Vertical distribution of ¹³⁷Cs, ²¹⁰Pb, ²²⁶Ra and ^{239,240}Pu in bottom sediments from the Southern Baltic Sea in the years 1998–2000

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Abstract This paper presents vertical distribution of ¹³⁷Cs, ²²⁶Ra and ^{239,240}Pu in bottom sediments collected from the Southern Baltic Sea in the years 1998–2000. In the southern part of Baltic Sea the highest concentrations of ¹³⁷Cs and ^{239,240}Pu were observed at the Gulf of Gdansk. Deposition of ¹³⁷Cs ranged from 1990±402 Bq m⁻² in the Bornholm Basin to 3260 ± 820 Bq m⁻² in the Gulf of Gdansk. Depositions of ^{239,240}Pu in the Bornholm Basin were 28.1–30.4 Bq m⁻² and in Gulf of Gdansk 162–174 Bq m⁻². The concentration peaks of long-lived radionuclides, owing to the sedimentation processes, show the maximum fallout period in different sediment layers. The observed differences in distribution of radiocaesium and plutonium along the profile confirm two main sources of contamination – Chernobyl fallout for ¹³⁷Cs and global fallout in case of ^{239,240}Pu. In chosen core samples from the Gdansk Basin vertical distribution of ²¹⁰Pb concentrations were determined. Evaluated sedimentation rates based on decrease of unsupported ²¹⁰Pb ranged in the Gdansk Basin from 1.9 to 2.3 mm year⁻¹. Calculations based on ^{239,240}Pu peaks show sedimentation rate, in the range from 1.6–2.2 mm year⁻¹ for P110 region.

Key words Baltic Sea • contamination • radionuclides • sedimentation rate

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Received: 10 September 2001, Accepted: 29 March 2002

Introduction

The total input of ¹³⁷Cs from the Chernobyl accident into the Baltic Sea area was estimated to be 4.7 PBq [6]. It was the main source of artificial radionuclides. Another source of radioactive contamination in this environment was the fallout after nuclear weapons tests. The Baltic Sea extends from 54°N to 66°N, which means that it belongs to the most intensive global fallout zone. The cumulative deposition of ¹³⁷Cs from this source was estimated to be 0.93 PBq [7]. The input of ¹³⁷Cs from discharges of Sellafield and La Hague transported by the inflow of saline water through the Danish Straits was estimated to be 0.38 PBq. The Baltic Sea nuclear installation input was estimated to be 0.65 TBq [7].

The activities of ¹³⁷Cs, as well as plutonium, in sediments reflect deposition directly to the Baltic Sea and through the drainage areas. However, these activities depend also on the sediment type and sedimentation rate at various locations. In the bottom sediments for 1991 inventories of 1.4 PBq for ¹³⁷Cs and 0.018 PBq of ^{239,240}Pu were estimated [4]. Compared to the early 1980s these inventories have increased by factors of 5.1 and 1.2, respectively [4].

The knowledge on vertical distributions of artificial radionuclides in bottom sediments gives opportunity to observe current contamination of the marine environment as well as its changes in time. Additionally, taking into account vertical distributions of ²¹⁰Pb and ^{239,240}Pu, the sedimentation rate in different regions of the Baltic Sea may be evaluated.

Experiments

Bottom sediment samples were collected from various regions of southern part of the Baltic Sea (Fig. 1), during the sampling cruises into the Baltic Sea with r/v "Baltica" organized once a year by the Institute of Meteorology and Water Management (July 1998, August 1999 and 2000). At each sampling station, five parallel core samples have been taken with a gravity corer (Niemisto type with inner diameter 55 mm). Core samples were sectioned into 1 or 2 cm sub-samples from 0 to 19 cm depth, and parallel sub-samples were combined for analyses. The determination of ¹³⁷Cs and ²²⁶Ra were performed each year and determinations of ²³⁸Pu and ^{239,240}Pu in two years period. ²¹⁰Pb was analyzed only in selected samples from the Gdansk Basin. The co-ordinates and depth of sampling stations are presented in Table 1.

