J Radioanal Nucl Chem (2010) 286:373–380
DOI 10.1007/s10967-010-0716-0

Polonium (^{210}Po), uranium (^{238}U) and plutonium ($^{239+240}$Pu) in the biggest Polish rivers

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Received: 13 July 2010 / Published online: 19 August 2010

Abstract In the paper the results of determination of ^{210}Po, ^{238}U and $^{239+240}$Pu radionuclides in the biggest Polish rivers are presented. Our results show the Vistula and the Oder as well as three Pomeranian Rivers are important sources of these radionuclides for the southern Baltic Sea.

Keywords ^{210}Po · ^{238}U · $^{239+240}$Pu · Vistula · Oder · Pomeranian Rivers · Inflow · Poland · Baltic Sea

Introduction

Alpha emitters play a significant role in radiological effects connected with their accumulation in organisms [1]. ^{210}Po is the most radiotoxic nuclide to man [2]. Natural concentration of polonium in the environment can be enhanced due to human activity (industry, fossil fuel combustion, phosphate fertilizers) [3]. ^{210}Po concentration in the atmosphere ranges from 13 to 240 μBq m^{-3} and its deposition varies between 10 and 500 Bq m^{-2} year^{-1} and depends on many processes such as rock erosion and coal combustion [4–6]. The main reservoir of polonium retention is the aquatic environment, e.g. the residence time of dissolved ^{210}Po in an estuary was estimated as 18–30 days [4].

In rocks, uranium isotopes are generally in radioactive equilibrium, but in natural waters the activity of ^{234}U is a little higher than that of ^{238}U (^{234}U/^{238}U activity ratio is 1.2–1.7) [7]. Uranium from rocks, soils and sediments is dissolved in water and transported to rivers [1, 8]. In seawater and river water, natural uranium exists predominantly in the dissolved form of uranyl carbonate anions [9].

Plutonium isotopes belong to the group of anthropogenic radionuclides. They are important from the radiological point of view due to their high radiotoxicity, long physical half-life and long residence times in biological system [10]. The principal source of plutonium in the environment is atmospheric fallout from nuclear weapon tests [1, 11]. Other plutonium sources such as releases from spent fuel reprocessing facilities are less important. Since 26 April 1986 also plutonium from the Chernobyl accident has to be taken into account [12].

The Vistula and the Oder Rivers

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 year^{-1}, and 50% of this amount is discharged by the Vistula and 34% with the Oder River [13, 14]. The Vistula and the Oder catchments cover more than 80% of Poland and transport about 2.1 × 10^6 m^3 of water per year with various fluvial material (i.e. 1.8 × 10^6 ton of salt from coal mines of the Upper Silesian Coal Factory) [15]. Also phosphate fertilizers have large influence on polonium and uranium concentrations in rivers [16]. Cultivated soils in Poland cover about 16.6 × 10^6 ha where about 18.7 kg ha^{-1} of phosphate fertilizers are used [14, 17].

The aim of this work was the determination of ^{210}Po, ^{238}U and $^{239+240}$Pu in the biggest Polish rivers and the calculation of total runoff of Po, U and Pu to the southern Baltic Sea.

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Materials and methods

Unfiltered river water grab samples of 60–200 dm^3 volume (3 per sampling point) were taken every quarter from Polish rivers (the Vistula, the Oder and Pomeranian—the Rega, the Pasłeka, the Słupia) between 2002 and 2004. The sampling sites are shown in Fig. 1. In laboratory, the samples were spiked immediately after their delivery with about 50 mBq of ^{209}Po, 100 mBq of ^{232}U and 5 mBq of ^{242}Pu as yield tracers. All the nuclides were coprecipitated with MnO_2. Po was electrodeposited on silver discs; U and Pu, after separation on Dowex anion-exchange resins, were electrolyzed on stainless steel discs [18, 19]. The activities of ^{210}Po, ^{238}U and $^{239+240}$Pu were measured using alpha spectrometry with semiconductor silicon detectors (Canberra-Packard, USA).

The minimum detectable activity (MDA) was 0.3 mBq of polonium and uranium for 2 days counting time and 0.1 mBq of plutonium for 10 days. The accuracy and precision of the radiochemical methods were estimated as better than 7% by participation in intercomparison exercises and analysis of IAEA reference materials. The polonium and uranium yield in analyzed samples ranged between 70–96%, for plutonium it was 40–75% [18, 19]. The results of ^{210}Po, ^{238}U and $^{239+240}$Pu activities are given with standard deviation (SD) calculated for 95% confidence intervals (2σ).

