runoff was characteristic for Narew (from 0.64 $GBq\cdot quarter^{-1}$ in summer to 7.47 $GBq\cdot quarter^{-1}$ in autumn; annual average 1.4 $GBq\cdot quarter^{-1}$). The seasonal inflow of polonium with the Vistula River was the highest in spring (Tab. 3), (Skwarzec and Jahnz 2007). In this period from Vistula drainage area to the Baltic Sea is transported polonium eluted from soil and contained in dry and wet atmospheric precipitation (Skwarzec et al. 2009b).
The results of the seasonal inflow of uranium with the Vistula River (Kiezmark) was

Table 5

Seasonal and annual runoff of $^{239+240}Pu$ from the Vistula River drainage

Sampling place	Winter	Spring	Summer	Autumn	All year $MBq\cdot year^{-1}$
	$MBq\cdot quarter^{-1}$				
the Vistula River					
Kraków	1.60	1.72	0.16	0.74	4.22
Sandomierz	9.58	21.60	1.89	1.47	34.54
Dęblin	10.0	54.7	6.30	6.27	77.27
Warszawa	6.59	11.80	5.00	20.70	44.09
Toruń	24.30	24.4	12.60	9.66	70.96
Grudziądz	20.30	32.3	2.91	5.88	61.39
Malbork	0.34	0.89	0.47	1.20	2.90
Kiezmark	15.70	50.80	3.38	16.10	86.00
Vistula tributaries					
Nida	0.43	1.33	0.19	0.35	2.30
Dunajec	1.86	9.44	0.59	1.68	13.57
San	15.31	23.5	1.56	2.33	42.70
Wieprz	3.02	2.64	0.17	1.42	7.25
Pilica	5.50	3.23	1.09	2.86	12.68
Narew (Pułtusk)	3.01	8.33	0.96	13.00	25.30
Bug	3.16	8.24	1.64	6.30	19.34
Bug with Narew	7.69	10.90	1.53	5.14	25.26
Drwęca	0.87	1.53	0.31	6.81	9.52
Brda	1.48	2.61	0.75	1.64	6.48
Bzura	0.73	1.16	0.29	0.81	2.99

higher in spring and winter, but lower in summer (Tab. 4). In spring season the highest value of uranium runoff in the Vistula River was observed in Kiezmark (93.3 GBq·quarter^{-1}), but amongst Vistula tributaries in Bug with Narew (53.8 GBq·quarter^{-1}). In summer the lowest values of ^{238}U flow in the Vistula River were noticed in Kraków (0.5 GBq·quarter^{-1}), but among Vistula tributaries in Drwęca and Nida (0.7 GBq·quarter^{-1}). In all year the highest runoff of ^{238}U in the Vistula River was observed in Kiezmark waters (222.8 GBq·year^{-1}), the lowest in Kraków waters (7.0 GBq·year^{-1}), (Tab. 4). Amongst the Vistula tributaries the maximum flow of uranium brought the Bug with Narew waters (81.2 GBq·year^{-1}), the minimum Brda waters (3.1 GBq·year^{-1}). Annually, the southern Baltic Sea is enriched by about 507 GBq uranium isotopes (Skwarzec et al. 2009b). Comparing with other radionuclides, annual ^{234}U and ^{238}U inflow to southern Baltic from Vistula catchments is higher than inflow of ^{210}Po. For comparison Mahanadi waters to the Bengal Bay in India is transporting 36 tone·year^{-1} of uranium (Ray et al. 1995). Saline main waters are source of natural isotopes and also chlorides and sulphates (Boryło et al. 2009). Annually with mine waters from Silesian Coal factory is transported 75 GB ^{226}Ra and 145 GBq ^{228}Ra to river waters. Mine waters type B transport every day 180 MBq of ^{226}Ra and 375 MBq of ^{228}Ra (Jasińska et al. 1996-1997, Chałupnik et al. 2001, Pociask-Karteczka et al. 2001, Chałupnik and Molenda 2002).

In case of plutonium during winter season the biggest amounts of $^{239+240}$Pu in the Vistula River flow through Toruń (24.30 MBq·quarter^{-1}), the lowest through small channel, a part of Vistula's delta – Nogat (Malbork) – (0.34 MBq·quarter^{-1}), (Tab. 5). Amongst Vistula tributaries the biggest plutonium flow was calculated for the long and wide mountain San River (15.31 MBq·quarter^{-1}), when the lowest in short Nida (0.43 MBq·quarter^{-1}). In spring season the biggest flow moved from Toruń to lower lowland part near Dęblin (54.70 MBq·quarter^{-1}) and Kiezmark – Vistula near delta – (50.80 MBq·quarter^{-1}), the lowest was calculated, similarly to winter, in short Nogat (Malbork), (0.89 MBq·quarter^{-1}). Amongst Vistula tributaries the biggest plutonium runoff in spring season was also observed in San (23.50 MBq·quarter^{-1}), when the lowest in lowland Bzura (1.16 MBq·quarter^{-1}). Summer season was characterized by the lowest plutonium flow during whole analyzed year. The biggest runoff was calculated in middle point of Vistula's stream – Toruń (12.60 MBq·quarter^{-1}) and was about two times lower than the biggest flow during winter and spring seasons, but the lowest flow was observed in the first point on Vistula – Kraków (0.16 MBq·quarter^{-1}). Amongst Vistula tributaries the biggest $^{239+240}$Pu runoff in summer season was also observed in big and wide lowland river Bug (1.64 MBq·quarter^{-1}) and as previously in San (1.56 MBq·quarter^{-1}), when the lowest in much shorter lowland Wieprz (0.17 MBq·quarter^{-1}). During autumn season plutonium flow was bigger that flows calculated for summer season. The biggest runoff was calculated in middle point of Vistula's stream – Warszawa (20.7 MBq·quarter^{-1}), but the lowest was observed in Kraków (0.74 MBq·quarter^{-1}). Amongst Vistula tributaries the biggest $^{239+240}$Pu flow in summer was observed in wide lowland river Narew (13.00 MBq·quarter^{-1}) when the lowest in Nida (0.35 MBq·quarter^{-1}). Annual calculations show that the biggest plutonium runoff was found near delta in Kiezmark (86.00 MBq·year^{-1}), the lowest amount flows through small channel Nogat (Malbork), (2.90 MBq·year^{-1}). Amongst

Table 6
Seasonal and annual surface runoff of ^{210}Po from the Vistula River tributaries drainages

Place	Winter	Spring	Summer	Autumn	All year [kBq·year^{-1}·km^{-2}]
	[kBq·quarter^{-1}·km^{-2}]				
Vistula tributaries					
Nida	20	230	100	30	380
Dunajec	210	1 370	60	80	1 720
San	60	420	30	130	640
Wieprz	190	300	10	80	580
Pilica	90	200	70	270	630
Narew (Pułtusk)	20	20	10	100	150
Bug	30	200	30	90	350
Bug with Narew	70	140	10	80	300
Drwęca	90	70	20	190	370
Brda	120	80	60	130	390
Bzura	110	160	30	110	410

Table 7
Seasonal and annual surface runoff of ^{238}U from the Vistula River tributaries drainages

Sampling location	Winter	Spring	Summer	Autumn	All year [kBq·year^{-1}·km^{-2}]
	[kBq·quarter^{-1}·km^{-2}]				
Nida	140	430	180	130	880
Dunajec	620	1 240	170	300	2 330
San	410	870	90	220	1 590
Wieprz	130	210	90	200	630
Pilica	100	290	100	420	910
Narew (Pułtusk)	80	210	20	120	430
Bug	70	580	50	60	760
Bug with Narew	140	470	30	70	710
Drwęca	60	480	120	480	1 140
Brda	60	170	90	350	670
Bzura	260	380	40	120	800

Table 8

Seasonal and annual surface runoff of $^{239+240}Pu$ from the Vistula River tributaries drainages

Sampling place	Winter	Spring	Summer	Autumn	All year Bq·km^{-2}·year^{-1}
	Bq·km^{-2}·quarter^{-1}				
Vistula tributaries					
Nida	110	345	49	91	595
Dunajec	273	1 387	87	247	1 994
San	908	1 395	93	138	2 534
Wieprz	290	253	16	136	695
Pilica	593	348	118	308	1 367
Narew (Pułtusk)	40	111	13	173	337
Bug	89	209	42	160	500
Bug with Narew	67	95	13	45	220
Drwęca	162	286	57	1 275	1 780
Brda	320	564	290	354	1 528
Bzura	94	148	38	104	384

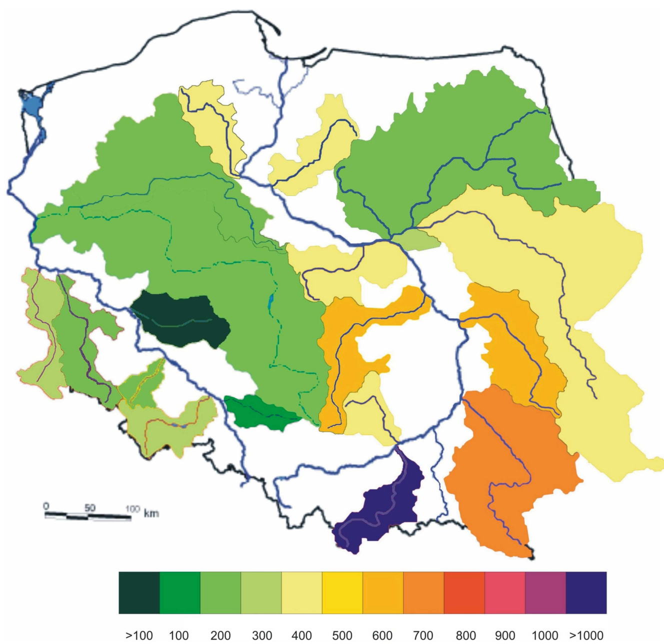

Fig. 2. Annual surface runoff of ^{210}Po from the Vistula and Odra tributaries drainages [kBq·km^{-2}·year^{-1}]

