The inflow of ^{234}U and ^{238}U from the River Odra drainage basin to the Baltic Sea*

OCEANOLOGIA, 52 (4), 2010.
pp. 705–725.

KEYWORDS
Uranium
^{234}U
^{238}U
River Odra
Poland

Bogdan Skwarzec*
Agnieszka Tuszkowska
Alicja Boryło

Department of Analytical Chemistry,
Faculty of Chemistry,
University of Gdańsk,
Sobieskiego 18/19, PL–80–952 Gdańsk, Poland;

e-mail: bosk@chem.univ.gda.pl

*corresponding author

Received 28 September 2009, revised 11 August 2010, accepted 2 September 2010.

Abstract

In this study the activity of uranium isotopes ^{234}U and ^{238}U in Odra river water samples, collected from October 2003 to July 2004, was measured using alpha spectrometry. The uranium concentrations were different in each of the seasons analysed; the lowest values were recorded in summer. In all seasons, uranium concentrations were the highest in Bystrzyca river waters (from 27.81 ± 0.29 Bq m^{-3} of ^{234}U and 17.82 ± 0.23 Bq m^{-3} of ^{238}U in spring to 194.76 ± 3.43 Bq m^{-3} of ^{234}U and 134.88 ± 2.85 Bq m^{-3} of ^{238}U in summer). The lowest concentrations were noted in the Mała Panew (from 1.33 ± 0.02 Bq m^{-3} of ^{234}U and 1.06 ± 0.02 Bq m^{-3} of ^{238}U in spring to 3.52 ± 0.05 Bq m^{-3} of ^{234}U and 2.59 ± 0.04 Bq m^{-3} of ^{238}U in autumn). The uranium radionuclides ^{234}U and ^{238}U in the water samples were not in radioactive equilibrium. The $^{234}U/^{238}U$ activity ratios were the highest in

* The authors would like to thank the Minister of Sciences and Higher Education for the financial support of this work under grant number: DS/8460-4-0176-0 and BW-8000-5-0249-9.

Odra water samples collected at Głogów (1.84 in autumn), and the lowest in water from the Noteć (1.03 in winter and spring). The $^{234}U/^{238}U$ activity ratio decreases along the main stream of the Odra, owing to changes in the salinity of the river's waters. Annually, 8.19 tons of uranium (126.29 GBq of ^{234}U and 100.80 GBq of ^{238}U) flow into the Szczecin Lagoon with Odra river waters.

1. Introduction

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). Naturally occurring uranium contains three alpha emitting radionuclides: ^{238}U (99.2745%), ^{235}U (0.7200%) and ^{234}U (0.0055%) (Bou-Rabee et al. 1995, Bagatti et al. 2003). 1 Bq of ^{238}U corresponds to 81.6 μg of natural uranium (1 μg of natural U contains 12.3 mBq of ^{238}U) (Sam et al. 1999). Uranium is widespread in nature and occurs in over 160 minerals, locally at high concentrations (between 50 and 90%) (Brzyska 1987, Meinrath et al. 2003). The radionuclides ^{234}U and ^{238}U are not in radioactive equilibrium in the natural environment; this disequilibrium is greater in sea water than in rocks, soils and sediments (Skwarzec 1995, Skwarzec et al. 2004). In southern Baltic waters the mean $^{234}U/^{238}U$ activity ratio was 1.17, whereas in terrestrial materials it was about 1.00 (Skwarzec 1995, Skwarzec et al. 2006). The uranium is leached from rocks, soils and sediments, after which the waters containing the element enter rivers. Under oxidizing conditions, natural uranium exists in river water predominantly in dissolved form as the uranium carbonate anions $[UO_2(CO_3)_2]^{2-}$ and $[UO_2(CO_3)_3]^{4-}$. The uranium concentration of natural river water increases linearly with salinity (Skwarzec 1995).

The principal sources of uranium in the natural environmental are the wet and dry atmospheric fallout of terrigenous matter, as well as human activities, particularly in agriculture (Skwarzec et al. 2002). Coal mining is also a source of huge amounts of waste containing large quantities of natural radionuclides, including uranium. Coals contain about 1% of the whole amount of trace elements and radionuclides in the environment (for example ^{238}U, ^{226}Ra, ^{210}Po and ^{210}Pb). Fly ash contains 1700 Bq kg^{-1} of ^{210}Po and 200 Bq kg^{-1} of ^{238}U. Post-combustion clinker is often used as a supplementary building material to stabilize roads, and as a soil fertilizer. During combustion some radionuclides are emitted to the atmosphere as a gas and radioactive dust, while others remain concentrated in ash (Nakaoka et al. 1984, Baxter 1996, Flues et al. 2002).

The Odra (Oder) is the second longest river in Poland and is one of the major sources of inputs to the Baltic environment. The Odra rises in the

Czech Republic, then flows through western Poland; 187 kilometres of its course form the Polish-German border. It eventually flows into the Szczecin Lagoon north of Szczecin, emptying into the Baltic Sea via three branches – the Dziwna, Świna and Peene. The Odra's drainage basin is 118 861 km^2 in area, 106 050 km^2 (89.2%) of which lies in Poland (Makinia et al. 1996). The Odra and its tributaries flow through the most highly industrialized (fertilizer plants, coal and uranium mines, steelworks, heat and power generating plants) and urbanized (the Lower Silesian Coal Basin) regions of Poland (MKOOpZ 1999). The enhanced concentrations of uranium radionuclides in the Odra and its tributaries may be due to the discharge of waters from coal mines (Michalik et al. 2002). Another source could be the phosphate fertilizers used in agriculture (Martínez-Aguirre et al. 1997, Carvalho et al. 2007): these contain uranium, which can be leached from the catchment area to rivers and lakes by running water. Elevated levels of uranium have been recorded in the water, soils and organisms around phosphogypsum spoil heaps in Poland and other countries (Boryło et al. 2009). A third important source of uranium in the natural environment, not only in the Odra drainage basin, is the atmospheric precipitation of terrigenous matter (Skwarzec et al. 2002, Ruszkiewicz & Świątkowski 2008).

Uranium is usually present in river water as the soluble uranyl ion $UO_2{}^{2+}$ (its progeny are highly radioactive) (Cothern & Lappenbusch 1983). Various uranium compounds can penetrate the blood from lung alveoli or through the gastrointestinal tract; exposure to high doses of uranium can damage the kidneys. However, when uranium is ingested in food or drink, most of it is excreted in the urine within a few days. Small amounts of uranium are taken up and retained by the bones and other tissues, from which they are slowly released. The long-term ingestion of uranium in drinking water led to elevated concentrations of this element in urine, which were detectable up to 10 months after exposure had stopped (ICRP 1994, Leggett 1994, Kenneth et al. 2004). Moreover, experiments carried out on animals showed that gastro-intestinal uranium absorption is much higher in newborns than in adults (Sullivan 1980, Sullivan & Gorham 1982). The preliminary radiological criterion for drinking-water recommended by the World Health Organization is 2 mg m^{-3} uranium; the level set by the US Environmental Protection Agency is 30 mg m^{-3} (WHO 1998, USEPA 2001).

The aim of this work was to determine the uranium isotopes ^{234}U and ^{238}U in river water samples taken from the Odra and its tributaries. The seasonal and annual flow was the basis upon which the total flow of uranium from the Odra drainage basin to the southern Baltic Sea was calculated.

