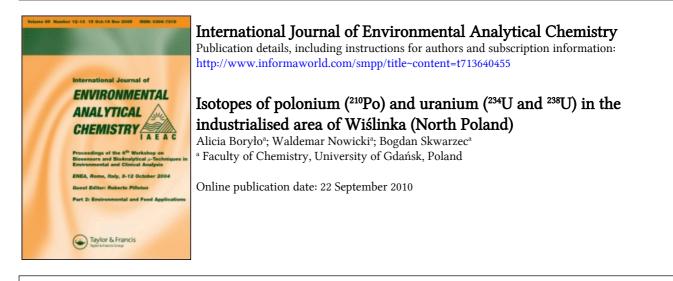
This article was downloaded by: On: *17 January 2011* Access details: *Access Details: Free Access* Publisher *Taylor & Francis* Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



To cite this Article Boryło, Alicia , Nowicki, Waldemar and Skwarzec, Bogdan (2009) 'Isotopes of polonium (²¹⁰Po) and uranium (²³⁴U and ²³⁸U) in the industrialised area of Wiślinka (North Poland)', International Journal of Environmental Analytical Chemistry, 89: 8, 677 – 685

To link to this Article: DOI: 10.1080/03067310802654605 URL: http://dx.doi.org/10.1080/03067310802654605

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: http://www.informaworld.com/terms-and-conditions-of-access.pdf

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan or sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.



Isotopes of polonium (²¹⁰Po) and uranium (²³⁴U and ²³⁸U) in the industrialised area of Wiślinka (North Poland)

Alicia Boryło*, Waldemar Nowicki and Bogdan Skwarzec

Faculty of Chemistry, University of Gdańsk, ul. Sobieskiego 18/19, 80-952 Gdańsk, Poland

(Received 30 July 2008; final version received 25 November 2008)

Phosphogypsum is very much enriched in natural polonium 210 Po, while the uranium radionuclides (234 U and 238 U) tend to be in waters around waste dumps. The migration and distribution of uranium and polonium radionuclides in analysed water samples are not uniform. The amounts of uranium isotopes in a retention reservoir around the waste dump are 100 times higher than in typical natural river waters. The concentration of uranium in surface water samples collected near Wiślinka (North Poland) ranged widely between 0.05 ± 0.02 and $430 \pm 5 \,\mu\text{g L}^{-1}$. In phosphogypsum samples, uranium concentrations varied in the range of 4.03 ± 0.08 and $0.65 \pm 0.05 \text{ mg kg}^{-1}$ in samples collected in 1997 and 2007, respectively. We observed that the activity ratio $^{234}\text{U}/^{238}\text{U}$ is approximately about one in the phosphogypsum (0.90 \pm 0.06 and 0.97 ± 0.03) and in the water of the immediate surroundings of waste heap $(1.00 \pm 0.06 \text{ and } 1.07 \pm 0.05)$, while in surface river water from Martwa Wisła River is higher than one (1.10 ± 0.07) and $1.16 \pm 0.13)$. The phosphogypsum samples contain much higher ²¹⁰Po activity in comparison with uranium radionuclides. The concentration of ²¹⁰Po in phosphogypsum samples is similar or slightly higher in 1997 and 2007 (600 ± 10 and 700 ± 10 Bq kg⁻¹, respectively). Finally, the principal sources of ²¹⁰Po, ²³⁴U, ²³⁸U radionuclides in the Wiślinka area waste dump are phosphorites and phosphogypsum, produced by Phosphoric Fertilisers Industry of Gdańsk.