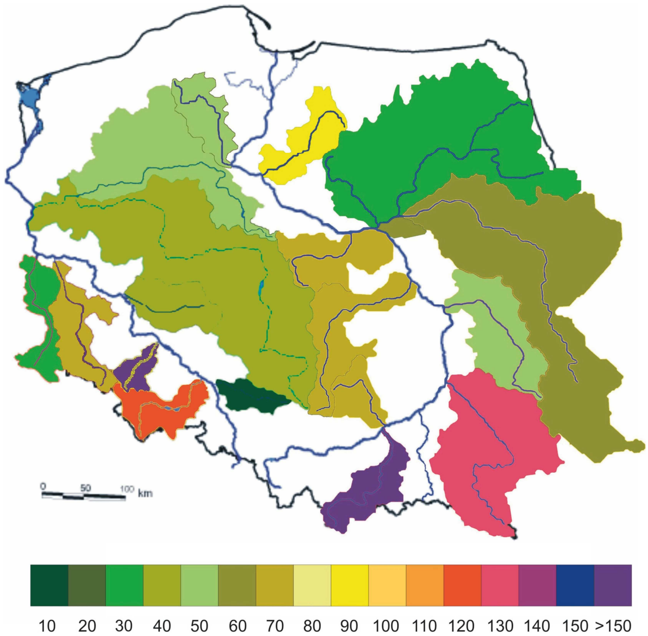

Fig. 3. Annual surface runoff of uranium from the Vistula and Odra tributaries drainages [g·km^{-2}·year^{-1}]

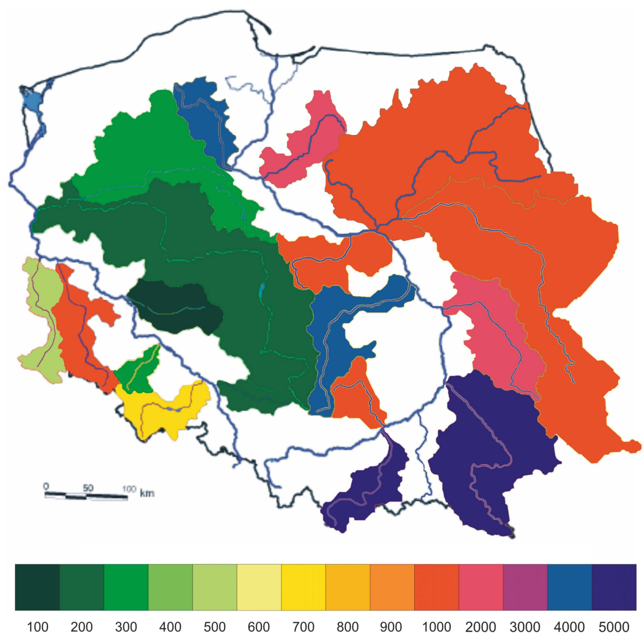

Fig. 4. Annual surface runoff of $^{239+240}$Pu from theVistula and Odra tributaries drainages [Bq·km^{-2}·year^{-1}]

Vistula tributaries the biggest annual plutonium flow was observed in wide-ranging mountain river San (42.70 MBq·year^{-1}) when the lowest in short Nida (2.30 MBq·quarter^{-1}). The total annual runoff of $^{239+240}$Pu from the Vistula River to Baltic Sea was calculated as 88.90 MBq.
On the basis of the Vistula tributaries area, there were calculated the seasonal and annual surface runoff of polonium, uranium and plutonium per unit of the area – obtained data are presented in Tables 6, 7 and 8, as well as in Figures 2, 3 and 4. The annual surface inflow of ^{210}Po from Vistula drainage was the highest in Dunajec (1 720 kBq·km^{-2}·year^{-1}) and San (640 kBq·km^{-2}·year^{-1}). The principal source of polonium in these rivers is rocks erosion and precipitation. The low-lying rivers contain polonium generally eluted from soil. (Skwarzec 2002). The lowest annual surface polonium was observed in Narew (150 kBq·km^{-2}·year^{-1}). The highest value o ^{238}U surface flow was noticed for Dunajec catchment (2 330 kBq·year^{-1}·km^{-2}), the lowest was transported to the Vistula River from Narew catchment (430 kBq·year^{-1}·km^{-2}), (Tab. 7, Fig. 3). In case of $^{239+240}$Pu the biggest values of total annual runoff were observed for mountain tributaries, such as San (2 534 Bq·km^{-2}·year^{-1}) and Dunajec (1 994 Bq·km^{-2}·year^{-1}), but the lowest annual runoff was found in Bug with Narew (220 Bq·km^{-2}·year^{-1}) and Bzura (384 Bq·km^{-2}·year^{-1}), (Tab. 8, Fig. 4).

Odra River catchment area

The concentration values of ^{210}Po, ^{238}U and $^{239+240}$Pu in water samples from the Odra River and their tributaries are presented in Table 9. The obtained data indicated the difference of analyzed radionuclides depending on seasons and place of taken samples. Generally, the polonium and uranium concentrations are higher in spring in comparison to winter. The highest value of ^{210}Po concentration in the Odra River was measured in water sample from Chałupki (3.64±0.23 Bq·m^{-3}) and Słubice (3.63±0.03 Bq·m^{-3}), but the lowest from Głogów (1.04±0.06 Bq·m^{-3}). Amongst the Odra tributaries maximum value of ^{210}Po concentration was observed in Bystrzyca (7.83±0.02 Bq·m^{-3}), but lowest in Barycz (0.60±0.09 Bq·m^{-3}), (Skwarzec and Tuszkowska 2008). In case of uranium the highest value of ^{228}U concentration in the Odra River was measured in water sample from Głogów (14.65±0.61 Bq·m^{-3}), but lowest from Chałupki (1.71±0.07 Bq·m^{-3}). Amongst the Odra tributaries maximum value of ^{238}U concentration was observed in Bystrzyca (134.9±2.85 Bq·m^{-3}), but lowest in Nysa Łużycka (1.39±0.06 Bq·m^{-3}). In the analyzed Odra River water samples there was observed radioactive disequilibrium between concentration of ^{234}U and ^{238}U and the value of ^{234}U/^{238}U activity ratio ranged between 1.03 (in Noteć) and 1.84 (in Odra), (Tuszkowska 2009). In case of plutonium $^{239+240}$Pu its highest concentration in the Odra River was measured in Chałupki (7.42±1.09 mBq·m^{-3}), but lowest in Widuchowa (1.24±0.34 mBq·m^{-3}). Among the Odra River tributaries, Bóbr was characterized by maximum value of $^{239+240}$Pu concentration (11.62±2.32 mBq·m^{-3}), but in Barycz plutonium concentration was the lowest (0.45±0.16 mBq·m^{-3}).
On the basis of average seasonal polonium, uranium and plutonium concentration in the Odra River and its tributaries, there were calculated average annual values of these radioelements concentration in analyzed rivers samples – the results are pre-

Table 9

Average concentration of ^{210}Po, ^{238}U and $^{239+240}Pu$ in the Odra River waters and its tributaries