2. Material and methods

River water samples (volume 60 dm^3) were collected seasonally (every three months) at five localities along the main stream of the Odra and at the mouths of eight of its tributaries from October 2003 to July 2004 (Figure 1, Table 1). The radiological methods for the determination of uranium in these samples were based on the coprecipitation of ^{234}U and ^{238}U with manganese dioxide, the mineralization and sequential separation and purification of the uranium on an anion exchange resin (Skwarzec 1997, 2009). Before radiochemical analysis, each sample was enriched with about 50 mBq of ^{232}U as the internal yield tracer. After coprecipitation with MnO_2 and mineralization, the dry residue was dissolved in 8 M nitric acid and the solution passed through an anion-exchange resin (Dowex 1x8; 50–100 mesh). The uranium adsorbed on the resin was eluted using 0.5 M nitric acid and the solution evaporated to dryness. The dry residue was dissolved in 1 M $(NH_4)_2SO_4$ (pH=1.5) and the uranium was purified of iron, cobalt and copper on Dowex 1x8 (100–200 mesh) in the sulphate form. After desorption and elution from the column, the uranium was electrolysed on a stainless steel disc (Skwarzec 1997, 2009). The radioactivity of ^{234}U and ^{238}U was measured using alpha spectrometry equipment with semiconductor silicon detectors and a 300 mm^2 active surface barrier (Canberra-Packard, USA). The accuracy and precision of the radiochemical methods were within 10% based on an international laboratory comparison using International Atomic Energy Agency reference materials (IAEA-384, IAEA-414). The uranium recoveries in the samples ranged between 60% and 95% (Tuszkowska 2009).

Figure 1. Water sample collection points

Table 1. River water sampling dates from the Odra drainage basin

Sampling location	River Odra			
	Autumn	Winter	Spring	Summer
Chałupki	18.10.2003	20.03.2004	22.05.2004	17.07.2004
Głogów	18.10.2003	20.03.2004	22.05.2004	17.07.2004
Słubice	25.10.2003	27.03.2004	29.05.2004	24.07.2004
Gozdowice	25.10.2003	27.03.2004	29.05.2004	24.07.2004
Widuchowa	25.10.2003	27.03.2004	29.05.2004	24.07.2004
	Tributaries of the River Odra			
	Autumn	Winter	Spring	Summer
Mała Panew	18.10.2003	20.03.2004	22.05.2004	17.07.2004
Nysa Kłodzka	18.10.2003	20.03.2004	22.05.2004	17.07.2004
Bystrzyca	18.10.2003	20.03.2004	22.05.2004	17.07.2004
Barycz	18.10.2003	20.03.2004	22.05.2004	17.07.2004
Bóbr	18.10.2003	20.03.2004	22.05.2004	17.07.2004
Nysa Łużycka	18.10.2003	20.03.2004	22.05.2004	17.07.2004
Warta	25.10.2003	27.03.2004	29.05.2004	24.07.2004
Noteć	25.10.2003	27.03.2004	29.05.2004	24.07.2004

The ^{238}U and ^{234}U concentrations in the river water samples are given with the standard deviation calculated for a 95% confidence interval.

3. Results

The concentrations of uranium ^{234}U, ^{238}U and total uranium in the Odra water samples are listed in Table 2. Uranium concentrations were the lowest in summer, but higher in autumn as a consequence of radionuclides being transported with water eluted from the soil, as well as materials leached from the Odra river bed and catchment area. In winter the higher ^{238}U concentrations were due to the elevated levels of this radionuclide in dry and wet atmospheric precipitation resulting from the combustion of coal, gas and oil during the heating season and in snowmelt (the samples collected in March). Among the Odra's tributaries, the Bystrzyca exhibited the highest uranium concentrations in each of the four seasons: from 27.81 ± 0.29 Bq m^{-3} of ^{234}U, 17.82 ± 0.23 Bq m^{-3} of ^{238}U and 1.45 ± 0.02 mg m^{-3} of total uranium in spring to 102.38 ± 2.24 Bq m^{-3} of ^{234}U, 73.13 ± 1.89 Bq m^{-3} of ^{238}U and 5.97 ± 0.15 mg m^{-3} of total uranium in winter (Table 2). The Bystrzyca is a typical mountain river, so the higher concentration of uranium in the river water is mainly the result of higher concentrations of this element in the bedrock. As a consequence, the Bystrzyca is the most highly contaminated river in the Sudeten region. In addition, the anthropogenic impact of uranium in river water is enhanced

Table 2. Average and annual concentrations of ^{234}U and ^{238}U in waters of the River Odra and its tributaries

Sampling location	Autumn		Winter	
	^{234}U [Bq m^{-3}]	^{238}U [Bq m^{-3}]	^{234}U [Bq m^{-3}]	^{238}U [Bq m^{-3}]
		River Odra		
Chałupki	9.28 ± 0.19	5.45 ± 0.14	2.43 ± 0.09	1.71 ± 0.07
Głogów	27.08 ± 0.50	14.69 ± 0.37	12.30 ± 0.11	7.59 ± 0.09
Słubice	18.62 ± 0.25	10.31 ± 0.19	14.14 ± 0.25	8.47 ± 0.20
Gozdowice	9.30 ± 0.11	6.73 ± 0.09	9.09 ± 0.31	7.60 ± 0.29
Widuchowa	10.34 ± 0.24	6.95 ± 0.19	7.19 ± 0.16	5.74 ± 0.14
		Tributaries of the River Odra		
Mała Panew	3.52 ± 0.05	2.59 ± 0.04	2.71 ± 0.07	1.92 ± 0.06
Nysa Kłodzka	19.92 ± 0.55	13.06 ± 0.44	9.68 ± 0.38	7.32 ± 0.33
Bystrzyca	75.76 ± 0.87	47.14 ± 0.69	102.38 ± 2.24	73.13 ± 1.89
Barycz	5.17 ± 0.11	4.31 ± 0.10	15.30 ± 0.47	13.00 ± 0.44
Bóbr	4.93 ± 0.12	3.42 ± 0.10	7.55 ± 0.16	5.27 ± 0.13
Nysa Łużycka	4.08 ± 0.13	3.72 ± 0.12	3.66 ± 0.04	3.38 ± 0.04
Warta	7.85 ± 0.12	7.05 ± 0.12	6.44 ± 0.70	5.96 ± 0.68
Noteć	8.54 ± 0.35	7.76 ± 0.30	7.17 ± 0.24	6.93 ± 0.24
	Spring		Summer	
	^{234}U [Bq m^{-3}]	^{238}U [Bq m^{-3}]	^{234}U [Bq m^{-3}]	^{238}U [Bq m^{-3}]
		River Odra		
Chałupki	4.11 ± 0.07	2.54 ± 0.06	9.60 ± 0.26	5.93 ± 0.21
Głogów	11.58 ± 0.26	7.51 ± 0.21	22.79 ± 0.75	14.65 ± 0.61
Słubice	17.75 ± 0.53	10.27 ± 0.40	17.00 ± 0.56	11.14 ± 0.45
Gozdowice	10.74 ± 0.28	8.40 ± 0.25	9.91 ± 0.34	7.45 ± 0.30
Widuchowa	18.39 ± 0.54	14.13 ± 0.47	9.38 ± 0.34	6.98 ± 0.29
		Tributaries of the River Odra		
Mała Panew	1.33 ± 0.02	1.06 ± 0.02	3.26 ± 0.13	2.19 ± 0.10
Nysa Kłodzka	13.60 ± 0.08	9.85 ± 0.07	16.76 ± 0.14	12.68 ± 0.13
Bystrzyca	27.81 ± 0.29	17.82 ± 0.23	194.76 ± 3.43	134.88 ± 2.85
Barycz	7.20 ± 0.19	6.39 ± 0.18	4.39 ± 0.12	3.69 ± 0.11
Bóbr	11.37 ± 0.33	8.83 ± 0.29	5.47 ± 0.16	3.41 ± 0.12
Nysa Łużycka	1.58 ± 0.06	1.39 ± 0.06	5.80 ± 0.07	4.89 ± 0.07
Warta	5.38 ± 0.19	4.89 ± 0.18	6.55 ± 0.19	6.18 ± 0.19
Noteć	3.28 ± 0.12	3.18 ± 0.12	7.00 ± 0.23	6.35 ± 0.21

