E3S Web of Conferences **1**, 11002 (2013)
DOI: 10.1051/e3sconf/20130111002

The concentrations of ^{241}Pu in the southern Baltic Sea

D. I. Strumińska-Parulska and B. Skwarzec

University of Gdańsk, Faculty of Chemistry, Sobieskiego 18, Gdańsk, POLAND, ***strumyk@chem.univ.gda.pl***

Abstract. The aim of the work was ^{241}Pu activities determination in different components (water, plankton and fish) of the southern Baltic Sea ecosystem. The determination of ^{241}Pu in the samples was done indirectly by activity measuring the increment in ^{241}Am from the decay of β-emitting ^{241}Pu in samples collected 10-15 years after the Chernobyl accident. Enhanced levels of ^{241}Pu were observed in all analyzed Baltic samples. The plutonium is also non-uniformly distributed between the organs and tissues of the analyzed fish; especially pelagic herring and cod as well as benthic flounder. The annual individual effective doses calculated on the basis of ^{241}Pu concentrations in fish indicated that the impact of the consumption of ^{241}Pu with Baltic fish on the annual effective dose for a statistical inhabitant of Poland was very small.

Key words: plutonium, ^{241}Pu, seawater, plankton, fish, Baltic Sea

Introduction

Most of all studies have focused on alpha emitting plutonium isotopes so far. ^{241}Pu is less important in terms of its radiotoxicity than the α-emitting plutonium radionuclides ^{238,239,240}Pu but is quite significant because of its huge contribution to the whole plutonium fallout. Moreover β-emitting ^{241}Pu ($T_{1/2}$=14.35 years) decays to the long-living, highly radiotoxic α-emitting ^{241}Am ($T_{1/2}$=432.2 years) (Mussalo et al., 1980). The main sources of plutonium in the marine environment are nuclear weapon tests, satellites and civil nuclear power plant accidents (Aarkrog, 1991). Since 26 April 1986 there has been a new source of plutonium, namely the Chernobyl accident that should be taken into consideration (Skwarzec, 1995; Strumińska and Skwarzec, 2006). Our previous experiments on air samples indicated extreme increase of ^{241}Pu amount in atmospheric dust; in April 1986 and the activity of ^{241}Pu reached 3643 mBq g^{-1} dw (Strumińska and Skwarzec, 2006). The available information about the bioaccumulation and distribution of ^{241}Pu in the Baltic Sea ecosystem and Poland territory is still very limited. The main purpose of the present work was to complete the present knowledge and estimate the further levels of the Baltic Sea environment contamination.

Materials and Methods

The samples of water, plankton and fish from the southern Baltic (the Gulf of Gdańsk, the Słupsk Bank, the Bornholm Deep and the Pomeranian Bay) were collected from 1997 to 2001 and the concentrations of ^{241}Pu were measured using the indirect method (Skwarzec, 1997; Skwarzec et al., 2001; Strumińska and Skwarzec, 2006). The plutonium samples were remeasured 10 years later using an alpha spectrometer Alpha Analyst Canberra Packard and the determination of ^{241}Pu was done by measuring the increment in ^{241}Am from the decay of β-emitting ^{241}Pu (Skwarzec et al., 2001; Strumińska, 2003). All ^{241}Pu activities were calculated on the sampling time on the basis of formula by Strumińska and Skwarzec (2006). The accuracy and precision of the plutonium analysis were less than 7% (1.5-6.4%), estimated by analysis of the IAEA standard materials.