Sampling location	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn
	^{210}Po±SD [Bq·m^{-3}]				^{238}U ± SD [Bq·m^{-3}]				$^{239+240}Pu$ ± SD [mBq·m^{-3}]			
the Odra River												
Chalupki	3.64 ± 0.23	1.62 ± 0.04	1.34 ± 0.05	1.38 ± 0.07	1.71 ± 0.07	2.54 ± 0.06	5.93 ± 0.21	5.45 ± 0.14	7.42 ± 1.09	2.35 ± 0.61	3.85 ± 0.96	4.54 ± 0.89
Glogów	1.80 ± 0.03	1.04 ± 0.06	1.42 ± 0.04	1.56 ± 0.09	7.59 ± 0.09	7.51 ± 0.21	14.65±0.61	14.69 ± 0.37	3.00 ± 0.49	3.17 ± 0.60	1.76 ± 0.35	3.45 ± 1.15
Slubice	3.63 ±0.03	3.32 ± 0.04	1.13 ± 0.06	1.50 ± 0.07	8.47 ± 0.20	10.27 ± 0.40	11.14±0.45	10.31 ± 0.19	2.67 ± 0.43	4.49 ± 0.86	3.60 ± 1.20	2.88 ± 0.96
Gozdowice	2.32 ± 0.04	2.48 ± 0.04	1.10 ± 0.05	1.64 ± 0.08	7.60 ± 0.29	8.40 ± 0.25	7.45 ± 0.30	6.73 ± 0.09	3.51 ± 0.85	3.25 ± 0.51	3.64 ± 0.74	3.76 ± 0.65
Widuchowa	1.13 ± 0.05	2.44 ± 0.04	1.79 ± 0.04	1.25 ± 0.08	5.74 ± 0.14	14.13 ± 0.47	6.98 ± 0.29	6.95 ± 0.19	1.24 ± 0.34	2.05 ± 0.47	2.00 ± 0.56	2.67 ± 0.49
Odra tributaries												
Mala Panew	1.71 ± 0.05	1.92 ± 0.04	1.57 ± 0.05	2.77 ± 0.07	1.92 ± 0.06	1.06 ± 0.02	2.19 ± 0.10	2.59 ± 0.04	0.85 ± 0.27	2.76 ± 0.36	3.04 ± 0.81	2.06 ± 0.37
Nysa Klodzka	3.19 ± 0.03	4.04 ± 0.03	2.00 ± 0.05	2.59 ± 0.12	7.32 ± 0.33	9.85 ± 0.07	12.68±0.13	13.08 ± 0.44	5.96 ± 0.87	4.32 ± 0.66	3.80 ± 0.62	3.20 ± 0.48
Bystrzyca	7.83 ± 0.02	2.73 ± 0.04	1.93 ± 0.05	1.09 ± 0.14	73.13 ± 1.89	17.82 ± 0.23	134.9±2.85	47.14 ± 0.69	7.10 ± 0.96	1.82 ±0.41	1.16 ± 0.31	1.05 ± 0.19
Barycz	1.29 ± 0.05	1.10 ± 0.05	0.60 ± 0.09	1.09 ± 0.03	13.00 ± 0.44	6.39 ± 0.18	3.69 ± 0.11	4.31 ± 0.10	1.97 ± 0.55	0.45 ±0.16	1.86 ± 0.59	2.18 ± 0.55
Bóbr	2.54 ± 0.04	2.03 ± 0.02	1.05 ± 0.05	1.89 ± 0.12	5,27 ± 0.13	8.83 ± 0.29	3.41 ± 0.12	3.42 ± 0.10	8.62 ± 1.88	7.24 ± 1.09	1.59 ± 0.28	11.62 ± 2.32
Nysa Luzycka	4.65 ± 0.03	2.81 ± 0.03	1.89 ± 0.04	5.21 ± 0.19	3.38 ± 0.04	1.39 ± 0.06	4.89 ± 0.07	3.72 ± 0.12	4.94 ± 0.65	4.20 ± 0.63	2.07 ± 0.47	4.76 ± 0.43
Warta	1.68 ±0.04	2.99 ± 0.06	1.60 ± 0.05	1.13 ± 0.06	5.96 ± 0.68	4.89 ± 0.18	6.18 ± 0.19	7.05 ± 0.12	2.37 ± 0.63	3.08 ± 1.26	4.10 ± 1.30	1.35 ± 0.25
Notec	1.00 ± 0.06	2.10 ± 0.02	0.71 ±0.07	2.09 ± 0.10	6.93 ± 0.24	3.18 ± 0.12	6.35 ± 0.21	7.76 ± 0.30	2.10 ± 0.70	4.39 ± 0.73	3.74 ± 0.80	3.67 ± 0.63

Table 10

Annual concentration of ^{210}Po, ^{238}U and $^{239+240}Pu$ in the Odra River waters and its tributaries

Sampling location	^{210}Po	^{238}U	$^{239+240}Pu$
	$Bq \cdot m^{-3}$	$Bq \cdot m^{-3}$	$mBq \cdot m^{-3}$
the Odra River			
Chałupki	2.00 ± 0.05	3.91 ± 0.12	4.59 ± 0,89
Głogów	1.46 ± 0.06	11.11 ± 0.32	2.85 ± 0.65
Słubice	2.39 ± 0.05	10.05 ± 0.31	3.41 ± 0.86
Gozdowice	1.89 ± 0.05	7.55 ± 0.23	3.54 ± 0.69
Widuchowa	1.65 ± 0.05	8.45 ± 0.27	1.99 ± 0.47
Odra tributaries			
Mała Panew	1.99 ± 0.05	1.94 ± 0.06	2.18 ± 0.45
Nysa Kłodzka	2.96 ± 0.06	10.73 ± 0.24	4.32 ± 0.66
Bystrzyca	3.39 ± 0.06	68.24 ± 1.42	2.78 ± 0.47
Barycz	1.02 ± 0.06	6.85 ± 0.21	1.62 ± 0.46
Bóbr	1.88 ± 0.06	5.23 ± 0.16	7.27 ± 1.39
Nysa Łużycka	3.64 ± 0.07	3.35 ± 0.07	3.99 ± 0.55
Warta	1.85 ± 0.05	6.02 ± 0.29	2.73 ± 0.86
Noteć	1.48 ± 0.06	6.06 ± 0.22	3.48 ± 0.72

sented in Table 10. Along the Odra River the highest value of studied isotopes appear in Słubice for polonium (2.39±0.05 $Bq \cdot m^{-3}$), Głogów for uranium (11.11±0.32 $Bq \cdot m^{-3}$) and Chałupki for plutonium (4.59±0.89 $mBq \cdot m^{-3}$). The lowest value of analyzed radionuclides were found in river water samples taken in Głogów (1.46±0.06 $Bq \cdot m^{-3}$ for ^{210}Po), Chałupki (3.91±0.12 $Bq \cdot m^{-3}$ for ^{238}U) and Widuchowa (1.99±0.47 for $^{239+240}Pu$). Amongst Odra tributaries the highest ^{210}Po, ^{238}U and $^{239+240}Pu$ concentration was found in Nysa Łużycka (3.64±0.07 $Bq \cdot m^{-3}$), Bystrzyca (68.24±1.24 $Bq \cdot m^{-3}$) and Bóbr (7.27±1.39 $mBq \cdot m^{-3}$) respectively, the lowest in Barycz (1.02±0.06 $Bq \cdot m^{-3}$), Mała Panew (1.94±0.06 $Bq \cdot m^{-3}$) and Barycz (1.62±0.46 $mBq \cdot m^{-3}$) respectively.

On the basis of average seasonal and annual flow of the Odra River and its tributaries there were calculated the seasonal and annual runoff of polonium, uranium and plutonium from Odra drainage – the results are presented in Tables 11, 12 and 13. The values of ^{210}Po seasonal flow, for the Odra River were the lowest in spring and lies in the range from 0.20 $GBq \cdot quarter^{-1}$ in Chałupki to 4.84 $GBq \cdot quarter^{-1}$ in Gozdowice. Amongst Odra tributaries the highest seasonal inflow was from Warta

Table 11

Seasonal and annual runoff of ^{210}Po from the Odra River drainage

Sampling location	Winter	Spring	Summer	Autumn	All year [GBq·year^{-1}]
	[GBq·quarter^{-1}]				
the Odra River					
Chałupki	0.21	0.54	0.24	0.20	1.19
Głogów	1.01	1.15	0.67	0.92	3.75
Słubice	1.41	3.36	3.08	1.06	8.91
Gozdowice	3.23	4.52	4.84	2.17	14.76
Widuchowa	2.49	2.23	4.82	3.57	13.11
Odra tributaries					
Mała Panew	0.05	0.03	0.04	0.03	0.15
Nysa Kłodzka	0.23	0.28	0.36	0.18	1.05
Bystrzyca	0.02	0.13	0.04	0.03	0.22
Barycz	0.012	0.014	0.012	0.007	0.045
Bóbr	0.22	0.29	0.23	0.12	0.86
Nysa Łużycka	0.38	0.34	0.20	0.14	1.06
Warta	0.96	1.41	2.51	1.36	6.24
Noteć	0.79	0.37	0.78	0.27	2.21

(2.51 GBq·quarter^{-1} in spring), but the lowest from Barycz (0.012 GBq·quarter^{-1} in autumn and spring). Annual flow of ^{210}Po from the Odra River was the highest in Gozdowice (14.76 GBq·year^{-1}), but the lowest in Chałupki (1.19 GBq·year^{-1}). The Odra drainage is the important source of ^{210}Po in the Southern Baltic Sea and boosts this region annually with 14.76 GBq ^{210}Po. Amongst Odra tributaries, the highest annually ^{210}Po runoff was characteristic for Warta (6.24 GBq·year^{-1}), but the lowest for Barycz (0.045 GBq·year^{-1}), (Skwarzec and Tuszkowska 2008). The data contained in Table 11 showed that seasonal inflow of polonium with the Vistula River was the highest in spring. In this period from drainage area to the Baltic Sea is transported polonium eluted from soil and contained in dry and wet atmospheric precipitation (Skwarzec et al. 2009b).