Table 2. (*continued*)

Sampling location	Annual average value	
	^{234}U [Bq m^{-3}]	^{238}U [Bq m^{-3}]
	River Odra	
Chałupki	6.36 ± 0.15	3.91 ± 0.12
Głogów	18.44 ± 0.41	11.11 ± 0.32
Słubice	16.88 ± 0.40	10.05 ± 0.31
Gozdowice	9.76 ± 0.26	7.55 ± 0.23
Widuchowa	11.33 ± 0.32	8.45 ± 0.27
	Tributaries of the River Odra	
Mała Panew	2.71 ± 0.07	1.94 ± 0.06
Nysa Kłodzka	14.99 ± 0.29	10.73 ± 0.24
Bystrzyca	100.18 ± 1.71	68.24 ± 1.42
Barycz	8.02 ± 0.22	6.85 ± 0.21
Bóbr	7.33 ± 0.19	5.23 ± 0.16
Nysa Łużycka	3.78 ± 0.08	3.35 ± 0.07
Warta	6.56 ± 0.30	6.02 ± 0.29
Noteć	6.50 ± 0.24	6.06 ± 0.22

by coal mining in the Lower Silesian Coal Basin and the use of phosphate fertilizers in agriculture (MKOOpZ 1999, Boryło et al. 2009).

The average annual concentrations of uranium in the river water samples were calculated from the average seasonal concentration of this element in the Odra and its tributaries,; the results are given in Tables 2 and 3, and Figures 2 and 3. In the Odra itself, the highest uranium concentration was recorded at Głogów (18.44 ± 0.41 Bq m^{-3} of ^{234}U, 11.11 ± 0.32 Bq m^{-3} of ^{238}U and 0.91 ± 0.03 mg m^{-3} of total uranium), and the lowest value at Chałupki (6.36 ± 0.15 Bq m^{-3} of ^{234}U, 3.91 ± 0.12 Bq m^{-3} of ^{238}U and 0.32 ± 0.01 mg m^{-3} of total uranium). Of all the Odra's tributaries, the Bystrzyca was characterized by the highest uranium concentration (100.18 ± 1.71 Bq m^{-3} of ^{234}U, 68.24 ± 1.24 Bq m^{-3} of ^{238}U and 5.57 ± 0.12 mg m^{-3} of total uranium).

^{234}U and ^{238}U are not in radioactive equilibrium in the water samples from the Odra drainage basin (Table 3). The annual average $^{234}U/^{238}U$ activity ratio in Odra mainstream water varied from 1.30 ± 0.05 at Gozdowice to 1.69 ± 0.07 at Słubice. All the river water samples from Widuchowa had a slightly higher $^{234}U/^{238}U$ activity ratio than those from

Table 3. Average annual total uranium and $^{234}U/^{238}U$ activity ratio in the waters of the River Odra and its tributaries

Sampling location	Autumn: U total [mg m^{-3}]	Autumn: $^{234}U/^{238}U$	Winter: U total [mg m^{-3}]	Winter: $^{234}U/^{238}U$
	River Odra			
Chałupki	0.44 ± 0.01	1.70 ± 0.06	0.14 ± 0.01	1.42 ± 0.08
Głogów	1.20 ± 0.03	1.84 ± 0.06	0.62 ± 0.01	1.62 ± 0.02
Słubice	0.84 ± 0.02	1.81 ± 0.04	0.69 ± 0.02	1.67 ± 0.05
Gozdowice	0.55 ± 0.01	1.38 ± 0.03	0.62 ± 0.02	1.20 ± 0.06
Widuchowa	0.57 ± 0.02	1.49 ± 0.05	0.47 ± 0.01	1.25 ± 0.04
	Tributaries of the River Odra			
Mała Panew	0.21 ± 0.01	1.36 ± 0.03	0.16 ± 0.005	1.41 ± 0.05
Nysa Kłodzka	1.07 ± 0.04	1.53 ± 0.07	0.60 ± 0.03	1.32 ± 0.08
Bystrzyca	3.85 ± 0.06	1.61 ± 0.03	5.97 ± 0.15	1.40 ± 0.05
Barycz	0.35 ± 0.01	1.20 ± 0.04	1.06 ± 0.04	1.18 ± 0.05
Bóbr	0.28 ± 0.01	1.44 ± 0.06	0.43 ± 0.01	1.43 ± 0.05
Nysa Łużycka	0.30 ± 0.01	1.10 ± 0.05	0.28 ± 0.01	1.08 ± 0.02
Warta	0.58 ± 0.01	1.11 ± 0.03	0.49 ± 0.06	1.08 ± 0.03
Noteć	0.63 ± 0.02	1.10 ± 0.03	0.57 ± 0.02	1.03 ± 0.05

Sampling location	Spring: U total [mg m^{-3}]	Spring: $^{234}U/^{238}U$	Summer: U total [mg m^{-3}]	Summer: $^{234}U/^{238}U$
	River Odra			
Chałupki	0.21 ± 0.01	1.62 ± 0.05	0.48 ± 0.02	1.62 ± 0.07
Głogów	0.61 ± 0.02	1.54 ± 0.05	1.20 ± 0.05	1.56 ± 0.07
Słubice	0.84 ± 0.03	1.73 ± 0.09	0.91 ± 0.04	1.53 ± 0.08
Gozdowice	0.69 ± 0.02	1.28 ± 0.05	0.61 ± 0.02	1.33 ± 0.07
Widuchowa	1.15 ± 0.04	1.30 ± 0.06	0.57 ± 0.02	1.34 ± 0.07
	Tributaries of the River Odra			
Mała Panew	0.09 ± 0.01	1.25 ± 0.03	0.18 ± 0.01	1.49 ± 0.09
Nysa Kłodzka	0.80 ± 0.01	1.38 ± 0.04	1.03 ± 0.01	1.32 ± 0.02
Bystrzyca	1.45 ± 0.02	1.56 ± 0.03	11.01 ± 0.23	1.44 ± 0.04
Barycz	0.52 ± 0.01	1.13 ± 0.04	0.30 ± 0.01	1.19 ± 0.05
Bóbr	0.72 ± 0.02	1.29 ± 0.06	0.28 ± 0.01	1.60 ± 0.07
Nysa Łużycka	0.11 ± 0.01	1.14 ± 0.07	0.40 ± 0.01	1.19 ± 0.02
Warta	0.40 ± 0.01	1.10 ± 0.06	0.50 ± 0.02	1.06 ± 0.04
Noteć	0.26 ± 0.01	1.03 ± 0.06	0.52 ± 0.02	1.10 ± 0.05