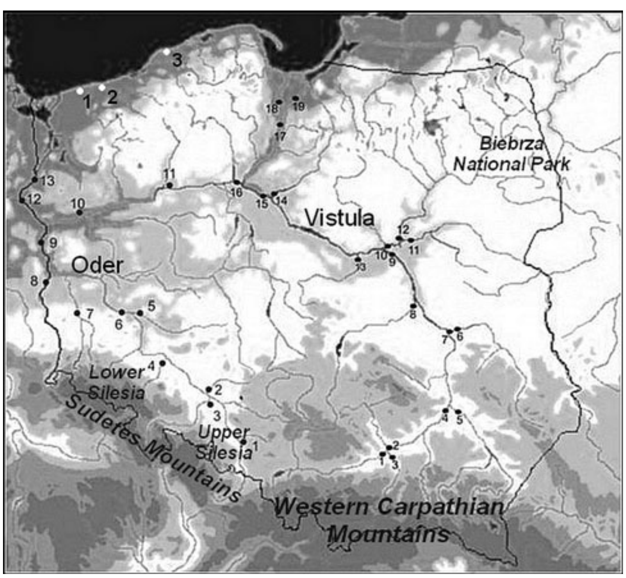

Fig. 1 Sampling sites. *The Vistula 1* Kraków, *2* Nida, *3* Dunajec, *4* Sandomierz, *5* San, *6* Wieprz, *7* Dęblin, *8* Pilica, *9* Warszawa, *10* Bug and Narew, *11* Bug, *12* Narew, *13* Bzura, *14* Drwęca, *15* Toruń, *16* Brda, *17* Grudziądz, *18* Leniwka, *19* Nogat. *The Oder 1* Chałupki, *2* Mała Panew, *3* Nysa Kłodzka, *4* Bystrzyca, *5* Barycz, *6* Głogów, *7* Bóbr, *8* Nysa Łużycka, *9* Słubice, *10* Warta, *11* Noteć, *12* Gozdowice, *13* Widuchowa. *The Pomeranian Rivers 1* Rega, *2* Parsęta, *3* Słupia

Results and discussion

The Vistula River

The activities of ^{210}Po, ^{238}U and $^{239+240}$Pu in water samples from the Vistula River and their tributaries in different seasons are presented in Table 1, where variations in the activities with season and sampling site can be seen. Generally, Po activities are higher in spring [20]. In samples from the Vistula catchment radioactive disequilibrium between uranium isotopes was observed, the values of ^{234}U/^{238}U activity ratio ranged from 1.00 (the Bug & the Narew) to 1.74 (the Vistula) [21].

Using seasonal and annual flow of the Vistula and its tributaries, the seasonal and annual runoff of polonium, uranium and plutonium from the Vistula drainage were calculated and are presented in Table 2. Annual flow of ^{210}Po in both Vistula's delta branches (the Leniwka and the Nogat) was 73.71 GBq year^{-1} (Table 2). It confirms that the Vistula drainage is an important source of ^{210}Po in the Baltic Sea [20].

The results of seasonal uranium and plutonium inflow with the Vistula (Kiezmark) were the highest in spring (Table 2). Annually, the south-eastern Baltic Sea is enriched with about 507 GBq uranium isotopes when, in comparison, the Mahanadi River transports 885.6 GBq year^{-1} of total uranium to the Bengal Bay (India) [6, 22]. The uranium inflow is mostly caused by human activity, mainly in agriculture and coal mining industry—a source of huge amount of waste containing large quantities of natural radionuclides [15]. Annually mine waters from the Silesian Coal Factory enrich the river with 75 GBq of ^{226}Ra and 145 GBq of ^{228}Ra [23–26]. Phosphate rocks contain usually uranium and radium (^{226}Ra) [27, 28]. During phosphate fertilizers production about 10% of the initial amount of ^{226}Ra, 20% of uranium and about 85% of ^{210}Po is found in the waste—phosphogypsum which is an important problem [16, 29, 30]. The vicinity of phosphogypsum waste located in Wiślinka (at delta of the Vistula) is strongly polluted and rich in polonium and uranium [31].