In case of uranium, the highest quantity of ^{238}U was transported in the Odra River stream especially in the river mouth in Widuchowa – from 13.04 GBq·quarter^{-1} in summer to 47.33 GBq·quarter^{-1} ^{238}U in spring (Tab. 12). The lowest flow of uranium was observed in the main stream in Chałupki (from 0.41 GBq·quarter^{-1} in spring to 3.23 GBq·quarter^{-1} in winter). In autumn the highest amount of uranium was ob-

Table 12

Seasonal and annual runoff of ^{238}U from the Odra River drainage

Sampling location	Winter	Spring	Summer	Autumn	All year [GBq·year^{-1}]
	[GBq·quarter^{-1}]				
the Odra River					
Chałupki	3.23	0.41	0.54	0.82	5.00
Głogów	23.45	11.75	7.16	9.52	51.88
Słubice	38.82	18.57	11.07	9.67	78.13
Gozdowice	45.71	27.01	14.09	13.27	100.08
Widuchowa	32.76	47.33	13.04	13.87	107.00
Odra tributaries					
Mała Panew	0.03	0.14	0.05	0.05	0.27
Nysa Kłodzka	1.68	2.71	1.13	1.16	6.68
Bystrzyca	3.08	1.08	4.33	0.78	9.27
Barycz	1.22	0.53	0.25	0.05	2.05
Bóbr	2.53	1.92	0.42	0.39	5.26
Nysa Łużycka	0.68	0.15	0.37	0.27	1.47
Warta	10.45	5.92	4.63	6.00	27.00
Noteć	4.75	1.39	1.99	2.93	11.06

served in Widuchowa (13.87 GBq·quarter^{-1}), the lowest in Chałupki (0.82 GBq·quarter^{-1}). Amongst Odra tributaries the highest runoff of uranium was transported by Warta's water (6.00 GBq·quarter^{-1}), the lowest in Barycz (0.05 GBq·quarter^{-1}). In winter the highest flow of uranium was carried by the Odra River water in Słubice (38.82 GBq·quarter^{-1}). Amongst Odra tributaries Warta transported the biggest amount of uranium to Odra's main stream (10.45 GBq·quarter^{-1}), the smallest – Mała Panew (0.03 GBq·quarter^{-1}). In spring similarly as in winter, among Odra tributaries Warta carried the biggest quantity of uranium (5.92 GBq·quarter^{-1}), against the smallest quantities carried by Mała Panew (0.14 GBq·quarter^{-1}) and Nysa Łużycka (0.15 GBq·quarter^{-1}). Summer season is characterized by much lower flow of uranium both in the Odra main stream and tributaries. Runoff of uranium content ranges from 0.54 GBq·quarter^{-1} in Chałupki to 14.09 GBq·quarter^{-1} in Gozdowice. Amongst Odra tributaries the highest flow value was for Warta (4.63 GBq·quarter^{-1}), the smallest was in water from Mała Panew (0.05 GBq·quarter^{-1}), (Tab. 12). Annually the Odra River water in Widuchowa transported the highest amount of ^{238}U (107.00 GBq·year^{-1}), the lowest in Chałupki (5.00 GBq·year^{-1}). Amongst Odra

Table 13

Seasonal and annual runoff of $^{239+240}$Pu from the Odra River drainage

Sampling location	Winter	Spring	Summer	Autumn	All year [MBq·year^{-1}]
	[MBq·quarter^{-1}]				
the Odra River					
Chałupki	14.00	0.38	0.35	0.69	15.42
Głogów	9.27	4.96	0.86	2.24	17.33
Słubice	12.24	8.12	3.58	2.70	26.64
Gozdowice	21.11	10.45	6.89	7.41	45.86
Widuchowa	7.08	6.87	3.74	5.33	23.02
Odra tributaries					
Mała Panew	0.02	0.36	0.06	0.04	0.48
Nysa Kłodzka	1.34	1.19	0.34	0.28	3.15
Bystrzyca	0.30	0.11	0.04	0.02	0.47
Barycz	0.18	0.04	0.13	0.02	0.37
Bóbr	4.13	1.57	0.19	1.33	7.22
Nysa Łużycka	1.00	0.46	0.16	0.35	1.97
Warta	4.16	3.73	3.07	1.15	12.11
Noteć	1.44	1.92	1.17	1.39	5.92

tributaries Warta distinguished from the others with the highest amount of uranium (27.00 GBq·year^{-1}), but the lowest quantity of ^{238}U was provided by Mała Panew (0.27 GBq·year^{-1}), (Tab. 12).

In case of plutonium flows of Odra waters and its main mouths the average quarter and annual $^{239+240}$Pu plutonium flows were calculated and presented in Table 13. During winter season the biggest amounts of $^{239+240}$Pu flow through Gozdowice (21.11 MBq·quarter^{-1}), but the lowest in Widuchowa (7.08 MBq·quarter^{-1}). Amongst Odra tributaries the biggest plutonium flow were calculated for Warta (4.16 MBq·quarter^{-1}) and Bóbr (4.13 MBq·quarter^{-1}). In spring season the biggest runoff was noticed in Gozdowice (10.45 MBq·quarter^{-1}), but the lowest in Chałupki (0.38 MBq·quarter^{-1}). Amongst Odra tributaries the biggest plutonium flow in spring season was also observed in Warta (3.73 MBq·quarter^{-1}), when the lowest in lowland Barycz (0.04 MBq·quarter^{-1}). Summer season is characterized by the lowest plutonium flow during all analyzed year. The biggest runoff was calculated in middle point of Odra's stream – Gozdowice (6.89 MBq·quarter^{-1}), but the lowest flow was observed in the first point Odra – Chałupki (0.35 MBq·quarter^{-1}). Amongst Odra tributaries the biggest $^{239+240}Pu$ flow in summer season was also observed in big and wide lowland river Warta (3.07 MBq·quarter^{-1}), when the lowest in much shorter lowland Mała Panew (0.06 MBq·quarter^{-1}). During autumn season plutonium flow in the Odra River was smaller than flows calculated for summer season. The biggest runoff was noticed in Gozdowice (7.41 MBq·quarter^{-1}), but the lowest flow was observed in Chałupki (0.69 MBq·quarter^{-1}). Amongst Odra tributaries the biggest $^{239+240}Pu$ flows in summer were observed in wide lowland rivers Noteć (1.39 MBq·quarter^{-1}) and Warta (1.15 MBq·quarter^{-1}), as well as in Bóbr (1.33 MBq·quarter^{-1}), while the lowest in Bystrzyca and Barycz (0.02 MBq·quarter^{-1}). Annual calculations show that the biggest plutonium flow in the Odra River was found in Gozdowice (45.86 MBq·year^{-1}), the lowest in Chałupki (15.42 MBq·year^{-1}). Amongst Odra tributaries the biggest annual plutonium flow was observed in Warta (12.11 MBq·year^{-1}), when the lowest in short Barycz (0.37 MBq·quarter^{-1}), (Tab. 13).

On the basis of the Odra tributaries area, there were calculated the seasonal and annual surface runoff of polonium, uranium and plutonium per unit of the area and obtained data are presented in Tables 14, 15 and 16, as well as on Figures 2, 3 and 4. The annual surface inflow of ^{210}Po from Odra drainage was the highest in mountain rivers Nysa Łużycka (245 kBq·km^{-2}·year^{-1}) and Nysa Kłodzka (229 kBq·km^{-2}·year^{-1}), but the lowest in Barycz (8 kBq·km^{-2}·year^{-1}). Seasonally, the lower values of polonium surface runoff were observed in summer in comparison with remaining quarters. The principal source of polonium in these rivers are rocks erosion and precipitation. The low-lying rivers contain polonium generally eluted from soil (Carvalho 1997, Skwarzec 2002). The highest annual values of ^{238}U surface flow were observed for Bystrzyca catchment (5 244 kBq·year^{-1}·km^{-2}), the lowest was transported to the Odra River from Mała Panew catchment (126 kBq·year^{-1}·km^{-2}), (Tab. 15, Fig. 3). Seasonally, the higher values of uranium surface runoff were ob-

Table 14

Seasonal and annual surface runoff of ^{210}Po from the Odra River tributaries drainages

Sampling location	Winter	Spring	Summer	Autumn	All year [kBq·year^{-1}·km^{-2}]
	[kBq·quarter^{-1}·km^{-2}]				
Mała Panew	25	15	17	14	71
Nysa Kłodzka	51	61	78	39	229
Bystrzyca	10	72	25	18	125
Barycz	2	3	2	1	8
Bóbr	37	49	39	20	145
Nysa Łużycka	88	78	47	32	245
Warta	18	26	46	25	115
Noteć	46	22	45	16	129

Table 15
Seasonal and annual surface runoff of ^{238}U from the Odra River tributaries drainages

Sampling location	Winter	Spring	Summer	Autumn	All year [kBq·year^{-1}·km^{-2}]
	[kBq·quarter^{-1}·km^{-2}]				
Mała Panew	24	16	64	22	126
Nysa Kłodzka	255	362	594	247	1 458
Bystrzyca	441	1 743	610	2 450	5 244
Barycz	9	220	96	45	370
Bóbr	67	430	326	71	894
Nysa Łużycka	63	158	36	85	342
Warta	110	192	109	85	496
Noteć	169	274	80	115	638

Table 16
Seasonal and annual surface runoff of $^{239*240}Pu$ from the Odra River tributaries drainages

Sampling location	Winter	Spring	Summer	Autumn	All year [Bq·year^{-1}·km^{-2}]
	[Bq·quarter^{-1}·km^{-2}]				
Mała Panew	19	7	168	30	224
Nysa Kłodzka	62	295	260	74	691
Bystrzyca	10	169	62	21	262
Barycz	4	33	7	23	67
Bóbr	226	704	267	33	1 230
Nysa Łużycka	81	230	108	36	455
Warta	21	76	68	56	221
Noteć	80	83	111	68	342

served in winter and spring in comparison with autumn and summer. Amongst Odra tributaries the extra highest values of surface runoff of ^{238}U were observed for Bystrzyca for all analyzed season, from 441 kBq·quarter^{-1}·km^{-2} in autumn to 2 450 kBq·quarter^{-1}·km^{-2} in summer. In case of $^{239+240}Pu$ the biggest value of total annual runoff was observed for Bóbr (1 230 Bq·km^{-2}·year^{-1}), but the lowest annual runoff was found in Barycz (67 Bq·km^{-2}·year^{-1}), (Tab. 16, Fig. 4). Seasonally, the highest values of surface runoff of plutonium were observed in winter, but lowest in autumn and summer.