Table 3. (*continued*)

Sampling location	Annual average value	
	U total [mg m^{-3}]	$^{234}U/^{238}U$
	River Odra	
Chałupki	0.32 ± 0.01	1.59 ± 0.07
Głogów	0.91 ± 0.03	1.64 ± 0.05
Słubice	0.82 ± 0.03	1.69 ± 0.07
Gozdowice	0.62 ± 0.02	1.30 ± 0.05
Widuchowa	0.69 ± 0.02	1.35 ± 0.06
	Tributaries of the River Odra	
Mała Panew	0.16 ± 0.01	1.38 ± 0.05
Nysa Kłodzka	0.88 ± 0.02	1.39 ± 0.05
Bystrzyca	5.57 ± 0.12	1.50 ± 0.04
Barycz	0.56 ± 0.02	1.18 ± 0.05
Bóbr	0.43 ± 0.01	1.44 ± 0.06
Nysa Łużycka	0.27 ± 0.01	1.13 ± 0.04
Warta	0.49 ± 0.03	1.09 ± 0.04
Noteć	0.50 ± 0.02	1.07 ± 0.05

Gozdowice. These differences can be explained by the specificity of the Odra estuary, which is influenced by sea waters from the southern Baltic. The Odra estuary is a mixing zone with abrupt thermodynamic changes, where sorption onto colloids and larger particle phases at low salinities affects the

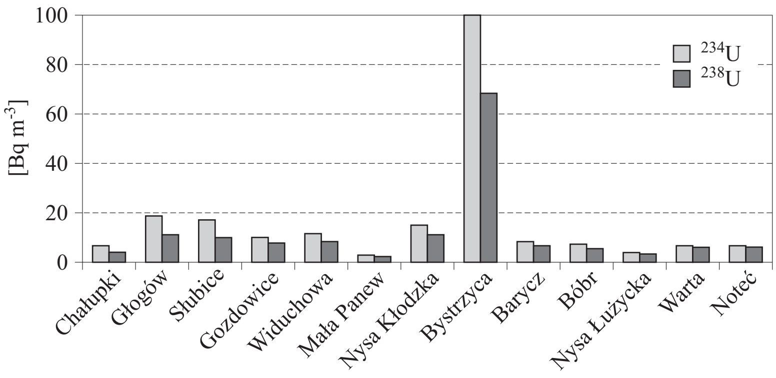

Figure 2. Average annual concentration of ^{234}U and ^{238}U in the waters of the Odra and its tributaries [Bq m^{-3}]

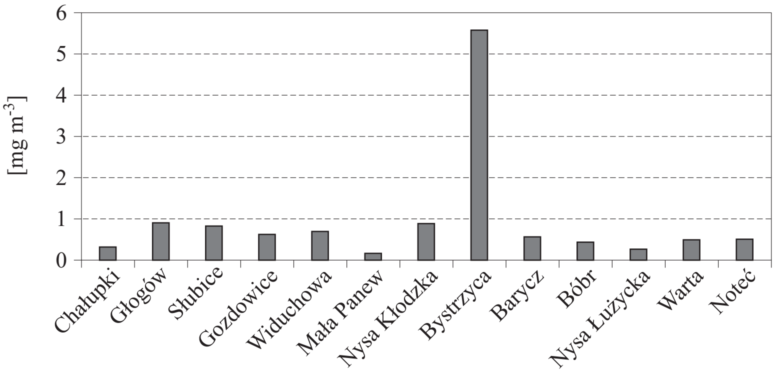

Figure 3. Average annual concentration of total uranium in the waters of the Odra and its tributaries [mg m^{-3}]

behaviour of uranium (Andersson et al. 2001). The highest $^{234}U/^{238}U$ activity ratio in the Odra's tributaries was recorded in the water samples from the Bystrzyca (from 1.40 ± 0.05 in winter to 1.61 ± 0.03 in autumn) and the Bóbr (from 1.29 ± 0.06 in spring to 1.60 ± 0.07 in summer); the activity ratios were lowest in water from the Warta (from 1.06 ± 0.04 in summer to 1.11 ± 0.03 in autumn) and the Noteć (from 1.03 ± 0.05 in winter and spring to 1.10 ± 0.03 in summer and autumn) (Table 3).

Table 4. Seasonal and average annual water flow in the River Odra and its tributaries

Sampling location	Autumn [m^3 s^{-3}]	Winter [m^3 s^{-3}]	Spring [m^3 s^{-3}]	Summer [m^3 s^{-3}]	Average value [m^3 s^{-3}]
			River Odra		
Chałupki	19	240	20.4	11.4	72.7
Głogów	81.5	393	199	61.5	183.7
Słubice	118	583	230	125	264
Gozdowice	248	765	409	238	415
Widuchowa	251	726	426	235	409.5
		Tributaries of the River Odra			
Mała Panew	2.4	2.2	16.5	2.6	6
Nysa Kłodzka	11.2	28.7	35	11.2	21.5
Bystrzyca	2.1	5.4	7.7	4	4.8
Barycz	1.38	11.9	10.6	8.5	8.1
Bóbr	14.4	61	27.6	15.4	29.6
Nysa Łużycka	9.16	25.5	14	9.45	14.5
Warta	107	223	154	94.2	144.5
Noteć	47.5	87.1	55.7	39.4	57.4

Table 5. Seasonal and annual runoff of ^{234}U and ^{238}U from the River Odra drainage basin

Sampling location	Autumn			Winter		
	^{234}U	^{238}U	U total	^{234}U	^{238}U	U total
	[GBq quarter^{-1}]		[kg quarter^{-1}]	[GBq quarter^{-1}]		[kg quarter^{-1}]
			River Odra			
Chałupki	1.40	0.82	66	4.59	3.23	264
Głogów	17.54	9.52	777	38.01	23.45	1916
Słubice	17.46	9.67	788	64.81	38.82	3163
Gozdowice	18.33	13.27	1084	54.67	45.71	3729
Widuchowa	20.63	13.87	1137	41.04	32.76	2683
			Tributaries of the River Odra			
Mała Panew	0.07	0.05	4	0.05	0.03	3
Nysa Kłodzka	1.77	1.16	95	2.18	1.65	135
Bystrzyca	1.25	0.78	64	4.31	3.08	251
Barycz	0.06	0.05	4	1.43	1.22	99
Bóbr	0.56	0.39	32	3.62	2.53	206
Nysa Łużycka	0.30	0.27	22	0.73	0.68	55
Warta	6.68	5.99	489	11.29	10.45	859
Noteć	3.22	2.93	238	4.91	4.75	387
	Spring			Summer		
	^{234}U	^{238}U	U total	^{234}U	^{238}U	U total
	[GBq quarter^{-1}]		[kg quarter^{-1}]	[GBq quarter^{-1}]		[kg quarter^{-1}]
			River Odra			
Chałupki	0.66	0.41	34	0.87	0.54	43
Głogów	18.12	11.75	954	11.14	7.16	587
Słubice	32.10	18.57	1519	16.89	11.07	904
Gozdowice	34.54	27.01	2219	18.75	14.09	1154
Widuchowa	61.60	47.33	3852	17.52	13.04	1065
			Tributaries of the River Odra			
Mała Panew	0.17	0.14	12	0.07	0.05	4
Nysa Kłodzka	3.74	2.71	221	1.49	1.13	92
Bystrzyca	1.68	1.08	88	6.25	4.33	353
Barycz	0.60	0.53	43	0.30	0.25	20
Bóbr	2.47	1.92	156	0.67	0.42	34
Nysa Łużycka	0.17	0.15	13	0.44	0.37	30
Warta	6.52	5.92	483	4.90	4.63	378
Noteć	1.44	1.39	114	2.19	1.99	162