The ¹³⁷Cs activity concentration was determined by gamma spectrometry with an HPGe detector, with energy resolution 1.8 keV for ⁶⁰Co (1332 keV) and a relative efficiency of 30%. The detector was connected to a multichannel analyser, Canberra, Series 90. Minimum detectable activity (MDA) for counting time 86,000 s was 0.06 Bg/sample (1.5 Bg kg⁻¹) [11]. Plutonium was separated by ion exchange, followed by electrodeposition onto stainless steel disks. ²⁴²Pu was used as an internal tracer for counting alpha activity and chemical recovery. Activity of plutonium was measured by alpha spectrometry using a PIPS detector with an efficiency of 32% placed in a vacuum chamber. MDA for counting time of 164,000 s was 0.2 mBq/sample (0.007 Bq kg⁻¹) [10]. Concentration of ²²⁶Ra was determined radiochemically using the emanation method (measurement of ²²²Rn in Lucas-type scintillation chambers) preceded by separation of radium [1]. MDA with the counting time of 21,600 s was equal to 0.73 mBq/sample (0.73 Bq kg⁻¹). Concentration of

 Table 1. Sampling stations for bottom sediment in HELCOM MORS programme.

Stations code	Sampling area	Depth (m)	Co-ordinates	
P110 P116 P1	Gulf of Gdansk Gdansk Deep	64–71 86–90 106–107	54°30'N;19°06.8'E 54°39.1'N;19°17.6'E 54°50'N:19°20'E	
P140 P5 P39	open sea area Bornholm Basin	88–89 88–91 63–68	55°33'N;18°24'E 54°15'N;15°59'E 54°44.5'N;15°08'E	

²¹⁰Pb was determined by beta activity measurement of ²¹⁰Bi deposited on a nickel disk. MDA with the counting time of 21,600 s was equal to 0.017 Bq/sample (3.4 Bq kg⁻¹) [1]. Minimum detectable activities were calculated at the probability of the type I and II errors equal to 0.05.

The reliability of applied methods was checked by participation in inter-comparison exercises organized by the IAEA and Risø National Laboratory (Table 2).

Results

The concentrations of 137 Cs in sediments differ depending on sampling site and sampling depth. The highest concentrations were found in the Gulf of Gdansk (Table 3). In the upper 0–3 cm layer of sediments, 137 Cs concentrations in the years 1998–2000 ranged from 220 to 393 Bq kg⁻¹ d.w. (P110 and P116) and their values decrease along the profiles (Fig. 2). In the sediments from the Bornholm Basin (P39 and P5), the 137 Cs concentrations were evidently lower and ranged from 77 to 129 Bq kg⁻¹ d.w. (Table 3). Similar values (87–117 Bq kg⁻¹ d.w.) were observed in the Pomeranian Bay by Bojanowski *et al.* [2]. In the years 1998–2000, the average deposition of 137 Cs for the Gulf of Gdansk was equal to 3260±820 Bq m⁻². Much lower deposition of 137 Cs, 1990±402 Bq m⁻², was found for



Fig. 1. Sampling sites of sediment for measurements of radioactive substances, 1998–2000.

Nuclide	CLRP Median AQCS (range accept		CLRP	Median AQCS values (range accepted)	
	IAF	EA 378	IAI	EA 379	
¹³⁷ Cs ²¹⁰ Pb*) ²²⁶ Ra ^{239,240} Pu	5.4 ± 0.5 207 ± 11.8 132 ± 15 0.120 ± 0.024	5.7 (3.2–6.5) 188 (153–191) 118 (103.2–132) 0.135 (0.11–0.15)	37.9 ± 3.0 212 ± 10.4 40.6 ± 5.6 3.6 ± 0.5	39.5 (37.9–45) 229 (204–240) 29 (22.1–48) 3.8 (3.2–4.1)	
	IAEA 300		IAEA 381		
¹³⁴ Cs ¹³⁷ Cs ²³⁸ Pu ^{239,240} Pu	$73.3 \pm 10.9 \\ 1097 \pm 142 \\ - \\ 3.2 \pm 0.4$	69 (57–94) 1053 (940–1101) - 3.43 (3.09–3.9)	0.496 ± 0.02 3.1 ± 0.4 13.2 ± 1.5	0.482 (0.48–0.50) 3.17 (3.1–3.5) 13.2 (13.0–14.0)	

Table 2. ¹³⁷Cs, ²¹⁰Pb, ²²⁶Ra and ^{239,240}Pu measurements in inter-comparison samples.