Keywords: polonium ²¹⁰Po; uranium ²³⁴U and ²³⁸U; concentration; phosphogypsum; surface water; industrialised areas; Wiślinka (Poland)

1. Introduction

One of the significant components of the Vistula River delta in the northern part of Poland is phosphogypsum waste dump near Wiślinka village. The phosphogypsum waste heap in Wiślinka was started 30 years ago and now this waste heap is 40 m high, more than 30 million tons in weight and covers over 85 hectares of land. The phosphogypsum is a waste product in the process of phosphoric acid production. The phosphorites – the main component for production – are transported to Gdańsk by Phosphoric Fertilisers Industry from North Africa (Morocco) [1]. Sulphuric acid – next component – is used for phosphorites degradation and production of superphosphate (hydrated calcium sulphate) enrichment. Phosphogypsum is transported by cargo boats from the Phosphoric Fertilisers Industry of Gdańsk to the Wiślinka waste dump. The plant authorities declare that

^{*}Corresponding author. Email: aborylo@chem.univ.gda.pl

phosphogypsum contains only very little amounts of trace metals and radioactive elements so it is not an environmentally hazardous substance. However, some of the components (trace metals and radionuclides) are classified as substances with mutagenic and carcinogenic action [2]. According to the Voivodship Inspectorate for Environmental Protection in Gdańsk, the phosphogypsum waste dump near Wiślinka is poisoning the Martwa Wisła River water [3]. Rainwater polluted from phosphogypsum waste dump should flow into the reservoir, but in practice water is probably leaking out into Vistula River.

Uranium is spread in nature, it occurs in over 160 minerals, locally at high concentrations (between 50 and 90%) [4,5]. Higher uranium concentrations are observed, for example, around phosphogypsum stacks [6]. The principal source of uranium in the natural environment is the atmospheric precipitation of terrigenic material, as well as river waters and fertilisers [7]. Natural uranium contains three alpha radioactive isotopes: 99.2745% of ²³⁸U, 0.7200% of ²³⁵U and 0.0054% of ²³⁴U [8,9]. One Bq ²³⁸U corresponds to 81.6 µg of natural uranium (1 µg natural U – 12.3 mBq of ²³⁸U) [10]. The ²³⁴U isotope belongs to the natural radioactive decay series of ²³⁸U [11]. The specific activity of natural uranium (²³⁴U, ²³⁵U and ²³⁸U) is about 25446 Bq g⁻¹ [9,5], but specific activity of ²³⁸U is lower (1.24 × 10⁴ Bq g⁻¹) in comparison with ²³⁴U (2.30 × 10⁸ Bq g⁻¹) [12]. The concentration of uranium in the natural environment is increasing as a result of human activity – industry, fossil fuel combustion, phosphate fertilisers in agriculture, domestic and industrial sewage [13].

The ²³⁴U and ²³⁸U radionuclides are not in the radioactive state equilibrium in the natural environment, higher disequilibrium has been noticed in the seawater rather than in the sediments [13,14]. In the southern Baltic (Gulf of Gdańsk) waters, the mean value of ²³⁴U²³⁸U activity ratio was found to be 1.17, while in the sediments is about 1.00 [13,15]. In the aquatic environment uranium exists on two oxidation degrees: U(IV) and U(VI). Chemical compounds of uranium in reduced form are sparingly soluble [13]. In oxidised form in sea and river water there are soluble carbonate complexes: $[UO_2(CO_3)_2]^{2-}$ [13].

Because uranium is a radioactive and toxic metal, it is a hazardous environmental pollutant. It is relatively highly toxic to humans, both chemically and radiologically (its progeny are highly radioactive). The compounds of uranium can penetrate the blood from alveoli pockets in the lungs or through the gastrointestinal (GI) tract as a result of consumption of contaminated water or food. The residence time of the insoluble uranium in the GI tract is estimated in years. Due to its heavy metal nature, the presence of uranium in the body causes damage to the kidney leading to nephritis [16]. The limit of permissible uranium in drinking water is $30 \,\mu g \, L^{-1}$ according to United States Environmental Protection Agency [17] and $15 \,\mu g \, L^{-1}$ according to the World Health Organization (WHO) [18].