Results and Discussion

Seawater

The results obtained for seawater samples are presented in Table 1. The highest total ^{241}Pu concentration in seawater was found in the Słupsk Bank (3.35±0.17 mBq dm^{-3}) and this area had the highest concentration of ^{241}Pu connected to suspended matter as well (1.94±0.12 mBq dm^{-3}). High concentrations of ^{241}Pu in the central part of the southern Baltic Sea can be a result of Baltic water circulation. In the southern Baltic Sea basins, the circulation has forms of separate circulation cells; thanks to the Earth rotation (Corliolis effect) the seawater moves anticlockwise (Groenwald, 2003). The highest activity of ^{241}Pu in colloidal fraction was observed in the Gdańsk

Bay. It is known the Vistula River carries a lot of plutonium; it is one of the biggest sources of plutonium in the southern Baltic (enriches the sea with 89.0 MBq of $^{239+240}$Pu annually) (Skwarzec et al., 2011). While $^{239+240}$Pu concentrations in the southern Baltic were increasing from the east to the west (Strumińska and Skwarzec, 2004), the total ^{241}Pu concentrations in seawater slightly increased from the west to the east. Obtained data showed increase of ^{241}Pu activities in comparison to our previous studies in May 1987 (Strumińska and Skwarzec, 2006). In water samples from the Gulf of Gdańsk and the Gdańsk Deep collected in 1999, the values of ^{241}Pu activities were almost 15 times higher (Table 1).

Table 1. $^{239+240}$Pu and ^{241}Pu concentrations in seawater, suspended matter and colloidal fraction from the southern Baltic collected in May 1999

Water fraction	$^{239+240}$Pu concentration* (µBq dm^{-3} ± SD)	^{241}Pu concentration (mBq dm^{-3} ± SD)
Gulf of Gdańsk (seaport)		
dissolved	2.8 ± 0.7	1.83 ± 0.09
suspended	1.6 ± 0.4	0.59 ± 0.04
colloidal	0.8 ± 0.2	0.22 ± 0.02
total	5.2 ± 0.8	2.57 ± 0.46
Gdańsk Deep		
dissolved	7.9 ± 0.6	2.19 ± 0.04
suspended	2.5 ± 0.2	0.16 ± 0.01
colloidal	0.7 ± 0.1	0.16 ± 0.01
total	11.1 ± 0.6	2.48 ± 0.04
Słupsk Bank		
dissolved	17.1 ± 2.3	1.56 ± 0.55
suspended	3.8 ± 0.9	1.94 ± 0.12
colloidal	1.1 ± 0.1	0.08 ± 0.01
total	20.9 ± 2.5	3.35 ± 0.17
Pomeranian Bay		
dissolved	145 ± 4	1.86 ± 0.03
suspended	4.0 ± 0.4	0.32 ± 0.01
colloidal	1.0 ± 0.2	0.05 ± 0.01
total	150 ± 4	2.21 ± 0.07

* (Strumińska and Skwarzec, 2004)

Despite all these years from the Chernobyl accident, the radiological effects are still observed in the environment and mean late plutonium inflow effect from Baltic catchment area.

Phyto- and Zooplankton

The results obtained for phyto- and zooplankton samples are presented in Table 2. The ^{241}Pu activity in phytoplankton sample from the Pomeranian Bay was 1.06±0.09 mBq g^{-1} dw. Within zooplankton samples the highest ^{241}Pu activity was found in samples from the central part of the southern Baltic (2.66±0.16 mBq g^{-1} dw) and from the Gdańsk Deep (2.64±0.70 mBq g^{-1} dw) (Table 2). In zooplankton samples, similar situation to seawater samples was noticed – the highest concentrations of ^{241}Pu were found in the central part of the southern Baltic Sea, and similarly to seawater it could be a result of Baltic water circulation.