*) determinations of ²¹⁰Pb in the IAEA samples

the Bornholm Basin. The differences in deposition of 137 Cs
between consecutive years, observed in particular sampling
sites, indicate that even at small area the radionuclide con-
centration may vary to a large degree.

Similarly as for radiocaesium the highest concentrations of plutonium in bottom sediments were found in the Gulf of Gdansk, however the maxima of ²³⁸Pu and ^{239,240}Pu were observed always in deeper layers. In core samples taken from station P110 the maximum of ^{239,240}Pu concentration, 7.2 ± 1.2 Bq kg⁻¹ d.w., was observed in 7–9 cm layer in 1998 and 6.1 ± 0.66 Bq kg⁻¹ d.w. in 5–7 cm layer in 2000 (Table 4a). In core samples from P116 station maxima of concentrations 5.27 ± 0.62 and 5.46 ± 0.59 Bq kg⁻¹ d.w. were found in 4-5 cm layers in both years (1998 and 1999). The concentrations of plutonium in the Bornholm Basin are uniform along the profiles even to the depth of 11 cm (Table 4b) and concentrations of ^{239,240}Pu ranged from 0.785 to 1.01 Bq kg⁻¹ d.w. in 1998. The deposition of ^{239,240}Pu were found to be 28.1–30.4 Bq m⁻² in the Bornholm Basin (P5), 95 Bq m⁻² in the Gdansk Deep (P1) and 162-174 Bq m⁻² in the Gulf of Gdansk (P110)(Table 4a). Similar values were found in previous years [10–12]. Comparable values (48 Bq m⁻²

for the Bornholm Basin and 98 Bq m⁻² for the Gdansk Deep) were determined also by Skwarzec and Bojanowski [9].

as secondary standard material.

The distributions of ²²⁶Ra concentration were similar along the profiles (Fig. 3), however, differences between particular sub-regions were observed. Lower concentration of ²²⁶Ra, 28±1.4 Bq kg⁻¹ d.w., were found in the Gulf of Gdansk (P110, P116) and higher, 40 ± 1.7 Bq kg⁻¹ d.w. and 46 ± 2.3 in the Gdansk Deep (P1) and the Bornholm Deep (P5), respectively.

Determinations of ²¹⁰Pb were performed in four core samples from the Gdansk Basin. ²¹⁰Pb concentration ranged from 338 to 434 Bq kg⁻¹ d.w. in the first 0–1 cm layer in the Gdansk Basin area and decreased exponentially along the profiles to the 40–85 Bq kg⁻¹ d.w. in the layer 17–19 cm depending on the sampling site (Table 5). The deposition of unsupported ²¹⁰Pb, for sediment layer 0–19 cm, observed in P110 sampling site was 9960±310 Bq m⁻² in 1998 and 7400±249 Bq m⁻² in 2000. In sampling site P1 this deposition was equal to 7020±191 Bq m⁻² and in P116 to 5480±173 Bq m⁻². The concentrations in the analyzed lowest layers were still 14.2–46.2 Bq kg⁻¹ d.w. depending of sampling site.



Fig. 2. Vertical distribution of ¹³⁷Cs in bottom sediments from the Southern Baltic Sea.

Table 3. Caesium-137 in layers of bottom sediment from the Gdansk Basin (P110, P116, P1) and from the Bornholm Basin (P5, P39), content of this radionuclide in particular layers and its total deposition.