Polonium ²¹⁰Po belongs to a natural uranium decay series starting from ²³⁸U but its fate depends on further members of this series, e.g. ²²⁶Ra and most of all on ²¹⁰Pb. ²²²Rn escaping from the Earth's surface constitutes the source of atmospheric ²¹⁰Po [19]. The main source of ²¹⁰Po in environment is ²¹⁰Pb and ²¹⁰Po falling to the ground with atmosphere. A small amount of ²¹⁰Po is formed *in situ* as a result of the radioactive decay of uranium contained in seawater. Apart from ²¹⁰Po formed by the decay of ²¹⁰Pb contained in the air, additional quantities are emitted directly from the Earth as a result of forest fires and volcanic eruptions [19]. Specific activity of polonium ²¹⁰Po is very high and equals 1.66×10^{14} Bq g⁻¹, and the half-life is 138.4 days [12]. Polonium, ²¹⁰Po, belongs to the most radiotoxic nuclide to human beings [7]. The limit of permissible polonium in drinking water is 0.1 Bq L^{-1} according to the WHO [18].

The objective of this study was the determination of uranium concentration in surface water samples and in phosphogypsum from the area around the phosphogypsum waste dump and from Martwa Wisła River in the northern part of Poland. Down the main current of Martwa Wisła River, water samples were collected in Przegalina, Sobieszewko and Górki Wschodnie. The analysed water samples were collected in 2007 and 2008, but the phosphogypsum samples in 1997 and 2007.

2. Experimental

The surface water samples (10 L) were collected near Wiślinka in November 2007 from the area around the phosphogypsum waste dump and from Martwa Wisła River to Wiślinka area in April 2008. Down the main current of the Martwa Wisła River, samples were collected in Przegalina (A), Sobieszewko (B) and Górki Wschodnie (C). The places of the surface water sampling are presented in Figures 1 and 2. Phosphogypsum samples were collected in March 1997 and November 2007.

The method of determination of polonium ²¹⁰Po and uranium (²³⁴U, ²³⁸U) in analysed samples was based on the method established by Skwarzec [20,21]. Before radiochemical analysis, about 100 mBq of ²⁰⁹Po and 100 mBq ²³²U was added to each sample as a yield tracer. Polonium was co-precipitated with manganese dioxide and after mineralisation, electro-deposited on silver disc [20,21]. After polonium electro-deposition, samples of surface water and phosphogypsum were mineralised using concentrated acids HNO₃ and HCl. When mineralised samples were clear, they were evaporated and dry residue was dissolved in 60 mL of 8 M HNO₃. The solution was introduced into a column filled with anion exchange resins AG 1–X 8 (50–100 mesh). The column was then washed with 90 mL

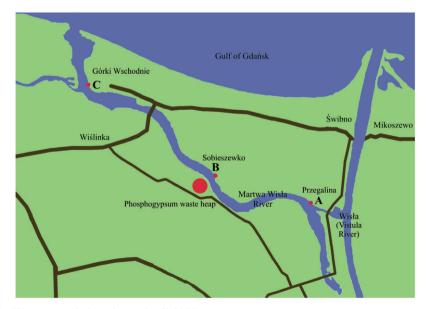


Figure 1. Water sample locations (April 2008).

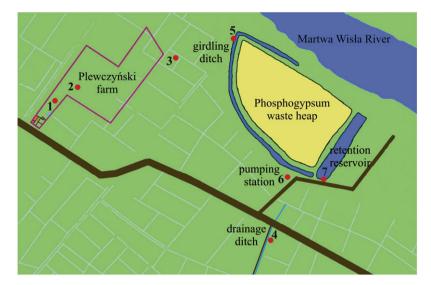


Figure 2. Water sample locations (November 2007).