Table 2. $^{239+240}$Pu and ^{241}Pu concentrations in phyto- and zooplankton from the southern Baltic Sea collected in May 1999

Sampling site	$^{239+240}$Pu concentration* (µBq g^{-1} dw ± SD)	^{241}Pu concentration (mBq g^{-1} dw ± SD)
Phytoplankton		
Pomeranian Bay	6.31 ± 0.89	1.06 ± 0.09
Zooplankton		
Gulf of Gdańsk	5.19 ± 0.57	1.56 ± 0.13
Gdańsk Deep	4.98 ± 0.69	2.64 ± 0.70
Open sea 1	5.15 ± 0.75	2.66 ± 0.16
Open sea 2	3.45 ± 0.76	2.24 ± 0.23
Oderbank	1.33 ± 0.21	1.10 ± 0.11

* (Strumińska, 2003)

Fish

The results of ^{241}Pu concentrations determination in fish from the southern Baltic are presented in Table 3. Generally the data show, similarly to $^{239+240}$Pu activities, significant differences in ^{241}Pu concentrations among all the species examined. The highest values of ^{241}Pu activities for whole organism were found in fish from *Perciformes:* benthic round goby (0.863±0.066 mBq g^{-1} ww) and pelagic perch (0.666±0.001 mBq g^{-1} ww). Both fish species feed on benthic invertebrates and small fish; especially round goby feeds mainly on blue mussel (*Mytilus trossulus*) and remobilizes heavy metals and radionuclides accumulated by these organisms (Kostrzewa et al., 2004). The lowest ^{241}Pu activity was found in flounder (0.104±0.009 mBq g^{-1} ww). The flounder is benthic fish but feeds on plankton and insects larvae when young and benthic invertebrates and small fish when adult. The differences in ^{241}Pu activity in the whole body in round goby and flounder could be caused by their diet. The plutonium was also non-uniformly distributed between the organs and tissues of the analyzed fish, especially pelagic herring and cod as well as benthic flounder. Most of ^{241}Pu in herring, cod and flounder was located in soft tissues (Table 3). The tendency found during ^{241}Pu analysis agreed to our previous studies on $^{239+240}$Pu. The amount of plutonium in fish alimentary system confirms its role in plutonium intake. The participation of fish alimentary system in plutonium bioaccumulation depends on the organ function; the fish's feeding habits and its location in the Baltic Sea.

Table 3. $^{239+240}Pu$ and ^{241}Pu concentrations in fish collected in the Gulf of Gdańsk in 1997