Pomeranian rivers

Three important Pomeranian rivers (Słupia, Parsęta and Rega) were examined in spring season on determination of ^{210}Po, ^{238}U and $^{239+240}$Pu and obtained results are presented in Table 17. The concentration of ^{210}Po lies between 3.82 Bq·m^{-3} in Rega to 5.50 Bq·m^{-3} in Parsęta, but of ^{238}U from 5.10 Bq·m^{-3} in Słupia to 8.97 Bq·m^{-3} in Rega, as well as of $^{239+240}$Pu ranged between 3.38 in Rega and 11.58 in Parsęta mBq·m^{-3}. On the basis of these results there were calculated the values of annual flow for the Pomeranian rivers – the data are presented in Table 18. The annual run-offs of analyzed radionuclide were: 6.00 GBq·year^{-1} for ^{210}Po, 7.36 GBq·year^{-1} for ^{238}U and 9.28 MBq·year^{-1} for $^{239+240}$Pu.

Table 17

Average concentration of ^{210}Po, ^{238}U and $^{239+240}$Pu in the Pomeranian rivers in spring

Sampling location	^{210}Po [Bq·m^{-3}]	^{238}U [Bq·m^{-3}]	$^{239+240}$Pu [mBq·m^{-3}]
Rega	3.82±0.24	8.97±0.52	3.38±0.98
Parsęta	5.50±0.33	6.19±0.30	11.58±3.49
Słupia	4.84±0.21	5.10±0.29	3.90±1.08

Table 18

The surface runoff of ^{210}Po, ^{238}U and $^{239+240}$Pu from the Pomeranian rivers in spring

Sampling location	^{210}Po	^{238}U	$^{239+240}$Pu
	[GBq·quarter^{-1}]		[MBq·quarter^{-1}]
Rega	0.18	0.41	0.16
Parsęta	0.87	0.96	1.80
Słupia	0.45	0.47	0.36
Annual runoff	6.00	7.36	9.28

The principal sources of polonium, uranium and plutonium radionuclides in aquatic environment are the wet and dry atmospheric fallout of terrigenic and radioactive particles from erosion process, as well as weapons nuclear bomb, in particularly artificial radionuclides (Skwarzec 2002). The ^{210}Po in precipitation varies with geographical location and ranged between 10 and 500 Bq·m^{-2}·year^{-1} and depends on natural processes of rocks erosion and also coal incinerating (Skwarzec et al. 2009b). The higher values are characteristic for atmospheric deposition on the area between 30° and 60° latitude. The Vistula, Odra and Pomeranian rivers drainage area lies in these geographical parameters. Moreover, coal mining is a source of huge amount of waste containing large quantities of natural radionuclides, also polonium and uranium. The coal contains about 1% of all amount of trace elements and radionuclides existing in environment (for example ^{238}U, ^{226}Ra, ^{210}Po, ^{210}Pb). During combustion some radionuclides are emitted to the atmosphere as a gas and radioactive dust, the others stay concentrated in ash (Flues et al. 2002, Nakaoka et al. 1984). In flying coal ash the concentrations are 1 700 $Bq \cdot kg^{-1}$ for ^{210}Po and 200 $Bq \cdot kg^{-1}$ for ^{238}U (Baxter 1996). The clinker after combustion is often used for roads stabilization, as a complement of building materials and for soil fertilization. The inflow of uranium from the river catchment area is caused by human activity on the drainage area, mainly in agriculture (using phosphate fertilizers) and mining industry (discharge of mine waters to river). In phosphate rocks there are usually radionuclides of uranium (^{234}U and ^{238}U) and radium (^{226}Ra), (Bem 2005). Phosphate rocks used to produce phosphate fertilizers contain from 1 300 to 1 500 $Bq \cdot kg^{-1}$ uranium ^{238}U and its decay products (Bolivar et al. 1995). During production of phosphate fertilizers about 10% of the initial amount of ^{226}Ra, 20% of uranium and about 85% of ^{210}Po is found in the waste phosphogypsum (Carvalho 1995, Carvalho et al. 2007). Phosphogypsum have enhanced concentration of natural radionuclides and it constitutes important problem in the pollution of soil groundwater and river water also in Poland (Boryło et al. 2009). In the neighborhood of phosphate fertilizers plant in Gdańsk (Northern Poland) concentration in soil and the Martwa Wisła River samples is much bigger in comparison to not contaminated samples (Boryło et al. 2009). Also surrounding of phosphogypsum waste located in Wiślinka (near Gdańsk and mouth of the Vistula River to Baltic Sea) is strongly polluted by polonium and uranium (Skwarzec et al. 2009a).

The average ^{238}U concentration in the Vistula, Odra and Pomeranian rivers and theirs tributaries is lower in comparison to values observed in Jucar and Ortigas Rivers in Spain and Turkey rivers (Rodriguez-Alvarez and Sanchez 1995, Vargas et al. 1995, Kumru 1997). Uranium concentration in the Kalixälven River (Sweden) was between 0.180±0.001 $\mu g \cdot kg^{-1}$ in spring time flood and 0.160±0.001 $\mu g \cdot kg^{-1}$ in summer and winter 1991-1992 (Andersson et al. 1995). Content of ^{238}U in Vistula in February 1993 was four-fold higher (0.724±0.001 $\mu g \cdot kg^{-1}$). Generally uranium content in rivers and sea is estimated as $5 \cdot 10^{-6}$-$2 \cdot 10^{-8}$%. Uranium concentration in sea waters is 3.2 $mg \cdot m^{-3}$, in river waters 0.4 $mg \cdot m^{-3}$, and $1 \cdot 10^{-7}$% in oceans (Skwarzec 1995). The uranium concentration in oceanic waters is constant. In river waters this concentration depends on weathering rate of rocky material (Ku et al. 1977). The activity ratio of uranium isotopes $^{234}U/^{238}U$ in natural water is not constant and this reason may by used in chronology, origin and inland waters share in coastal mixing and circulation of sea water processes (Skwarzec 1995, Skwarzec et al. 2004). The Vistula, Odra and Pomeranian Rivers waters are characterized by the higher values of uranium concentration and higher values of $^{234}U/^{238}U$ activity ratio than waters of southern Baltic waters (1.17). It is caused also by human activity on the drainage area of these rivers. Rivers waters are consumed by the people living on this area. The average isotopes uranium ^{234}U and ^{238}U concentration in drinking water from Polish rivers (1992-1996) was between 10.2-15.0 and 7.3-8.3 $Bq \cdot m^{-3}$ respectively, but the value of activity ratio $^{234}U/^{238}U$ was 1.23±0.40 (Pietrzak-Flis et al. 1997). Drinking water can be a source of 75.1% ^{238}U and 76.9% ^{234}U found in the human organism. Yearly annual effective dose is 5.95±0.21 μSv, the highest part comes from ^{226}Ra (72.4%). Uranium (^{234}U and ^{238}U) and thorium (^{232}Th, ^{230}Th and ^{235}Th) are source of 14.1% and 13.5% alpha radionuclides in human body (Pietrzak-Flis et al. 2001, Skwarzec et al. 2001, 2003a).

Inflow of ^{210}Po, ^{238}U and $^{239+240}$Pu radionuclides from Vistula, Odra and Pomeranian rivers to the Baltic Sea

On the basis of annual flow of analyzed radionuclide from Vistula, Odra and Pomeranian rivers, there was calculated annual inflow of ^{210}Po, ^{238}U and $^{239+240}$Pu to the southern Baltic Sea – obtained results are presented in Table 19 (Jahnz 2007, Skwarzec

Table 19

Annual runoff of ^{210}Po, ^{234}U, ^{238}U, ^{238}Pu and $^{239+240}$Pu from Vistula, Odra and the Pomeranian rivers drainages to the Baltic Sea