The Oder River

The activities of ^{210}Po, ^{238}U and $^{239+240}$Pu in samples from the Oder River and its tributaries are presented in Table 3, where similar features can be found as in Table 1 for the Vistula. The highest activities of ^{210}Po were again observed in spring season [32]. In the Oder water radioactive disequilibrium between ^{234}U and ^{238}U ranged from 1.03 (the Noteć) to 1.84 (the Oder). The highest activities of $^{239+240}$Pu were determined in autumn.

The seasonal and annual runoff of polonium, uranium and plutonium are shown in Table 4. The highest values of ^{210}Po flow in the Oder were found in winter and spring and

Table 1 Average activity concentrations of ^{210}Po, ^{238}U and $^{239+240}Pu$ in the Vistula River waters and their tributaries (±2σ uncertainties)

Sampling site	^{210}Po ± SD (Bq m^{-3})				^{238}U ± SD (Bq m^{-3})				$^{239+240}Pu$ ± SD (mBq m^{-3})			
	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn
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
Dęblin	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
Toruń	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
Grudziądz	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	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	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	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 and Narew	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
Drwęca	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

Table 2 Seasonal, annual runoff of ^{210}Po, ^{238}U and $^{239+240}Pu$ from the Vistula River drainage and annual surface runoff from the Vistula tributaries

Sampling site	^{210}Po (GBq quarter^{-1})				^{238}U (GBq quarter^{-1})				$^{239+240}Pu$ (MBq quarter^{-1})				All year			Annual surface runoff		
	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	^{210}Po (GBq year^{-1})	^{238}U (GBq year^{-1})	$^{239+240}Pu$ (MBq year^{-1})	^{210}Po (kBq year^{-1} km^{-2})	^{238}U (kBq year^{-1} km^{-2})	$^{239+240}Pu$ (Bq year^{-1} km^{-2})
Vistula River																		
Kraków	0.66	1.94	0.22	0.78	3.7	2.6	0.5	0.2	1.60	1.72	0.16	0.74	3.60	7.0	4.22	–	–	–
Sandomierz	2.18	10.38	1.66	2.23	3.1	21.4	4.1	9.7	9.58	21.60	1.89	1.47	16.45	45.3	34.54	–	–	–
Dęblin	4.48	37.46	4.63	5.94	5.2	32.9	10.8	16.5	10.0	54.7	6.30	6.27	52.51	65.4	77.27	–	–	–
Warszawa	4.02	23.08	4.32	10.53	19.9	44.5	9.3	23.1	6.59	11.80	5.00	20.70	41.95	96.8	44.09	–	–	–
Toruń	15.08	30.95	7.66	4.61	40.7	61.8	24.4	15.9	24.30	24.4	12.60	9.66	58.30	142.8	70.96	–	–	–
Grudziądz	16.96	34.26	6.82	3.88	76.4	69.7	11.1	25.4	20.30	32.3	2.91	5.88	61.92	182.6	61.39	–	–	–
Malbork	0.36	0.54	0.54	0.70	2.1	1.8	1.3	2.6	0.34	0.89	0.47	1.20	2.14	7.8	2.90	–	–	–
Kiezmark	19.67	40.82	4.31	6.77	78.8	93.3	17.3	33.4	15.70	50.80	3.38	16.10	71.57	222.8	86.00	–	–	–
Vistula tributaries																		
Nida	0.08	0.87	0.37	0.11	0.5	1.7	0.7	0.5	0.43	1.33	0.19	0.35	1.43	3.4	2.30	380	880	595
Dunajec	1.43	9.29	0.41	0.57	4.3	8.4	1.2	2.0	1.86	9.44	0.59	1.68	11.70	15.9	13.57	1,720	2,330	1,994
San	0.98	7.09	0.46	2.16	6.8	14.7	1.5	3.7	15.31	23.5	1.56	2.33	10.69	26.7	42.70	640	1,590	2,534
Wieprz	1.94	3.08	0.07	0.80	1.3	2.2	1.0	2.0	3.02	2.64	0.17	1.42	5.89	6.5	7.25	580	630	695
Pilica	0.88	1.88	0.67	2.05	0.9	2.7	1.0	3.9	5.50	3.23	1.09	2.86	5.48	8.5	12.68	630	910	1,367
Narew	1.51	1.70	0.64	7.47	5.9	16.1	1.2	8.9	3.01	8.33	0.96	13.00	11.32	32.1	25.30	150	430	337
Bug	1.05	7.81	1.32	3.53	10.8	36.5	2.2	1.2	3.16	8.24	1.64	6.30	13.71	50.7	19.34	350	760	500
Bug and Narew	7.59	16.12	1.19	9.63	15.8	53.8	3.4	8.2	7.69	10.90	1.53	5.14	34.53	81.2	25.26	300	710	220
Drwęca	0.46	0.37	0.10	1.04	0.3	2.6	0.7	2.6	0.87	1.53	0.31	6.81	1.97	5.9	9.52	370	1,140	1,780
Brda	0.55	0.38	0.29	0.62	0.3	0.8	0.4	1.6	1.48	2.61	0.75	1.64	1.84	3.1	6.48	390	670	1,528
Bzura	0.89	1.28	0.25	0.84	2.0	2.9	0.3	1.0	0.73	1.16	0.29	0.81	3.26	6.2	2.99	410	800	384