Sample	Sample		$^{239+240}Pu$ concentration*	^{241}Pu concentration	Part of total ^{241}Pu	$^{241}Pu/^{239+240}Pu$ activity ratio
	Wet weight (g)	Contribution (%)	(µBq g^{-1} ww ± SD)	(mBq g^{-1} ww ± SD)	(%)	
Herring (*Clupea harengus*) (n=29)						
muscle	1177	53.8	0.24 ± 0.05	0.034 ± 0.004	5.8	146 ± 34
skeleton	515	23.5	1.01 ± 0.11	0.090 ± 0.006	6.7	89 ± 11
skin	211	9.6	1.12 ± 0.14	0.102 ± 0.005	3.1	91 ± 12
gills	61	2.8	37.7 ± 3.50	3.83 ± 0.11	33.7	101 ± 10
stomach	47	2.1	2.31 ± 0.42	0.227 ± 0.014	1.5	98 ± 19
intestine	76	3.5	4.10 ± 0.39	0.532 ± 0.024	5.8	130 ± 14
liver	3	0.1	9.36 ± 1.13	7.70 ± 0.70	3.3	822 ± 124
gonads	63	2.9	4.53 ± 0.42	0.556 ± 0.032	5.1	123 ± 14
rest	34	1.6	17.2 ± 1.80	7.12 ± 0.21	35.0	415 ± 46
whole body	2187	100.0	2.22 ± 0.11	0.317 ± 0.013	100.0	143 ± 9
Cod (*Gadus morhua*) (n=5)						
muscle	1660	48.6	0.29 ± 0.06	0.047 ± 0.004	9.0	163 ± 35
skeleton	988	28.9	0.91 ± 0.20	0.135 ± 0.012	15.4	148 ± 35
skin	238	7.0	0.55 ± 0.08	0.180 ± 0.011	4.9	328 ± 51
gills	111	3.3	6.22 ± 0.78	0.336 ± 0.036	4.3	54 ± 9
stomach	73	2.1	1.98 ± 0.26	0.089 ± 0.011	0.8	45 ± 8
intestine	76	2.2	70.5 ± 3.60	6.96 ± 0.23	61.0	99 ± 6
liver	234	6.9	0.40 ± 0.09	0.024 ± 0.003	0.7	60 ± 14
gonads	25	0.7	4.08 ± 0.70	1.012 ± 0.050	2.9	248 ± 44
rest	9	0.3	1.12 ± 0.23	1.102 ± 0.097	1.1	984 ± 218
whole body	3414	100.0	2.35 ± 0.11	0.254 ± 0.013	100.0	108 ± 8
Perch (*Perca fluviatilis*) (n=12)						
muscle	209	38.1	0.52 ± 0.09	0.197 ± 0.018	11.3	381 ± 76
skeleton	186	34.1	0.50 ± 0.13	0.238 ± 0.023	12.2	473 ± 130
skin	65	11.9	3.25 ± 0.61	0.479 ± 0.175	8.5	147 ± 61
gills	212	7.9	3.65 ± 0.98	0.208 ± 0.028	12.1	57 ± 17
stomach	9	1.7	5.51 ± 1.23	4.16 ± 0.27	10.4	755 ± 176
intestine	10	1.8	26.7 ± 4.80	4.09 ± 0.65	11.3	153 ± 37
liver	7	1.3	6.62 ± 1.91	5.01 ± 0.45	10.1	756 ± 229
gonads	5	0.9	14.4 ± 2.90	7.85 ± 0.72	11.1	549 ± 123
rest	12	2.3	4.98 ± 1.29	3.79 ± 0.26	13.0	762 ± 203
whole body	547	100.0	1.96 ± 0.03	0.666 ± 0.001	100.0	340 ± 42
Flounder (*Platichthys flesus*) (n=14)						
muscle	1120	38.2	0.13 ± 0.02	0.006 ± 0.001	2.4	48 ± 9
skeleton	1041	35.5	0.67 ± 0.10	0.049 ± 0.007	16.6	73 ± 15
skin	366	12.5	0.63 ± 0.10	0.147 ± 0.006	17.6	231 ± 38
gills	87	3.0	5.32 ± 0.63	0.286 ± 0.023	8.1	54 ± 8
stomach	53	1.8	2.03 ± 0.32	0.171 ± 0.027	3.0	84 ± 19
intestine	113	3.9	7.11 ± 0.53	0.893 ± 0.051	33.0	126 ± 12
liver	74	2.5	1.51 ± 0.20	0.238 ± 0.027	5.8	158 ± 27
gonads	48	1.6	1.68 ± 0.29	0.344 ± 0.035	5.4	204 ± 41
rest	31	1.1	3.80 ± 0.61	0.810 ± 0.104	8.2	213 ± 44
whole body	2933	100.0	0.94 ± 0.05	0.104 ± 0.009	100.0	111 ± 11
Round goby (*Neogobius melanostomus*) (n=17)						
muscle	272	31.3	0.88 ± 0.10	0.294 ± 0.019	10.7	336 ± 44
skeleton	369	42.5	3.24 ± 0.27	0.191 ± 0.017	9.4	59 ± 7
skin	70	8.0	14.9 ± 1.70	1.079 ± 0.119	10.0	72 ± 12
gills	60	6.9	22.5 ± 1.50	2.046 ± 0.201	16.3	91 ± 11
stomach	7	0.9	36.3 ± 3.80	9.88 ± 0.65	9.8	272 ± 34
intestine	13	1.5	51.2 ± 7.70	6.15 ± 0.52	10.6	120 ± 34
liver	47	5.4	5.97 ± 0.84	1.719 ± 0.127	10.8	288 ±46
gonads	14	1.6	34.2 ± 3.20	6.56 ± 0.36	12.4	192 ± 21
rest	17	1.9	8.88 ± 1.42	4.54 ± 0.21	10.2	512 ± 85
whole body	868	100.0	6.52 ± 0.57	0.863 ± 0.066	100.0	132 ± 15