Table 5. (*continued*)

Sampling location	Annual average value		
	^{234}U	^{238}U	U total
	[GBq quarter^{-1}]		[kg quarter^{-1}]
	River Odra		
Chałupki	7.52	4.99	0.41
Głogów	84.81	51.88	4.23
Słubice	131.27	78.14	6.37
Gozdowice	126.29	100.08	8.19
Widuchowa	140.79	107.00	8.74
	Tributaries of the River Odra		
Mała Panew	0.36	0.27	0.02
Nysa Kłodzka	9.19	6.65	0.54
Bystrzyca	13.50	9.27	0.76
Barycz	2.38	2.05	0.17
Bóbr	7.32	5.25	0.43
Nysa Łużycka	1.64	1.47	0.12
Warta	29.39	26.99	2.21
Noteć	11.76	11.06	0.90

The seasonal and annual uranium runoff from the Odra drainage basin was calculated on the basis of the average seasonal and annual water flow of the Odra and its tributaries (Table 4) – the results are listed in Table 5. The highest quantity of uranium was transported by the Odra itself, especially at Widuchowa (from 17.52 GBq quarter^{-1} of ^{234}U, 13.04 GBq quarter^{-1} of ^{238}U and 1065 kg quarter^{-1} of total uranium in summer to 61.60 GBq quarter^{-1} of ^{234}U, 47.33 GBq quarter^{-1} of ^{238}U and 3852 kg quarter^{-1} of total uranium in spring), whereas the lowest levels were recorded at Chałupki (from 0.66 GBq quarter^{-1} of ^{234}U, 0.41 GBq quarter^{-1} of ^{238}U and 34 kg quarter^{-1} of total uranium in spring to 4.59 GBq quarter^{-1} of ^{234}U, 3.23 GBq quarter^{-1} of ^{238}U and 264 kg quarter^{-1} of total uranium in winter). Among the Odra's tributaries the highest uranium runoff was transported by the Warta (from 4.90 GBq quarter^{-1} of ^{234}U, 4.63 GBq quarter^{-1} of ^{238}U and 378 kg quarter^{-1} of total uranium in summer to 11.29 GBq quarter^{-1} of ^{234}U, 10.49 GBq quarter^{-1} of ^{238}U and 859 kg quarter^{-1} of total uranium in winter); the lowest levels were in the Mała Panew (from 0.05 GBq quarter^{-1} of ^{234}U, 0.03 GBq quarter^{-1} of ^{238}U and 3 kg quarter^{-1} of total uranium in winter to 0.17 GBq quarter^{-1} of ^{234}U, 0.14 GBq quarter^{-1} of ^{238}U and 12 kg quarter^{-1} of total uranium in spring). Annually, the highest amount of uranium transported by the Odra was measured at Widuchowa (140.79 GBq year^{-1} of ^{234}U, 107.00 GBq year^{-1} of ^{238}U and 8.74 tons year^{-1} of total uranium) and the lowest at Chałupki (7.52 GBq year^{-1} of ^{234}U, 4.99 GBq year^{-1} of ^{238}U and 0.41 tons year^{-1} of total uranium). Of the Odra's tributaries, the Warta carried the largest quantities of uranium (29.39 GBq year^{-1} of ^{234}U, 26.99 GBq year^{-1} of ^{238}U and 2.21 tons year^{-1} of total uranium), while the smallest amounts were carried by the Mała Panew (0.36 GBq year^{-1} of ^{234}U, 0.27 GBq year^{-1} of ^{238}U and 0.02 tons year^{-1} of total uranium) (Table 5). The results of this investigation thus indicate that the River Odra is an important source of uranium in the southern Baltic environment. Annually, the Szczecin Lagoon receives 126.29 GBq of ^{234}U and 100.80 GBq of ^{238}U (8.19 tons of total uranium) (Table 6).

Table 6. Inflow of uranium from the River Odra to the Baltic Sea

Season	Total inflow		
	^{234}U	^{238}U	U total
	[GBq year^{-1}]		[ton year^{-1}]
autumn	18.33	13.27	1.08
winter	54.67	45.71	3.73
spring	34.54	27.01	2.22
summer	18.75	14.09	1.15
all year	126.29	100.80	8.19

The seasonal and annual surface runoff of uranium per unit area was calculated on the basis of the area of the Odra drainage basin; the relevant data are presented in Table 7 and in Figures 4 and 5. Seasonally, the surface runoff of uranium in the main stream of the Odra was higher in winter and spring than in autumn and summer. During the year the highest surface runoffs of ^{234}U, ^{238}U and total uranium in the Odra drainage basin were recorded at Słubice, the lowest at Gozdowice. Annually, the uranium surface runoff lies between 75 and 119 g year^{-1} km^{-2} (from 911 to 1460 kBq year^{-1} km^{-2} of ^{238}U). Among the Odra's tributaries the greatest surface runoff of uranium for all seasons was recorded in the catchment area of the Bystrzyca (^{238}U – from 441 kBq year^{-1} km^{-2} in autumn to 2450 kBq year^{-1} km^{-2} in summer), but the least in the catchment areas of the Barycz and Mała Panew. Also, the highest annual surface flows were recorded in the catchment area of the Bystrzyca (7639 kBq year^{-1} km^{-2} of ^{234}U, 5244 kBq year^{-1} km^{-2} of ^{238}U and 428 g year^{-1} km^{-2} of total uranium), and the lowest in that of the Mała Panew (168 kBq year^{-1} km^{-2}

Table 7. Seasonal and annual surface runoff of ^{234}U and ^{238}U from the catchment areas of the Odra and its tributaries