Table 3 Average activity concentrations of ^{210}Po, ^{238}U and $^{239+240}Pu$ in the Oder River waters and their tributaries (±2σ uncertainties)

Sampling site	^{210}Po ± SD (Bq m^{-3})				^{238}U ± SD (Bq m^{-3})				$^{239+240}Pu$ ± SD (mBq m^{-3})			
	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn
Oder River												
Chałupki	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
Głogów	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
Słubice	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
Oder tributaries												
Mała 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 Kłodzka	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 Łużycka	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
Noteć	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 4 Seasonal, annual runoff of ^{210}Po, ^{238}U and $^{239+240}Pu$ from the Oder River drainage and annual surface runoff from the Oder tributaries

Sampling site	^{210}Po (GBq quarter^{-1})				^{238}U (GBq quarter^{-1})				$^{239+240}Pu$ (MBq quarter^{-1})				All year			Annual surface runoff		
	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	^{210}Po (GBq year^{-1})	^{238}U (GBq year^{-1})	$^{239+240}Pu$ (MBq year^{-1})	^{210}Po (kBq year^{-1} km^{-2})	^{238}U (kBq year^{-1} km^{-2})	$^{239+240}Pu$ (Bq year^{-1} km^{-2})
Oder River																		
Chałupki	0.54	0.24	0.20	0.21	3.23	0.41	0.54	0.82	14.00	0.38	0.35	0.69	1.19	5.00	15.42	–	–	–
Głogów	1.15	0.67	0.92	1.01	23.45	11.75	7.16	9.52	9.27	4.96	0.86	2.24	3.75	51.88	17.33	–	–	–
Słubice	3.36	3.08	1.06	1.41	38.82	18.57	11.07	9.67	12.24	8.12	3.58	2.70	8.91	78.13	26.64	–	–	–
Gozdowice	4.52	4.84	2.17	3.23	45.71	27.01	14.09	13.27	21.11	10.45	6.89	7.41	14.76	100.08	45.86	–	–	–
Widuchowa	2.23	4.82	3.57	2.49	32.76	47.33	13.04	13.87	7.08	6.87	3.74	5.33	13.11	107.00	23.02	–	–	–
Oder tributaries																		
Mała Panew	0.03	0.04	0.03	0.05	0.03	0.14	0.05	0.05	0.02	0.36	0.06	0.04	0.15	0.27	0.48	71	126	224
Nysa Kłodzka	0.28	0.36	0.18	0.23	1.68	2.71	1.13	1.16	1.34	1.19	0.34	0.28	1.05	6.68	3.15	229	1,458	691
Bystrzyca	0.13	0.04	0.03	0.02	3.08	1.08	4.33	0.78	0.30	0.11	0.04	0.02	0.22	9.27	0.47	125	5,244	262
Barycz	0.014	0.012	0.007	0.012	1.22	0.53	0.25	0.05	0.18	0.04	0.13	0.02	0.045	2.05	0.37	8	370	67
Bóbr	0.29	0.23	0.12	0.22	2.53	1.92	0.42	0.39	4.13	1.57	0.19	1.33	0.86	5.26	7.22	145	894	1,230
Nysa Łużycka	0.34	0.20	0.14	0.38	0.68	0.15	0.37	0.27	1.00	0.46	0.16	0.35	1.06	1.47	1.97	245	342	455
Warta	1.41	2.51	1.36	0.96	10.45	5.92	4.63	6.00	4.16	3.73	3.07	1.15	6.24	27.00	12.11	115	496	221
Noteć	0.37	0.78	0.27	0.79	4.75	1.39	1.99	2.93	1.44	1.92	1.17	1.39	2.21	11.06	5.92	129	638	342