* (Skwarzec et al., 2001; Strumińska, 2003)

Radiation Doses

It has been estimated that the yearly intake of ^{241}Pu via the consumption of Baltic fish by Poles (about 5 kg year^{-1}) equaled the intake of average value of 2.2 Bq per capita. According to muscles only (filet) the average consumption value decreased to 0.6 Bq. The annual individual effective dose values calculated on this basis (using the conversion factor given by the UNSCEAR (2000) – 4.8 nSv Bq^{-1} for ^{241}Pu) are 10.6 nSv per whole fish and 2.8 nSv according to muscles (filet). The total natural radiation exposure in Poland leads to an annual effective dose of 2.8 mSv (including ^{222}Rn) (Jagielak et al, 1997). This indicates the impact of the consumption of ^{241}Pu with Baltic fish on the annual effective dose for a statistical inhabitant of Poland is very small.

Acknowledgments

The authors would like to thank the Ministry of Science and Higher Education for the financial support under grant DS/8120-4-0176-12.

References

1. Aarkrog A. Source of terms and inventories of anthropogenic radionuclides. Environmental Science and Technology Department/Ecology Section. Risø National Laboratory, Roskilde, Denmark; 1991.
2. Groenwald M. Geografia morza. Gdańsk: Podkowa; 2003
3. Jagielak J, Biernacka M, Henschke A, Sosińska A. Radiologiczny Atlas Polski. Warszawa: PIOŚ, CLOR, PAA; 1997.
4. Kostrzewa J, Grabowski M, Zięba G. Nowe inwazyjne gatunki ryb w wodach Polski. Arch Pol Fish 2004;12(2):21.
5. Mussalo H, Jaakkola T, Miettinen, JK. Distribution of fallout plutonium in Southern Finns. Health Phys 1980;39(2):245.
6. Skwarzec B. Polon, uran i pluton w ekosystemie południowego Bałtyku. 6. Rozprawy i monografie, IO PAN. Sopot; 1995.
7. Skwarzec B. Radiochemical methods for the determination of polonium, radiolead, uranium and plutonium in environmental samples. Chem Anal (Warsaw) 1997;42:107.
8. Skwarzec B, Strumińska DI, Boryło A. Bioaccumulation and distribution of plutonium in fish from Gdansk Bay. J Environ Radioact 2001;55:167.
9. Skwarzec B, Jahnz-Bielawska A, Strumińska-Parulska DI. The inflow of ^{238}Pu and $^{239+240}Pu$ from the Vistula River catchment area to the Baltic Sea. J Environ Radioact 2011;102:728.
10. Strumińska DI. Nagromadzanie plutonu w planktonie i rybach oraz jego bilans w Zatoce Gdańskiej. PhD thesis. Gdańsk University, Faculty of Chemistry; 2003.
11. Strumińska DI, Skwarzec B. Plutonium concentrations in water from the southern Baltic Sea and their distribution in cod (*Gadus morhua*) skin and gills. J Environ Radioact 2004;72:355.
12. Strumińska DI, Skwarzec B. Plutonium ^{241}Pu concentrations in southern Baltic Sea ecosystem. J Radioanal Nucl Chem 2006;268(1):59.
13. Strumińska-Parulska DI, Skwarzec B. Plutonium isotopes ^{238}Pu, $^{239+240}Pu$, ^{241}Pu and $^{240}Pu/^{239}Pu$ atomic ratios in the southern Baltic Sea ecosystem. Oceanologia 2010;52(3):499.
14. UNSCEAR. Sources and effects of ionizing radiation, Vol I: Sources, Appendix A 2000.