of 8 M HNO₃, eluted solution contained uranium, iron and alkaline earth elements. Next, the uranium fraction was dissolved in 10 mL of 9 M HCl and introduced into a column filled with anion exchange resins AG 1–X 8 (100–200 mesh). The column was washed with 60 mL of 9 M HCl (in order to remove Cs, Sr, Ra, Ni), and next, U, Fe, Co and Cu retained by the resin were washed with 60 mL of 0.5 M HCl. The solution eluted was evaporated and dissolved in 10 mL of 1 M (NH₄)SO₄ (pH = 1.5). In order to separate uranium from Fe, Co and Cu, the solution was introduced into a column filled with anion exchange resins AG 1–X 8 (100–200 mesh), and washed with 60 mL of 0.5 M HCl. The solution (NH₄)SO₄ (pH = 1.5); uranium retained by resins was eluted with 50 mL of 0.5 M HCl. The solution containing uranium was evaporated and dry residue was mineralised with 2 mL of 1 : 1 mixture of concentrated HNO₃ and HCl. After evaporation, residue was dissolved in 5 mL of 0.75 M (NH₄)₂SO₄ (pH = 2) and transferred into electrolysis cell and the electrolysis was carried out on steel disc during 90 min at a constant current of 1.0 A.

Activity of ²¹⁰Po, ²³⁴U and ²³⁸U were measured using alpha spectrometry equipment with semiconductor silicon detectors and 300 mm² active surface barrier (Canberra-Packard, USA). Polonium samples were measured for two days and their activity were calculated on the electro-deposition day. Uranium samples were measured for 2–3 days. Uranium samples were measured for 3–5 days. The uranium yield in analysed samples ranged from 50 to 90%. The results of ²¹⁰Po, ²³⁴U and ²³⁸U concentration in analysed samples are given with standard deviation (SD) calculated for 95% confidence intervals. The trueness and precision of the radiochemical method were evaluated using International Atomic Energy Agency (IAEA) reference materials (IAEA-384, IAEA-385, IAEA-414) and estimated that the absolute relative bias is <10%.

3. Results and discussion

The results of ²³⁴U and ²³⁸U measurements in surface water samples are given in Tables 1 and 2. The highest concentrations of uranium isotopes and total uranium in samples

Sampling location	234 U (mBq dm $^{-3}$)	238 U (mBq dm $^{-3}$)	Total uranium (µg dm ⁻³)	Activit y ratio $^{234}U/^{238}U$
Wiślinka 1 Wiślinka 2 Wiślinka 3 Wiślinka 4 Wiślinka 5 Wiślinka 6	$\begin{array}{c} 0.9 \pm 0.05 \\ 1 \pm 0.05 \\ 19 \pm 0.5 \\ 1.5 \pm 0.05 \\ 33 \pm 1 \\ 55 \pm 1 \end{array}$	$\begin{array}{c} 0.6 \pm 0.05 \\ 0.75 \pm 0.05 \\ 16 \pm 0.5 \\ 1.3 \pm 0.05 \\ 31 \pm 1 \\ 51 \pm 1 \end{array}$	$\begin{array}{c} 0.05 \pm 0.02 \\ 0.06 \pm 0.03 \\ 1.3 \pm 0.05 \\ 0.1 \pm 0.05 \\ 2.6 \pm 0.1 \\ 4.5 \pm 0.1 \end{array}$	$\begin{array}{c} 1.5 \pm 0.1 \\ 1.30 \pm 0.1 \\ 1.20 \pm 0.05 \\ 1.15 \pm 0.05 \\ 1.1 \pm 0.05 \\ 1.1 \pm 0.05 \\ 1.1 \pm 0.05 \end{array}$

Table 1. The uranium concentration and values of the ${}^{234}\text{U}/{}^{238}\text{U}$ activity ratio in surface water samples near Wiślinka in November 2007 (area around the phosphogypsum waste dump).

Table 2. The uranium concentration and values of the ${}^{234}\text{U}/{}^{238}\text{U}$ activity ratio in surface water samples from Martwa Wisła river to Wiślinka area in April 2008.