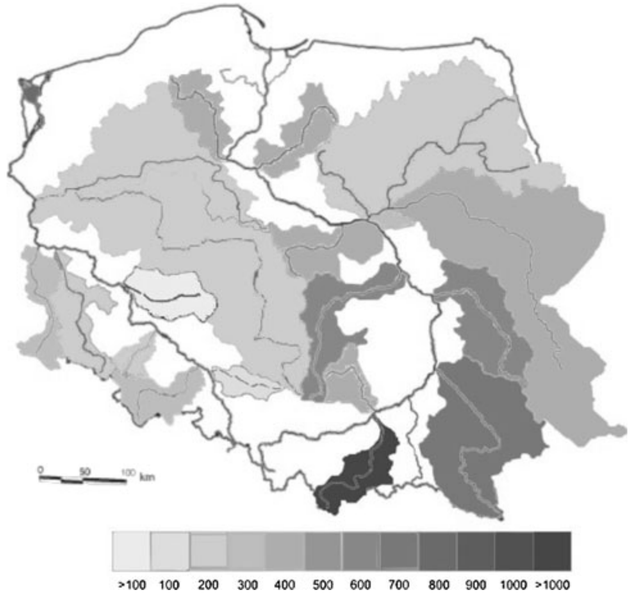

Fig. 2 Annual surface runoff of ^{210}Po from the Vistula and the Oder tributaries drainages (kBq km^{-2} year^{-1})

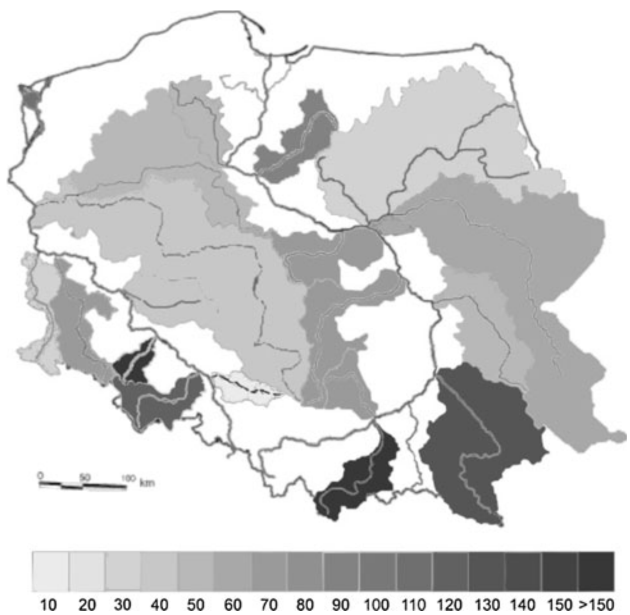

Fig. 3 Annual surface runoff of uranium from the Vistula and the Oder tributaries drainages (g km^{-2} year^{-1})

the Oder is an important source of ^{210}Po for the Baltic Sea [6]. The annual surface runoff of polonium, uranium and plutonium per square kilometer of Oder tributaries catchments is presented in Table 4 as well as in Figs. 2, 3 and 4.

The Pomeranian Rivers

Three Pomeranian Rivers (the Słupia, the Parsęta, the Rega) were examined in spring 2004 to determine the