Radionuclide	Vistula drainage	Odra drainage	Pomeranian rivers drainage	Total
^{210}Po [GBq·year^{-1}]	73.7	14.8	6.0	94.5
^{234}U [GBq·year^{-1}]	276.8	126.3	8.2	411.3
^{238}U [GBq·year^{-1}]	230.6	100.8	7.4	338.8
U total [ton·year^{-1}]	18.8	8.2	0.6	27.6
^{238}Pu [MBq·year^{-1}]	10.1	9.5	2.6	22.2
$^{239+240}$Pu [MBq·year^{-1}]	88.9	45.9	9.3	144.1

and Jahnz 2007, Skwarzec and Tuszkowska 2008, Tuszkowska 2009). Annually, the southern Baltic Sea is enriched by 73.7 GBq of ^{210}Po (71.6 GBq inflow to the Gdańsk Bay, and 2.1 GBq inflow to the Vistula Lagoon), about 507 GBq of ^{234}U and ^{238}U (490 GBq going to the Gdańsk Bay via the Leniwka River and 16.5 GBq to the Vistula Lagoon via the Nogat River), as well as 10.1 MBq of ^{238}Pu and 88.9 MBq of $^{239+240}$Pu flow with the Vistula River waters. From this amount 9.96 MBq ^{238}Pu (96.7%) and 86.0 MBq $^{239+240}$Pu (96.6%) go with the Leniwka River to the Gulf of Gdańsk, 0.39 MBq ^{238}Pu (3.3%) and 2.90 MBq $^{239+240}$Pu (3.4%) go with Nogat to the Vistula Lagoon. The inflow of analyzed radionuclides from the Odra River is smaller with comparison to the Vistula River. The annual flow of polonium, uranium and plutonium from the Odra River to the Szczecin Lagoon was calculated as 14.8 GBq of ^{210}Po, 126.3 GBq of ^{234}U, 100.8 GBq of ^{238}U, was well as 9.5 MBq of ^{238}Pu and 45.9 MBq of $^{239+240}$Pu. The inflow of the analyzed radionuclides for Pomeranian rivers is less important in comparison with Vistula or Odra rivers. From Rega, Pasłęka, Słupia rivers every year to the Southern Baltic is going 6 GBq of ^{210}Po, 8.2 GBq of ^{234}U and 7.4 GBq of ^{238}U as well as 2.5 MBq of ^{238}Pu and 9.3 MBq of $^{239+240}$Pu. Total annual runoff of polonium, uranium and plutonium from Vistula, Odra and Pomeranian rivers to the Baltic Sea was calculated as about 95 GBq of

^{210}Po, 750 GBq of $^{234+238}$U and 166 MBq of $^{238+239+240}$Pu. These data indicated, that Vistula, Odra and Pomeranian rivers are the important sources of polonium, uranium and plutonium radionuclides in the southern Baltic environment.

Polonium, uranium and plutonium radionuclides in the southern Baltic Sea

Polonium ^{210}Po

The analysis of seawater samples revealed that the mean value of ^{210}Po concentration in Baltic water was 0.59 mBq·dm^{-3}, 80% of which consisted of soluble forms. There were significant differences in the concentrations of dissolved polonium in the samples analyzed. The concentration of ^{210}Po in suspended matter was 74 Bq·kg^{-1} d.w. (Skwarzec and Bojanowski 1988, Skwarzec 1995, 1997b).
Amongst analyzed radionuclides ^{210}Po is mostly accumulated in land and Baltic organisms. Mean values of the bioconcentration factor (BCF) in the Baltic organisms, calculated on the basis of the polonium content, fell within the range 1.5·10^3-3.2·10^4 (Tab. 20). In 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 < Malaco-

Table 20
^{210}Po activity concentration and bioconcentration factor (BCF) in Baltic organisms in Poland

Organisms	^{210}Po concentration Bq.kg^{-1} d.w.	BCF·10^3
Baltic organisms		
Phytoplankton	41	4.2
Phytobenthos	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	27	11
– alimentary track	420	70
– muscles	4.5	1.5

straca < Bivalves (soft tissues), (Skwarzec and Bojanowski 1988, Skwarzec and Falkowski 1988, Skwarzec 1995, 1997b, Skwarzec et al. 2003c, 2006). The data on *Saduria entomon* Crustaceans and *Mya arenaria* Bivalves indicated ^{210}Po is not uniformly distributed in their bodies. The 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 fish, the organs directly connected with digestion (intestine, liver, spleen, pyloric caeca) contain much higher ^{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 values of ^{210}Po/^{210}Pb are observed in animals (Skwarzec 1988, 1995, 1997b, Skwarzec and Falkowski 1988). Moreover, about 30% of ^{210}Po consumed by statistical Polish habitant is uptaken from fish and the annual radioactive dose is 43 μSv (Skwarzec 2002).

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

The uranium concentrations lay within the range from 0.68 to 0.85 $\mu g \cdot dm^{-3}$ in surfacebaltic water samples. The mean value of ^{234}U/^{238}U activity ratio in Baltic seawater was found to be 1.17. The uranium concentration in seawater increased, but values of ^{234}U/^{238}U activity ratio decreased with salinity (Skwarzec 1995). Uranium, in contrary to polonium, is not highly accumulated by Baltic organisms. Marine al-

Table 21
The average values of ^{238}U concentrations, bioconcentration factor (BCF) and ^{234}U/^{238}U activity ratio in Baltic organisms

Organisms	^{238}U concentration $Bq \cdot kg^{-1}$ d.w.	BCF	^{234}U/^{238}U activity ratio
Baltic organisms			
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	

gae and benthic animals concentrated uranium isotopes to a small extent only. The values of BCF ranged from 0.4 to 82 (Tab. 21). The mean value of 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 is increasing in the sequence muscle < skeleton < viscera (Skwarzec 1995, 1997b). The average value of $^{234}U/^{238}U$ activity ratio in Baltic suspended matter is about 1.0, i.e. similar to that in sediments. This indicated that terrigenic material is the general source of uranium in the Baltic sediments. When the values of $^{234}U/^{238}U$

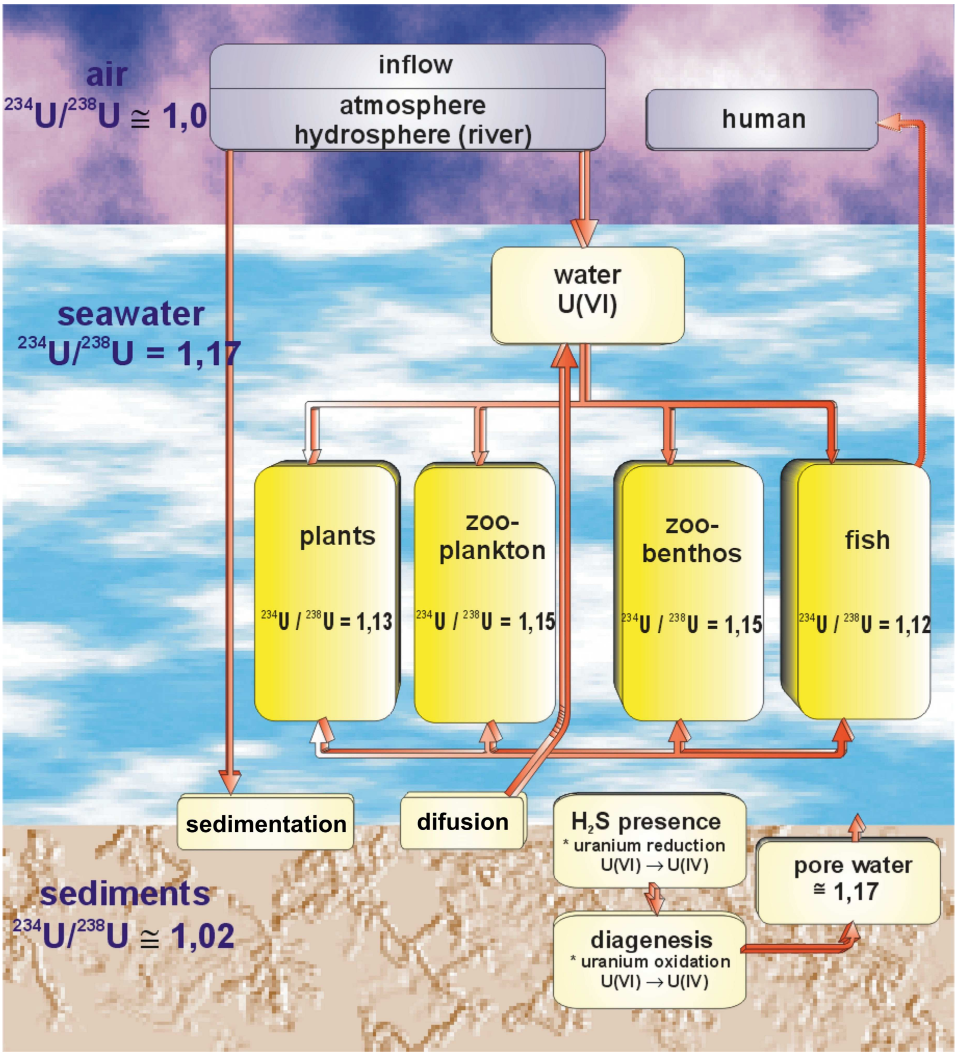

Fig. 5. $^{234}U/^{238}U$ activity ratio in the Baltic Sea ecosystem

activity ratio in Baltic organisms, in the 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 source of uranium in Baltic biota (Fig. 5). The concentration of uranium in sediments from the southern Baltic increases with depth core, probably indicating the diffusion from sediments to water through interstitial water, where uranium concentration is much higher than in bottom water (Skwarzec et al. 2004). The values of $^{234}U/^{238}U$ activity ratio in sediments from reduction area of the southern Baltic (the Gdańsk Deep and the Bornholm Deep) indicated that the reduction process of U(VI) to U(IV) and removal of autogenic uranium from seawater to sediments constitute a small part only in the Gdańsk Deep (Skwarzec et al. 2002, 2004). The most important processes in geochemical uranium migration in the Southern Baltic Sea ecosystem are terrigenous material and river (Vistula) suspended matter and its diffusion from sediments to bottom water (Skwarzec 2002).