Sampling location	234 U (mBq dm $^{-3}$)	238 U (mBq dm $^{-3}$)	Total uranium (µg dm ⁻³)	$\underset{^{234}\text{U}/^{238}\text{U}}{\text{Activity ratio}}$
Przegalina (A) Sobieszewko (B) Górki Wschodnie (C) Wiślinka 5 Wiślinka 7a Wiślinka 7b	$12 \pm 0.3 \\11 \pm 0.5 \\14 \pm 1 \\25 \pm 1 \\558,600 \pm 50 \\10,800 \pm 50$	$11 \pm 0.3 \\ 10.00 \pm 0.5 \\ 12 \pm 1 \\ 25 \pm 1 \\ 5250 \pm 50 \\ 11,050 \pm 50$	$\begin{array}{c} 0.9 \pm 0.02 \\ 0.8 \pm 0.04 \\ 1 \pm 0.1 \\ 2.1 \pm 0.1 \\ 430 \pm 5 \\ 900 \pm 4 \end{array}$	$\begin{array}{c} 1.15 \pm 0.05 \\ 1.10 \pm 0.1 \\ 1.2 \pm 0.1 \\ 1.00 \pm 0.1 \\ 1.1 \pm 0.05 \\ 1 \pm 0.05 \end{array}$

collected in November 2007 were observed in water from the Wiślinka area: in the pumping station (Wiślinka 6) and in the girdling ditch (Wiślinka 5) (Table 1). Also higher uranium concentration was found in the drainage ditch (Wiślinka 3). The relatively lower uranium concentration was in the open drainage ditch (Wiślinka 4) (Table 1). This open ditch located at the opposite side of road from Gdańsk to Mikoszewo is not connected with retention reservoir immediate surroundings of phosphogypsum waste dump. The lowest concentration of ²³⁴U, ²³⁸U and total uranium in water samples was collected relatively far away from the phosphogypsum waste dump: Wiślinka 1 and Wiślinka 2 (Table 1). The uranium isotopes ²³⁴U and ²³⁸U are not in the radioactive equilibrium state, but higher disequilibrium has been noticed in natural water [15]. The values of ²³⁴U/²³⁸U activity ratio in water samples from Wiślinka 1 and 2 (Table 1) are higher than average value in oceanic (1.14) and Baltic waters (1.17). These values are typical for surface land area waters (rivers and lakes). In the natural underground waters, the ²³⁴U/²³⁸U activity ratio lied in very wide range 0.51–9.02 [22].

The lower uranium concentrations in samples taken in April 2008 were noted in water from Martwa Wisła River samples: in Sobieszewko (station B), Przegalina (station A) and Górki Wschodnie (station C) (Table 2). The higher uranium concentration was confirmed in the girdling ditch (Wiślinka 5). The maximum uranium concentration in analysed water samples taken in April 2008, hundred times higher than was observed in the pumping station and girdling ditch, was found in retention reservoir. This fact can be explained by the lixiviation of uranium from phosphogypsum waste dump to water. Two water samples were collected in the retention reservoir: surface water (Wiślinka 7a) and bottom water (Wiślinka 7b) (Table 2). The concentration of uranium in the bottom water was much higher than in surface water: The value of $^{234}U/^{238}U$ activity ratio in these samples ranged between 0.98 in bottom water and 1.06 in surface water. The obtained result probably indicates a diffusion process of uranium from surface water to bottom water. This way of uranium elimination causes the increase of the 234 U/ 238 U activity ratio in surface water. The uranium ²³⁸U concentration in analysed water samples taken in retention reservoir (Wiślinka 7a and 7b) is clearly higher than uranium content in: Baltic water $(8.20 \text{ mBq dm}^{-3})$, drinking and bottled water from Gdańsk agglomeration $(0.80-2.80 \text{ mBq dm}^{-3})$, beer from Poland $(4.94 \text{ mBq dm}^{-3})$ and Vistula River water $(5.25-11.49 \text{ mBg dm}^{-3})$ [15].