Sampling location	Autumn			Winter		
	^{234}U [kBq quarter^{-1} km^{-2}]	^{238}U [kBq quarter^{-1} km^{-2}]	U total [g quarter^{-1} km^{-2}]	^{234}U [kBq quarter^{-1} km^{-2}]	^{238}U [kBq quarter^{-1} km^{-2}]	U total [g quarter^{-1} km^{-2}]
River Odra						
Chałupki	301	176	14	983	692	57
Głogów	482	261	21	1044	644	53
Słubice	326	181	15	1211	725	59
Gozdowice	167	121	10	498	416	34
Widuchowa	187	125	10	371	296	24
Tributaries of the River Odra						
Mała Panew	32	24	2	22	16	1
Nysa Kłodzka	388	255	21	478	362	30
Bystrzyca	709	441	36	2440	1743	142
Barycz	10	9	1	259	220	18
Bóbr	96	67	5	616	430	35
Nysa Łużycka	69	63	5	171	158	13
Warta	122	110	9	207	192	16
Noteć	186	169	14	283	274	22
	Spring			Summer		
	^{234}U [kBq quarter^{-1} km^{-2}]	^{238}U [kBq quarter^{-1} km^{-2}]	U total [g quarter^{-1} km^{-2}]	^{234}U [kBq quarter^{-1} km^{-2}]	^{238}U [kBq quarter^{-1} km^{-2}]	U total [g quarter^{-1} km^{-2}]
River Odra						
Chałupki	141	87	7	187	115	9
Głogów	498	323	26	306	197	16
Słubice	600	347	28	316	207	17
Gozdowice	315	246	20	171	128	11
Widuchowa	557	428	35	158	118	10
Tributaries of the River Odra						
Mała Panew	81	64	5	32	22	2
Nysa Kłodzka	820	594	48	327	247	20
Bystrzyca	952	610	50	3538	2450	200
Barycz	108	96	8	54	45	4
Bóbr	420	326	27	114	71	6
Nysa Łużycka	41	36	3	101	85	7
Warta	120	109	9	90	85	7
Noteć	83	80	7	126	115	9

Table 7. (*continued*)

Sampling location	Annual average value		
	^{234}U	^{238}U	U total
	[kBq quarter^{-1} km^{-2}]		[g quarter^{-1} km^{-2}]
	River Odra		
Chałupki	1612	1070	87
Głogów	2330	1425	116
Słubice	2453	1460	119
Gozdowice	1151	911	75
Widuchowa	1273	967	79
	Tributaries of the River Odra		
Mała Panew	168	126	10
Nysa Kłodzka	2014	1457	119
Bystrzyca	7639	5244	428
Barycz	431	370	31
Bóbr	1246	894	73
Nysa Łużycka	381	342	28
Warta	539	495	41
Noteć	678	638	52

of ^{234}U, 126 kBq year^{-1} km^{-2} of ^{238}U and 10 g year^{-1} km^{-2} of total uranium). Seasonally, the surface runoff of uranium was higher in winter and spring than in autumn and summer. In the Odra's tributaries the highest values of ^{238}U surface runoff were reported from the Bystrzyca (from 441 kBq quarter^{-1} km^{-2} in autumn to 2450 kBq quarter^{-1} km^{-2} in summer).

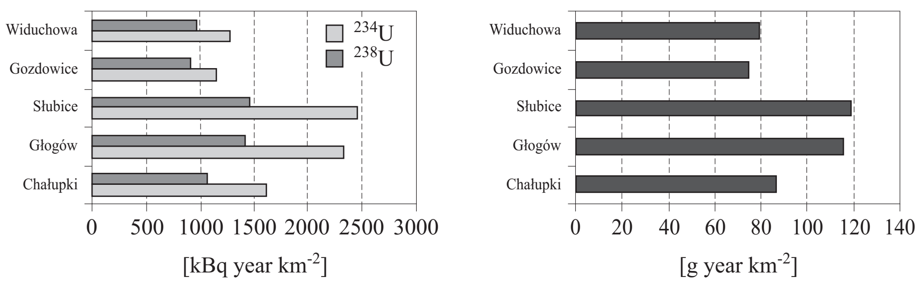

Figure 4. Annual surface runoff of uranium: a) ^{234}U and ^{238}U [kBq year km^{-2}], b) total uranium from the Odra drainage basin [g year km^{-2}]

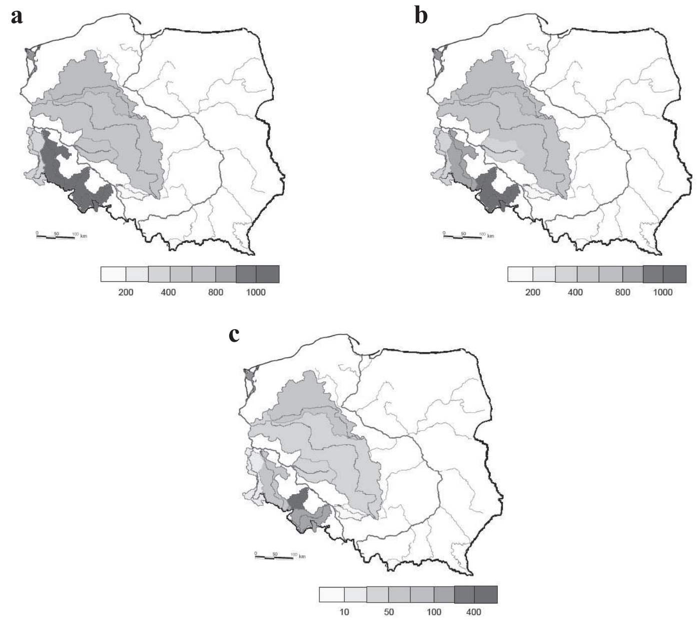

Figure 5. Annual surface runoff of uranium: a) ^{234}U [kBq year^{-1} km^{-2}], b) ^{238}U [kBq year^{-1} km^{-2}] and c) total uranium [g year^{-1} km^{-2}] from the Odra drainage basin

4. Discussion and conclusions

The inflow of uranium from the Odra drainage basin to the Baltic is due to human activities there, mainly in agriculture (phosphate fertilizers) and the mining industry (discharge of mine waters into the river). Phosphate-bearing rocks usually contain radionuclides of uranium (^{234}U and ^{238}U) and radium (^{226}Ra), the uranium concentration being around 130 ppm (Bem 2005). The phosphate-bearing rocks used to produce phosphate fertilizers contain from 1300 to 1500 Bq kg^{-1} of uranium ^{238}U and its decay products (Bolivar et al. 1995). During the production of these fertilizers ca 10% of the initial amount of ^{226}Ra, 20% of the uranium and about 85% of the ^{210}Po ends up in phosphogypsum spoil (Carvalho 1995, Carvalho et al. 2007). Having elevated concentrations of natural radionuclides, phosphogypsum can seriously pollute ground waters and river waters; this is also the case in Poland (Boryło et al. 2009). In the neighbourhood of the phosphate fertilizer plant in Gdańsk (near the mouth of the river Vistula) and at Police (Odra mouth), radionuclide concentrations in soil and river water samples are much higher than in uncontaminated samples (Boryło et al. 2009). Moreover, the surroundings of the phosphogypsum spoil tip at Wiślinka (near Gdańsk) are badly polluted by polonium and uranium (Skwarzec 2009). Saline waters from underground coal mines usually contain the natural radioactive isotopes ^{226}Ra and ^{228}Ra. In the Odra drainage basin coal mines generally produce type A water, which contains high concentrations of radium and barium, but not of sulphate ions; this type of water contains highly radioactive deposits formed by the co-precipitation of barium and radium. It was calculated that about 20 MBq per day of ^{226}Ra and 10 MBq per day of ^{228}Ra enter Odra waters (Tomza & Lebecka 1981, Chałupnik et al. 2001). But despite the fact that uranium used to be mined in the Kowary, Radoniów and Kletno areas, there were no gross increases in uranium concentrations in the waters of the rivers Bóbr and Nysa Kłodzka assayed near the mining region.