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

The average level of $^{239+240}Pu$ in Baltic seawater was found to be 4.8 $mBq \cdot m^{-3}$ (Tab. 22), near 60% of which constituted filterable forms (≤ 0.45 μm). The concentration of plutonium in the suspended matter was 1.5 $Bq \cdot kg^{-1}$ d.w., and the distribution coeffi-

Table 22

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

Organisms	$^{239+240}Pu$ concentration $Bq \cdot kg^{-1}$ d.w.	$BCF \cdot 10^3$
Baltic seawater	4.8 $mBq \cdot m^{-3}$	
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

cient (DC) of plutonium was of the order of $4{\cdot}10^5$ (Skwarzec 1995, 1997b, Skwarzec et al. 2006). The plutonium concentration in southern Baltic changes horizontally from east to west, and is the highest in the Pomeranian Bay, where richer in plutonium water inflows. The concentration of $^{239+240}Pu$ in seawater from the Pomeranian Bay ranged 150 $mBq{\cdot}m^{-3}$ and the Gdańsk Bay it ranged only 5.2 $mBq{\cdot}m^{-3}$ (Skwarzec and Strumińska 2001, Strumińska and Skwarzec 2005).

Concerning plutonium it was noticed it is highly accumulated in Baltic organisms and BCF values range from 25 to $27{\cdot}10^3$. The Baltic plants and benthic animals concentrated plutonium isotopes to various extents (Tab. 22). The $^{239+240}Pu$ concentrations in the Baltic organisms ranged from 14 $mBq{\cdot}kg^{-1}$ d.w. (fish) to 957 $mBq{\cdot}kg^{-1}$ d.w. (Priapulida). In the seaweeds the highest plutonium concentrations were found in *Pylaiella litoralis* collected in the Puck Bay in 1987. In the zoobenthos, plutonium concentrations were higher in Priapulida and Polychaeta and lower in Entomostraca, Malacostraca and Bivalves (soft tissues), (Skwarzec and Bojanowski 1992, Skwarzec 1995). 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 were found in muscles. Participation of evaluated Chernobyl-derived plutonium in Baltic fish reached from 20% for *Neogobius melanostomus* to 70% for herring *Clepea harrengus* (Strumińska and Skwarzec 2004, 2005).

The analysis of $^{238}Pu/^{239+240}Pu$ activity ratios in Baltic flora and fauna showed increasing with time Chernobyl plutonium participation in the total content, noticed especially in fish. It indicates plutonium remobilization from sediments to bottom water via benthic organisms and the effect of plutonium late inflow from Baltic catchment's area. It is especially important in the Gdańsk Bay because the experiments show this basin is more contaminated with plutonium (Tab. 23). The sediments of the Gdańsk Bay contain 6.4% of $^{239+240}Pu$ deposited in sediments of whole Baltic Sea but the area of the Gdańsk Bay constitutes only about 1.2% (Skwarzec et al. 2003b).

The main source of $^{239+240}Pu$ inflow in the Gdańsk Bay and the Gdańsk Basin is the Vistula River, which enriches these regions with 78% of total plutonium. The total

Table 23

$^{239+240}Pu$ plutonium inventories in 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)		1	0,5
water and suspended matter (GBq)	200	2.3	1.2
organisms (MBq)	a few GBq	3.81	~0.1
area (km^2)	415 266	4 940	1.2
capacity (km^3)	21 721	291.2	1.3

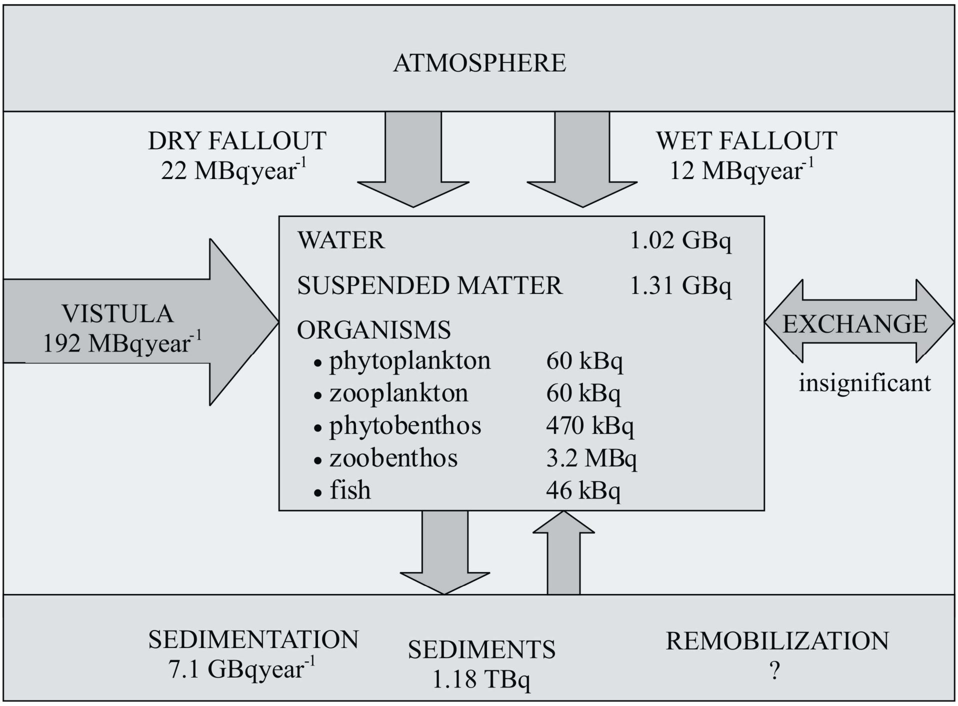

Fig. 6. The scheme of plutonium inventories in the Gdańsk Bay

$^{239+240}$Pu amount deposited in the Gdańsk Bay and the Gdańsk Basin is 1.18 TBq and 3.76 TBq respectively (Fig. 6 and 7), (Skwarzec et al. 2003b). Almost whole plutonium is deposited in Baltic sediments. Because the area of the Gdańsk Bay and the 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 the Gdańsk Bay (with suspended matter) there is about 2.3 GBq (0.2% of total amount) and 9.92 GBq (0.3% of total amount) in the Gdańsk Basin. In both cases 56% of $^{239+240}$Pu is associated with suspended matter. Organisms in the Gdańsk Bay contain 3.81 MBq and 7.45 MBq $^{239+240}$Pu in the Gdańsk Basin. From this value in the 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 the 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 (Fig. 6 and 7), (Skwarzec et al. 2003b).

Air filters analysis after Chernobyl, at 26 April 1986 accident indicated, that ^{241}Pu concentration increased suddenly over 3 500 times, then was decreasing slowly and in November 1986 got the level before the accident. The ^{241}Pu activity concentration in air increased from value near 1 $Bq{\cdot}kg^{-1}$ d.w. to 3 643 $Bq{\cdot}kg^{-1}$ d.w. and then slowly decreased. (Strumińska and Skwarzec 2005). Its concentration in seawater from the Gdańsk Bay was 0.23 $Bq{\cdot}m^{-3}$ and value of ^{241}Pu/$^{239+240}$Pu activity ratio was the same

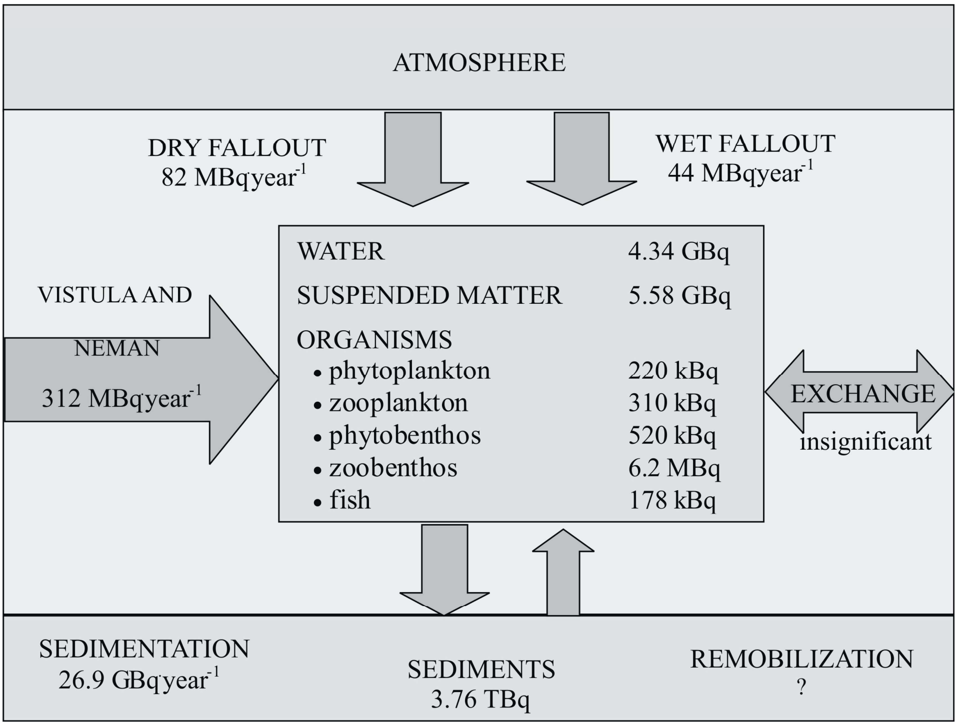

Fig. 7. The scheme of plutonium inventories in the Gdańsk Basin

Table 24

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

Sample	^{241}Pu concentration ($Bq \cdot m^{-3}$)	$^{241}Pu/^{239+240}Pu$ 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 25
^{241}Pu concentration in phyto- and zoobenthos from the Puck Bay collected in 1986 and 1987