The concentration of ²³⁴U and ²³⁸U in phosphogypsum samples are presented in Table 3. The higher values of uranium concentrations were measured in phosphogypsum samples collected in 1997 and the lower in samples taken in 2007 (Table 3). This difference is probably caused by change of phoshorite sources. The high amounts of radionuclides are in phosphorites from sea-bottom (phosphate) [1]. The values of 234 U/ 238 U activity ratio in analysed phosphogypsum samples lie between 0.90 and 0.97. Our results of ²³⁴U and ²³⁸U concentration in phosphogypsum samples are higher in comparison with values for Senegal phosphogyspum $(16\pm 2 \text{ and } 16\pm 2 \text{ Bg kg}^{-1})$ and lower in comparison with samples from Morocco $(225 \pm 13 \text{ and } 240 \pm 14 \text{ Bg kg}^{-1})$ [23].

The concentrations of polonium ²¹⁰Po in phosphogypsum samples are given in Table 3. The phosphogypsum samples are very much enriched in ²¹⁰Po in comparison to ²³⁴U and ²³⁸U activity. The similar activity of ²¹⁰Po were observed in phosphogypsum from Portugal (586 Bq kg⁻¹), Northern Florida (427-648 Bq kg⁻¹) [24] as well as Senegal (529 Bq kg^{-1}) and Morocco (779 Bq kg^{-1}) [23].

Polonium concentration, similar to uranium concentration, was higher in water samples from the Wiślinka area (Table 4). The maximum ²¹⁰Po concentration was found in the retention reservoir (Wiślinka 7a), the lowest was measured in the pumping station (Wiślinka 6) and girdling ditch around the phosphogypsum waste dump (Wiślinka 5).

Table 3. Average polonium 210 Po and uranium concentration and values of the 234 U/ 238 U activity	Į
ratio in phosphogypsum samples (five samples).	

Samples	210 Po (Bq kg ⁻¹)	^{234}U (Bq kg ⁻¹)	^{238}U (Bq kg ⁻¹)	Total uranium $(mg kg^{-1})$	Activity ratio ²³⁴ U/ ²³⁸ U
Phosphogypsum (1997) Phosphogypsum (2007)	$\begin{array}{c} 700\pm10\\ 600\pm10 \end{array}$	$\begin{array}{c} 50\pm1\\ 7.2\pm0.5\end{array}$	$\begin{array}{c} 48\pm1\\8\pm0.5\end{array}$	4 ± 0.1 0.65 ± 0.05	$\begin{array}{c} 0.97 \pm 0.03 \\ 0.90 \pm 0.06 \end{array}$

Table 4. Average polonium ²¹⁰Po concentration in surface water samples.

Samples location	210 Po (mBq dm $^{-3}$)	
Przegalina (A) Sobieszewko (B) Górki Wschodnie (C) Wiślinka 5 Wiślinka 6 Wiślinka 7a	$ \begin{array}{r} 6 \pm 0.5 \\ 1.2 \pm 0.3 \\ 53 \pm 1 \\ 5.50 \pm 0.5 \\ 53 \pm 3 \\ 200 \pm 10 \end{array} $	

The minimum activity of ²¹⁰Po was observed in water samples from Martwa Wisła (Table 4): Sobieszewko (station B), Górki Wschodnie (station C) and Przegalina (station A). We observed considerably higher values of ²¹⁰Po concentration in analysed water samples from Wiślinka area in comparison to Baltic water (0.59 mBq dm^{-3}), drinking and bottled water from Gdańsk agglomeration ($0.48-1.28 \text{ mBq dm}^{-3}$), beer from Poland (4.63 mBq dm^{-3}) and Vistula river water ($1.94-3.21 \text{ mBq dm}^{-3}$) [13,15,25].