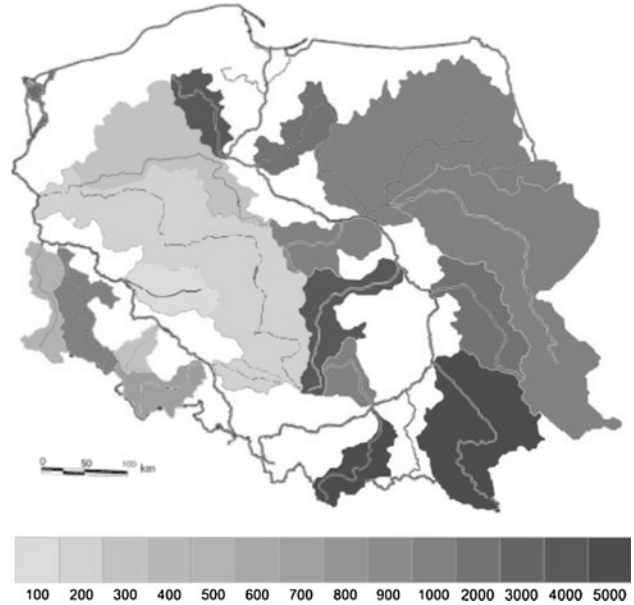

Fig. 4 Annual surface runoff of $^{239+240}$Pu from the Vistula and the Oder tributaries drainages (Bq km^{-2} year^{-1})

concentrations of ^{210}Po, ^{238}U and $^{239+240}$Pu and the results are presented in Table 5. On the basis of activities the seasonal flow was calculated and the data are also given in Table 5.

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

On the basis of annual flow of analyzed radionuclides from the rivers studied, the annual inflow of ^{210}Po, ^{238}U and $^{239+240}$Pu to the southern Baltic Sea was calculated and the results are presented in Table 6 [20, 21, 32, 33].

Annually, the south-eastern Baltic is enriched with 73.7 GBq of ^{210}Po, about 507 GBq of ^{234}U and ^{238}U and 88.9 MBq of $^{239+240}$Pu flow with the Vistula River. From this amount 86.0 MBq $^{239+240}$Pu (96.6%) go with the Leniwka to the Gdańsk Bay. The inflow of analyzed radionuclides from the Oder River is smaller in comparison with the Vistula River. The annual flows of Po, U and Pu with the Oder River to the Szczecin Lagoon (southern Pomeranian Bay) were calculated as 14.8 GBq of ^{210}Po, 100.8 GBq of ^{238}U and 45.9 MBq of $^{239+240}$Pu. The inflow of analyzed radionuclides with the Pomeranian Rivers is less important in comparison with the Vistula or the Oder River. Total annual runoff of polonium, uranium, plutonium with the Vistula, the Oder and the 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 indicate, that the Vistula, the Oder and to a

Table 5 Activity concentration and the surface runoff of ^{210}Po, ^{238}U and $^{239+240}$Pu in the Pomeranian Rivers in spring

Sampling site	Average activity concentrations			Surface runoff		
	^{210}Po (Bq m^{-3})	^{238}U (Bq m^{-3})	$^{239+240}$Pu (mBq m^{-3})	^{210}Po (GBq quarter^{-1})	^{238}U (GBq quarter^{-1})	$^{239+240}$Pu (MBq quarter^{-1})
Rega	3.82 ± 0.24	8.97 ± 0.52	3.38 ± 0.98	0.18	0.41	0.16
Parsęta	5.50 ± 0.33	6.19 ± 0.30	11.58 ± 3.49	0.87	0.96	1.80
Słupia	4.84 ± 0.21	5.10 ± 0.29	3.90 ± 1.08	0.45	0.47	0.36

Table 6 Annual runoff of ^{210}Po, ^{238}U and $^{239+240}$Pu from the Vistula, the Oder and the Pomeranian Rivers to the Baltic Sea

Radionuclide/quantity	Vistula River	Oder River	Pomeranian Rivers	Total
^{210}Po (GBq year^{-1})	73.7	14.8	6.0	94.5
^{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
$^{239+240}$Pu (MBq year^{-1})	88.9	45.9	9.3	144.1

lesser extent the Pomeranian Rivers are important sources of polonium, uranium and plutonium radionuclides in the southern Baltic Sea environment.

Conclusions

On the basis of the study we can conclude that the annual surface runoff of ^{210}Po, ^{238}U and $^{239+240}$Pu from the Vistula drainage was higher in mountain catchments.

The results of our investigation indicated the Vistula, the Oder and to a lesser extent the Pomeranian Rivers as important sources of polonium, uranium and plutonium radionuclides in the southern Baltic Sea 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.

Acknowledgments The authors would like to thank the Ministry of Science and Higher Education of the financial support of this work under grant DS/8460-4-0176-0.

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