If we exclude the water samples taken from the Bystrzyca, the average uranium concentration in the Odra and its tributaries is similar to that in the Vistula and in drinking water drawn from central Poland, but is lower than the values reported from the Rivers Jucar and Ortigas in Spain and from Turkish rivers (Rodriguez-Alvarez & Sanchez 1995, Vargas et al. 1995, Kumru 1997, Pietrzak-Flis et al. 1997). Odra waters are consumed by the people living in the surrounding areas. The respective average uranium ^{234}U and ^{238}U concentrations in drinking water drawn from Polish rivers (1992–1996) lie between 10.2–15.0 and 7.3-8.3 Bq m^{-3}; the ^{234}U/^{238}U activity ratio was 1.23 ± 0.40 (Pietrzak-Flis et al. 1997). Drinking water may be the source of 75.1% of ^{238}U and 76.9% of ^{234}U found in the human body. The effective annual radiation dose for humans has been calculated at 5.95 ± 0.21 μSv, the largest contribution coming from ^{226}Ra (72.4 %). Uranium (^{234}U and ^{238}U) and thorium (^{232}Th, ^{230}Th and ^{235}Th) radionuclides were the respective sources of 14.1% and 13.5% alpha emitters in the human body (Pietrzak-Flis et al. 2001, Skwarzec et al. 2001, 2003). Consumption of water from the Bystrzyca by wild animals and humans could be dangerous because the primary radiological criterion for drinking-water is a gross alpha radioactivity of 100 Bq m^{-3} or a gross beta radioactivity of 100 Bq m^{-3} (WHO 1998). The higher concentrations of uranium in the Bystrzyca can be explained by the outflow of mine waters, the proximity of quarries and the application of phosphorus fertilizers in agriculture, as well as elevated surface runoff and a greater subterranean water flow resulting from higher rainfall. Our results for the Bystrzyca are comparable with those obtained in the United States by Longtin (1988), where the mean uranium concentration was 1.86 mg m^{-3} and 3% of the samples analysed had a uranium concentration of > 10 mg m^{-3} (Kenneth et al. 2004).

The $^{234}U/^{238}U$ activity ratio in natural waters is not constant: it can be applied in chronology, and used to determine the origin of waters and to track hydrological processes (Skwarzec 1995, Skwarzec et al. 2004). The waters of the River Odra have higher uranium concentrations and higher $^{234}U/^{238}U$ activity ratios than those of the southern Baltic (1.17). There was a radioactive disequilibrium between the activities of ^{234}U and ^{238}U in all the river water samples analysed for each season (Table 3). The $^{234}U/^{238}U$ activity ratio ranged from 1.07 to 1.69 and decreased from the Odra's source to its estuary, probably because of the increasing salinity of the river's water as it approaches the sea (Chabaux et al. 2003). Generally, rivers originating in mountain areas have higher $^{234}U/^{238}U$ activity ratios than rivers with lowland sources. The $^{234}U/^{238}U$ activity ratio of waters of rivers in humid regions is much the same as that in the bedrocks that they drain (Kronfeld & Vogel 1991, Kronfeld et al. 2004).

The results of this investigation indicate that the River Odra is an important source of uranium radionuclides in the southern Baltic environment. Annually, the southern Baltic Sea receives 126.29 GBq of ^{234}U and 100.80 GBq of ^{238}U (8.19 tons of total uranium). For comparison, Vistula river waters transport to the Baltic Sea ca 276.80 GBq year^{-1} of ^{234}U, 230.60 GBq year^{-1} of ^{238}U (18.80 ton total uranium every year) (Skwarzec et al. 2010); in contrast, the River Mahandi in India transports ca 36 tons of uranium per year (Ray et al. 1995).

The study shows that the high uranium concentrations in the Odra drainage basin are the result of using phosphate fertilizers in agriculture and the discharge of saline waters from coal mines. It was calculated that about 30% of the total uranium entering the Baltic Sea from Poland do so via Odra river waters.

References

Andersson P. S., Porcelli D., Gustafsson Ö., Ingri J., Wasserburg G. J., 2001, *The importance of colloids for the behavior of uranium isotopes in the low-salinity zone of a stable estuary*, Geochim. Cosmochim. Ac., 65 (1), 13–25.

Bagatti D., Cantone M. C., Giussani A., Veronese I., Roth P., Werner E., Höllriegl V., 2003, *Regional dependence of urinary uranium baseline levels in non-exposed subjects with particular reference to volunteers from Northern Italy*, J. Environ. Radioactiv., 65 (3), 357–364.

Baxter M. S., 1996, *Technologically enhanced radioactivity: Another view*, J. Environ. Radioactiv., 32 (1–2), 3–17.

Bem H., 2005, *Radioaktywność w środowisku naturalnym (Radioacivity in the natural environment)*, Komisja Ochrony Środowiska, PAN, Łódź, 100 pp.

Bolivar J. P., García-Tenorio R., García-León M., 1995, *Fluxes and distribution of natural radionuclides in the production and use of fertilizers*, Appl. Radiat. Isotopes, 46 (6/7), 717–718.

Boryło A., Nowicki W., Skwarzec B., 2009, *Isotopes of polonium (^{210}Po) and uranium (^{234}U and ^{238}U) in the industrialised area of Wiślinka (North Poland)*, Int. J. Environ. An. Ch., 89 (8–12), 677–685.

Bou-Rabee F., Bakir Y., Bem H., 1995, *Contribution of uranium to gross alpha radioactivity in some environmental samples in Kuwait*, Environ. Int., 21 (3), 293–298.

Brzyska W., 1987, *Lantanowce i aktynowce*, WNT, Warszawa, 172 pp.

Carvalho F. P., 1995, *^{210}Pb and ^{210}Po in sediments and suspended matter in the Tagus estuary, Portugal; local enhancement of natural levels by wastes from phosphate ore processing industry*, Sci.Total Environ., 159 (2–3), 201–214.

Carvalho F. P., Oliveira J. M., Lopes I., Batista A., 2007, *Radionuclides from past uranium mining in rivers of Portugal*, J. Environ. Radioactiv., 98 (3), 298–314.

Chabaux F., Riotte J., Dequincey O., 2003, *U-Th-Ra fractionation during weathering and river transport*, [in:] *Uranium-series geochemistry*, B. Bourdon, G. M. Henderson, C. C. Lundstrom & S. P. Turner (eds.), Mineral. Soc. Amer., 533–576.

Chałupnik S., Michalik B., Wysocka M., Skubacz K., Mielnikow A., 2001, *Contamination of settling ponds and rivers as a result of discharge of radium-bearing waters from Polish coal mines*, J. Environ. Radioactiv., 54 (1), 85–98.

Cothern C. R., Lappenbusch W. L., 1983, *Occurrence of uranium in drinking water in US*, Health Phys., 45 (1), 89–99.

Flues M., Moraes V., Mazzilli B. P., 2002, *The influence of a coal-fired plant operation on radionuclides in soil*, J. Environ. Radioactiv., 63 (3), 285–294.

ICRP – International Commission on Radiological Protection, 1994, *Age-dependent doses to members of the public from intake of Radionuclides. Part 3. Ingestion dose coefficients*, ICRP Publ. 69, Pergamon Press, New York, 74 pp.

Kenneth K. G., Orloff G., Mistry K., Charp P., Metcalf S., Marino R., Shelly T., Melaro E., Donohoe A. M., Jones R. L., 2004, *Human exposure to uranium in groundwater*, Environ. Res., 94 (3), 319–326.