The contents of ²¹⁰Po, ²³⁴U and ²³⁸U radionuclides in phosphogypsum are immediately connected with the phosphoric acid production technology and concentration of these radionuclides in phosphorites. The radionuclides are separated during the production of phosphoric acid. The uranium radionuclides stay behind in produced phosphoric acid, while the radium is present in phosphogypsum [3]. The lower uranium concentration in phosphogypsum samples and the higher polonium concentration permit to suppose that the uranium isotopes tend to be associated with the phosphoric acid fraction, while the 210 Po is bound to the phosphogypsum fraction [26]. About 80% of uranium and 3% of polonium stay in the filtered phosphoric acid fraction, while 15% of uranium and 90% of polonium remain with the phosphogypsum [26]. On the other hand, the concentration of natural alpha radionuclides in phosphorites lies in a very wide range. The phosphoric rock from the Southwest of Spain is rich in uranium isotopes, radium ²²⁶Ra and polonium ²¹⁰Po. Phosphorites contain 1300 to 1500 Bq kg⁻¹ of ²³⁸U and its daughter products [26]. The activities of ²¹⁰Po in Central and Northern Florida ranged between 860 and 1355 Bq kg⁻¹ in phosphate [24]. The activities 705 ± 50 , 745 ± 53 and 984 ± 80 Bq kg⁻¹ were measured in phosphate rocks from Senegal, 1005 ± 72 , 1043 ± 75 and $1303 \pm 93 \text{ Bq kg}^{-1}$ from Morocco as well as 1040 ± 77 , 1069 ± 79 and $931 \pm 66 \text{ Bq kg}^{-1}$ from Togo for ²³⁸U, ²³⁴U and ²¹⁰Po, respectively. The activities of ²³⁸U in phosphate rock from Florida ranged between 692 Bq kg⁻¹ (Northern Florida) and 1263 Bq kg⁻¹ (Central Florida) [24]. After production, 9% of the initial amount of ²²⁶Ra and 20% of uranium are deposited in waste heaps or industrial-used phosphogypsum [27].

4. Conclusion

The results obtained of uranium and polonium concentration in surface water from the area around the phosphogypsum waste dump and phosphogypsum samples allow us to conclude that the source of these radionuclides for the Wiślinka area are phosphorites and phosphogypsum, produced by Phosphoric Fertilisers Industry of Gdańsk. The higher concentrations of uranium and polonium radionuclides were determined for girdling ditch and the pumping station from the Wiślinka area $(2.1 \pm 0.1, 2.6 \pm 0.1 \text{ and } 4.5 \pm 0.1 \mu g L^{-1}$ for uranium and 5.50 ± 0.5 and $53 \pm 3 \text{ mBq L}^{-1}$ for ²¹⁰Po), but the highest were found in the retention reservoir $(430 \pm 5 \,\mu\text{g}\,\text{dm}^{-3}$ for total uranium in surface water, 900 ± 5 in bottom water and $200 \pm 10 \,\text{mBq}\,\text{L}^{-1}$ for ²¹⁰Po). The very high ²¹⁰Po concentration was found in phosphogypsum samples (600 ± 10 and 700 ± 10 Bq kg⁻¹). The observed differences between uranium concentration in phosphogypsum samples in 1996 and 2007 are probably a result of different phosphorites origins. The uranium is bound initially to the phosphogypsum and then is probably leached from the waste dump to the retention reservoir water. The processes of uranium leaching and precipitation seem to be responsible for the highest observed concentration in waters from the retention reservoir, girdling ditch and pumping station. The $^{234}U/^{238}U$ activity ratio is approximately one in the phosphogypsum samples, while the quotient is higher in waters. The polonium 210 Po is

more efficiently incorporated in phosphogypsum, but liberation to water is harder. The lower concentration of uranium and polonium radionuclides in water from the Martwa Wisła River indicated that the process of their release through canals in ramparts to the Vistula River water is not significant.

The influence of natural radionuclides contained in the phosphogypsum waste dump near Wiślinka village on the natural ecosystem (population, fauna and flora) should be realised and the determination of other higher radiotoxic nuclides, like radium ²²⁶Ra and radiolead ²¹⁰Pb, should be carried out.

Acknowledgements

The authors would like to thank the Polish Ministry of Higher Education and Sciences of the financial support of this work under grant: DS-8210-4-0086-8 and BW/8000-5-0109-8. We wish to thank particularly Bogusław Plewczyński for his assistance in sampling research material.