Organism	^{241}Pu concentration (Bq·kg^{-1} d.w.)	^{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

as in the moment of accident and equaled 140. Plutonium is strongly accumulated in Baltic organisms (Tab. 24), (Strumińska and Skwarzec 2006). Amongst all analyzed organisms higher ^{241}Pu activity concentrations were found in brown algae *Pylaiella litoralis* (1.01 Bq·kg^{-1} d.w.), Priapulida *Halicryptus sinulosus* (9.20 Bq·kg^{-1} d.w.) and Polychaeta *Antinöella sarsi* (7.71 Bq·kg^{-1} d.w.). Analyzed ^{241}Pu/$^{239+240}$Pu activity ratio indicates that a few years after Chernobyl accident it is too early to estimate the impact of this accident on the Baltic Sea environment (Strumińska and Skwarzec 2006). In the case of Baltic flora, the ^{241}Pu activity concentration in analyzed seaweed from the Puck Bay ranged from 0.22 Bq·kg^{-1} d.w. for seed plants *Elodea canadiensis* and 0.24 Bq·kg^{-1} d.w. for *Zostera marina*, to 1.01 Bq·kg^{-1} d.w. for brown algae *Pylaiella litoralis* (Tab. 25). 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. The values of ^{241}Pu/$^{239+240}$Pu activity ratios were from 5.4 for green algae *Cladophora rupestris* to 35 for seed plant *Potamogeton pectinalis* (Tab. 25). It indicates that after Chernobyl accident in 1986 there was no significant amount of plutonium in phytobenthos from the Puck Bay (Strumińska and Skwarzec 2006). In case of Baltic zoobenthos organisms the ^{241}Pu activity concentrations vary from 0.13 Bq·kg^{-1} d.w. for Crustacean *Saduria entomon* and 0.20 Bq·kg^{-1} d.w. for *Balanus improvisus*, to 7.71 Bq·kg^{-1} d.w. for Polychaeta *Antinöella sarsi* and 9.20 Bq·kg^{-1} d.w. for Priapulida *Halicryptus spinulosus* (Tab. 25). The values of the ^{241}Pu/$^{239+240}$Pu activity ratios are versatile 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. These results indicate that plutonium in zoobenthos organisms is accumulated from sediments and interstitial water inversely to phytobentos organisms, where plutonium can be adsorbed from seawater (Strumińska and Skwarzec 2005, 2006).

CONCLUSIONS

On the basis of done study we can conclude, that the annual surface inflow of ^{210}Po from Vistula drainage was the highest in Dunajec (1 720 kBq·km^{-2}·year^{-1}) and San (640 kBq·km^{-2}·year^{-1}), but the lowest was observed in Narew (150 kBq·km^{-2}·year^{-1}). The highest value of ^{238}U surface flow was noticed for Dunajec catchment (2 330 kBq·year^{-1}·km^{-2}), the lowest was transported to the Vistula River from Narew catchment (430 kBq·year^{-1}·km^{-2}). In case of $^{239+240}$Pu the biggest values of total annual runoff were observed for mountain tributaries, such as San (2 533 Bq·km^{-2}·year^{-1}) and Dunajec (1 994 Bq·km^{-2}·year^{-1}), but the lowest annual runoff was found in Bug with Narew (220 Bq·km^{-2}·year^{-1}) and Bzura (384 Bq·km^{-2}·year^{-1}). The annual surface inflow of ^{210}Po from Odra drainage was the highest in mountain rivers Nysa Kłodzka (245 kBq·km^{-2}·year^{-1}) and Nysa Łużycka (229 kBq·km^{-2}·year^{-1}), but the lowest in Barycz (8 kBq·km^{-2}·year^{-1}). Seasonally, the lower values of polonium surface runoff were observed in summer in comparison with remaining quarters. The highest annual values of ^{238}U surface flow were observed for Bystrzyca catchment (5 244 kBq·year^{-1}·km^{-2}), the lowest was transported to the Odra River from Mała Panew catchment (126 kBq·year^{-1}·km^{-2}). Seasonally, the higher values of surface runoff of uranium were observed in winter and spring in comparison with autumn and summer. Amongst Odra tributaries extra highest values of ^{238}U surface runoff were observed for Bystrzyca for all analyzed seasons, from 441 kBq·quarter^{-1}·km^{-2} in autumn to 2 450 kBq·quarter^{-1}·km^{-2} in summer. In case of $^{239+240}$Pu the biggest value of total annual runoff was observed for Warta (1 230 Bq·km^{-2}·year^{-1}), but the lowest annual runoff was found in Barycz (67 Bq·km^{-2}·year^{-1}). Seasonally, the highest values of plutonium surface runoff were observed in winter, but lowest in autumn and summer. Results of obtained investigation indicated, that Vistula, Odra and Pomeranian rivers are the important sources of polonium, uranium and plutonium radionuclides in the southern Baltic environment. Annually, the southern Baltic Sea is enriched by 95 GBq of ^{210}Po, 750 GBq of $^{234+238}$U and 160 MBq of $^{238+239+240}$Pu. In the southern Baltic environment polonium and plutonium are strongly accumulated by organisms. The values of bioconcentration factor (BCF) range between $1.5 \cdot 10^3$-$3.2 \cdot 10^4$ for ^{210}Po and from 25 to $27 \cdot 10^3$ for $^{239+240}$Pu. Polonium and plutonium in

Baltic organisms are not uniformly distributed. Most of accumulated ^{210}Po and $^{239+240}$Pu in analyzed Baltic invertebrates and fish is located in soft tissues (hepatopancreas, alimentary track). Some organisms are good bioindicators for radioactive contamination of land and Baltic environment, especially the brown algae *Pylaiella litoralis* and Priapulida *Halicryptus spinulosus* are very good bioindicators for plutonium contamination of the Southern Baltic Sea ecosystem. The impact of Chernobyl plutonium on Baltic organisms is in 2003 significant and equals from 10% to 60%. 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 $^{239+240}$Pu activity inventory within the Baltic Sea sediments. Uranium in comparison to polonium and plutonium is not highly accumulated by Baltic organisms and BCF values for ^{238}U in biota ranged between 1 and 80. The analysis of ^{234}U/^{238}U activity ratio indicates the main sources of uranium in Baltic organisms are its dissolved forms in seawater. The most important processes in geochemical uranium migration in southern Baltic ecosystem are terrigenous material and rivers (Vistula, Odra and Pomeranian rivers) suspended matter and its diffusion from sediment to bottom water through interstitial water.

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RADIONUKLIDY POLONU, URANU I PLUTONU W ŚRODOWISKU WODNYM POLSKI I POŁUDNIOWEGO BAŁTYKU

Streszczenie

W pracy przestawiono wyniki badań nad występowaniem naturalnych (polon ^{210}Po, uran ^{234}U i ^{238}U) i sztucznych (pluton ^{238}Pu, $^{239+240}$Pu i ^{241}Pu) radionuklidów alfa promieniotwórczych w środowisku wodnym Polski i południowego Bałtyku, jak również rozpoznanie procesu ich nagromadzania w morskim łańcuchu troficznym. Wyniki badań wykazały, że Wisła i Odra oraz Rega, Słupia i Parsęta są ważnymi źródłami analizowanych radionuklidów w południowym Bałtyku. Całkowity roczny spływ polonu, uranu i plutonu wodami Wisły, Odry i rzek Przymorza został obliczony na 95 GBq dla ^{210}Po, 750GBq dla $^{234+238}$U oraz 166 MBq dla $^{238+239+240}$Pu.

Badania zawartości polonu ^{210}Po, uranu ^{234}U i ^{238}U, jak również plutonu ^{238}Pu, $^{239+240}$Pu oraz ^{241}Pu w organizmach bałtyckich wykazały silne nagromadzanie w nich radionuklidów, szczególnie polonu i plutonu. Wartości współczynnika nagromadzania (BCF) polonu i plutonu w organizmach południowego Bałtyku mieszczą się w szerokim przedziale wartości od 25 do 27000. Bałtyckie wodorosty i organizmy zwierzęce nagromadzają uran w niewielkim

stopniu. Z kolei w osadach bałtyckich stężenie uranu wzrasta z głębokością rdzeni, co związane jest z dyfuzją izotopów ^{234}U, ^{235}U i ^{238}U za pomocą wody porowej do wody naddennej. Analiza wartości stosunku aktywności $^{234}U/^{238}U$ w osadach wykazała, że możliwa redukcja U(VI) do U(IV) i usuwanie autogenicznego uranu z wody morskiej do osadów w redukcyjnych rejonach Bałtyku: Głębi Gdańskiej i Głębi Bornholmskiej zachodzi w niewielkim stopniu.