Kronfeld J., Godfrey-Smith D. I., Johannessen D., Zentilli M., 2004, *Uranium series isotopes in the Avon Valley, Nova Scotia*, J. Environ. Radioactiv., 73 (3), 335–352.

Kronfeld J., Vogel J. C., 1991, *Uranium isotopes in surface waters from southern Africa*, Earth Planet. Sci. Lett., 105 (1–3), 65–73.

Kumru M. N., 1997, *Possible uranium rich areas in the Aegean region of Turkey*, Appl. Radiat. Isotopes, 48 (2), 295–299.

Legett R. W., 1994, *Basis for the ICRP's age-specific biokinetic model for uranium*, Health Phys., 67 (6), 589–610.

Longtin J. P., 1988, *Occurrence of radon, radium and uranium in ground water*, J. Am. Water Works Ass., 80 (7), 84–93.

Makinia J., Dunnette D., Kowalik P., 1996, *Water pollution in Poland*, Eur. Water Pollut. Contr., 6 (2), 26–33.

Martinez-Aguirre A., Garcia-Orellana I., Garcia-León M., 1997, *Transfer of natural radionuclides from soils to plants in a marsh enhanced by the operation of non-nuclear industries*, J. Environ. Radioactiv., 35 (2), 149–171.

Meinrath A., Schneider P., Meinrath G., 2003, *Uranium ores and depleted uranium in the environment with a reference to uranium in the biosphere from the Erzgebrige/Sachsen Germany*, J. Environ. Radioactiv., 64 (2–3), 175–193.

Michalik B., Chałupnik S., Wysocka M., Skubacz K., 2002, *Ecological problem of the coal industry and the ways to solve them*, J. Min. Sci., 38 (6), 601–607.

MKOOpZ – Międzynarodowa Komisja Ochrony Odry przed Zanieczyszczeniem, 1999, *Program szybkiego działania dla ochrony rzeki Odry przed zanieczyszczeniem 1997–2002*, MKOOpZ, Wrocław.

Nakaoka A., Fukushima M., Takagi S., 1984, *Environmental effects of natural radionuclides from coal-fired power plants*, Health Phys., 47 (3), 407–416.

Pietrzak-Flis Z., Rosiak L., Suplińska M. M., Chrzanowski E., Dembinska S., 2001, *Daily intakes of ^{238}U, ^{234}U, ^{232}Th, ^{230}Th, ^{238}Th and ^{226}Ra in the adult population of central Poland*, Sci. Total Environ., 273 (1–3), 163–169.

Pietrzak-Flis Z., Suplińska M. M., Rosiak L., 1997, *The dietary intake of ^{238}U, ^{234}U, ^{232}Th, ^{230}Th, ^{238}Th and ^{226}Ra from food and drinking water by inhabitants of the Wałbrzych region*, J. Radioanal. Nucl. Ch., 222 (1–2), 189–193.

Ray S. B., Mohanti M., Somayajulu B. L. K., 1995, *Uranium isotopes in the Mahandai River – estuarine system, India*, Estuar. Coast. Shelf Sci., 40 (6), 635–645.

Rodriguez-Alvarez M. J., Sanchez F., 1995, *Behavior of uranium along the Jucar river (eastern Spain). Determination of $^{234}U/^{238}U$ and $^{235}/^{238}U$ ratios*, J. Radioanal. Nucl. Chem., 190, 113–120.

Ruszkiewicz K., Świątkowski A., 2008, *Zagrożenie radioaktywne polskich rzek oraz Morza Bałtyckiego. Ochrona wód przed zanieczyszczeniem*, Gosp. Wod., 1, 17–22.

Sam A. K., Ahmed M. M. O., El Khangi F. A., El Nigumi Y. O., Holm E., 1999, *Radiological and chemical assessment of Uro and Kurun rock phosphates*, J. Environ. Radioactiv., 42 (1), 65–75.

Skwarzec B., 1995, *Polon uran i pluton w ekosystemie południowego Bałtyku*, Rozpr. Monogr. 6, IO PAN, Sopot, 183 pp.

Skwarzec B., 1997, *Radiochemical methods for the determination of polonium, radiolead, uranium and plutonium in the environmental samples*, Chem. Anal. Warsaw, 42 (1), 107–115.

Skwarzec B., 2009, *Determination of radionuclides in aquatic environment*, [in:] *Analytical measurement in the aquatic environments*, J. Namieśnik & P. Szefer (eds.), CRC Press, Boca Raton–London–New York, 241–259.

Skwarzec B., Boryło A., Strumińska D. I., 2002, *^{234}U and ^{238}U isotopes in water and sediments of the southern Baltic*, J. Environ. Radioactiv., 61 (3), 345–363.

Skwarzec B., Boryło A., Strumińska D. I., 2004, *Activity disequilibrium between ^{234}U and ^{238}U isotopes in southern Baltic*, J. Water Air Soil Poll., 159 (1), 165–173.

Skwarzec B., Jahnz-Bielawska A., Boryło A., 2010, *The inflow of uranium ^{234U} and ^{238}U from Vistula River catchment area to the Baltic Sea*, Radiochim. Ac., (in press).

Skwarzec B., Strumińska D. I., Boryło A., 2001, *The radionuclides ^{234}U, ^{238}U and ^{210}Po in drinking water in Gdańsk agglomeration (Poland)*, J. Radioanal. Nucl. Ch., 250 (2), 315–318.

Skwarzec B., Strumińska D. I., Boryło A., 2003, *Radionuclides of ^{210}Po, ^{234}U and ^{238}U in drinking bottled mineral mater in Poland*, J. Radioanal. Nucl. Ch., 256 (2), 361–363.

Skwarzec B., Strumińska D. I., Boryło A., 2006, *Radionuclides of iron (^{55}Fe), nickel (^{63}Ni), polonium (^{210}Po), uranium (^{234}U, ^{235}U, ^{238}U) and plutonium (^{238}Pu, $^{239+240}Pu$, ^{241}Pu) in Poland and Baltic Sea environment*, Nukleonika, 51 (Supp. 2), 45–51.

Sullivan M. F., 1980, *Absorption of actinide elements from the gastro-intestinal tract of neonatal animals*, Health Phys., 38 (2), 173–185.

Sullivan M. F., Gorham L. S., 1982, *Further studies on the absorption of actinide elements from the gastro-intestinal tract of neonatal animals*, Health Phys., 43 (4), 509–519.

Tomza I., Lebecka J., 1981, *Radium-bearing waters in coal mines: occurrence. methods of measurements and radiation hazard*, Proc. Int. Conf. on Radiation Hazards in Mining, Golden, CO, Kingsport Press.

Tuszkowska A., 2009, *Spływ radionuklidów polonu, uranu i plutonu z dorzecza Odry*, Ph. D. thesis, Fac. Chem., Univ. Gdańsk.

USEPA – U. S. Environmental Protection Agency, 2001, Office of Water, Current Drinking Water Standards, IOWS.

Vargas M. J., Tome F. V., Schanchez A. M., 1995, *Behavior of uranium, thorium and ^{226}Ra in surface waters from a river passing through a granite region in the southwest of Spain*, Nucl. Geophys., 9, 567–578.

WHO – World Health Organisation, 1998, *Guidelines for drinking-water quality*, Geneva.