References

- Polskigips.pl: Fosfogips, Przemysłowe zastosowanie fosfogipsu <http://www.polskigips.pl/ index.php?id=125>.
- [2] Wiślinka ABC fosfogipsu. < http://wislinka.fosfory.pl/index.php?option=com_content&task=view&id=41&Itemid=9>.
- [3] Raport o stanie środowiska województwa pomorskiego w 2002 roku. < http://www.gdansk. wios.gov.pl/~web/test/raporty/rpt/rpt2002x.html>.
- [4] W. Brzyska, Lantanowce i aktynowce (Wydawnictwa Naukowo-Techniczne, Warszawa, 1987).
- [5] A. Meinrath, P. Schneider, and G. Meinrath, J Environ. Radioact. 64, 175 (2003).
- [6] A. Martínez-Aguirre, I. García-Orellana, and M. García-León, J. Environ. Radioact. 35 (2), 149 (1997).
- [7] B. Skwarzec, A. Boryło, and D.I. Strumińska, J. Environ. Radioact. 61, 345 (2000).
- [8] F. Bou-Rabee, Y. Bakir, and H. Bem, Environ. Int. 21, 293 (1995).
- [9] D. Bagatti, M.C. Cantone, A. Giussani, I. Veronese, P. Roth, E. Werner, and V. Höllriegl, J. Environ. Radioact. 65, 357 (2003).
- [10] A.K. Sam, M.M.O. Ahmed, F.A. El Khangi, Y.O. El Nigumi, and E. Holm, J. Environ. Radioact. 42, 65 (1999).
- [11] A.N. Niesmiejanow, Radiochemia (Państwowe Wydawnictwo Naukowe, Warszawa, 1975).
- [12] E. Browne and F.B. Firestone, in *Table of Radioactive Isotopes*, edited by V.S. Shirley (John Wiley and Sons, New York, 1986).
- [13] B. Skwarzec, Polon, uran i pluton w ekosystemie południowego Bałtyku (Rozprawy i monografie Instytutu Oceanologii PAN, 6, Sopot, 1995).
- [14] B. Skwarzec, A. Boryło, and D.I. Strumińska, Water, Air and Soil Pollut. 159, 165 (2004).
- [15] B. Skwarzec, D.I. Strumińska, and A. Boryło, Nukleonika. 51, 45 (2006).
- [16] A.C. Miller, W.F. Blakely, V. Livengood, T. Whittaker, J. Xu, W.J. Ejnik, M.M. Hamilton, E. Parlette, T.S. John, H.M. Gestenberg, and H. Hsu, Environ. Health Perspec. 106, 465 (1998).
- [17] http://epa.gov/safewater/radionuclides/index.html
- [18] World Health Organisation, Guidelines for Drinking Water Quality, 3rd edn < www.who.int/ water_sanitation_health/dwq/gdwq0506_9.pdf>.
- [19] B. Skwarzec, Ambio. 26 (2), 113 (1997).
- [20] B. Skwarzec, Stud. Mater. Oceanol. 162, 87 (1992).
- [21] B. Skwarzec, Chemia Analityczna. 42, 107 (1997).
- [22] P. Szefer, Stud. Mater. Oceanol. 51, 133 (1987).

- [23] J.P. Bolivar, R. García-Tenorio, and M. García-León, J. Radioanal. Nucl. Chem. 214, 77 (1996).
- [24] C.D. Hull and W.C. Burnett, J. Environ. Radioact. 32, 213 (1996).
- [25] B. Skwarzec and A. Jahnz, J. Environ. Sci. Health A 42, 2117 (2007).
- [26] J.P. Bolivar, R. García-Tenorio, and M. García-León, Appl. Radiat. Isot. 46, 6/7, 717 (1995).
- [27] R. Hedvall and B. Erlandsson, J. Environ. Radioact. 32, 19 (1996).