		¹³⁷ Cs					
Station	Layer	1998		199	99	2000	
	(cm)	Activity (Bq kg ⁻¹ d.w.)	Content ^{a)} (Bq m ⁻²)	Activity (Bq kg ⁻¹ d.w.)	Content (Bq m ⁻²)	Activity (Bq kg ⁻¹ d.w.)	Content (Bq m ⁻²)
P110	0 - 1	$312 \pm 8.70^{\text{b}}$	$409 \pm 36.0^{\circ}$	361 ± 13.0	713±68.4	272±8.72	311±28.6
	1 - 2	299 ± 11.4	648 ± 63.5	328 ± 13.8	795 ± 81.1	232 ± 8.60	632 ± 61.8
	2 - 3 3 - 4	255 ± 9.50 162 ± 7.42	011 ± 02.4 449 ± 47.6	249 ± 9.71 165 + 8 56	465 ± 52.0	103 ± 7.00 93 4 + 3 46	362+351
	4 - 5	102 ± 6.10	311±37.3	105 ± 0.50 108 ± 5.39	315 ± 34.6	78.2 ± 4.70	280 ± 33.6
	5 - 7	77.6 ± 4.96	429 ± 40.3	88.8 ± 2.91	554 ± 34.9	63.9 ± 3.71	456 ± 40.1
	7-9	69.6 ± 2.92	454±32.7	61.1 ± 2.93	469 ± 36.2	35.0 ± 2.10	308 ± 27.8
	9 – 11	41.5 ± 2.62	325 ± 30.0	19.9 ± 1.61	198 ± 22.0	12.1 ± 1.15	181 ± 22.8
	11 - 13 13 15	$12.6 \pm 1.4 /$ 3.08 ± 0.720	112 ± 10.5 31 7 + 8 47	5.12 ± 0.830 2.59 ± 0.565	52.0 ± 10.0 25.8 ± 6.41	5.19 ± 1.04	53.6 ± 12.4
	15 - 15 15 - 17	2.04 ± 0.300	22.4 ± 3.96	2.59 ± 0.505 3.56 ± 0.797	25.8 ± 0.41 35.6 ± 8.98	<mda <mda< td=""><td>_</td></mda<></mda 	_
	17 – 19	2.17 ± 0.353	21.2 ± 4.10	2.33 ± 0.596	21.8 ± 6.22	<mda< td=""><td>_</td></mda<>	_
	0 – 19		38±130 ^{d)}		4290±150		3140 ± 115
P116	$ \begin{array}{ccc} 0 - 1 \\ 1 & 2 \end{array} $	374 ± 13.1	269 ± 25.6	332 ± 9.62	506 ± 45.0	329 ± 11.2	146 ± 13.7
	1 - 2 2 - 3	257 ± 10.2 220 ± 11.0	302 ± 35.5 306 ± 33.7	333 ± 9.17 267+8.28	443 ± 38.3 444 ± 40.4	393 ± 11.0 326 ± 11.1	398 ± 33.0 330 ± 31.0
	3 - 4	153 ± 9.80	277 ± 34.4	140 ± 4.77	236 ± 22.2	230 ± 9.45	337 ± 34.0
	4 - 5	125 ± 7.99	262 ± 32.5	103 ± 7.43	204 ± 26.9	159 ± 8.57	266 ± 30.4
	5 - 7	85.9 ± 4.64	356 ± 29.9	64.7 ± 4.60	263 ± 26.5	134 ± 3.61	591±33.7
	7 - 9	41.0 ± 2.42	206 ± 18.4	34.5 ± 1.90	178 ± 15.1	116 ± 3.59	451 ± 27.5
	9 – 11 11 13	18.0 ± 1.79 5.07 ± 0.674	$114 \pm 14./$ 30.6 ± 4.08	19.8 ± 0.814 10.2 + 1.12	88.4 ± 6.28 54 1 + 7 58	84.6 ± 2.45 61.8 ± 2.78	366 ± 21.6 314 ± 23.6
	11 - 15 13 - 15	3.14 ± 0.517	21.9 ± 4.27	4.11 ± 0.621	24.5 + 4.44	40.0 ± 2.16	200+16.8
	15 – 17	<mda< td=""><td>_</td><td>2.04 ± 0.468</td><td>12.3 ± 3.39</td><td>26.6 ± 1.12</td><td>154±11.1</td></mda<>	_	2.04 ± 0.468	12.3 ± 3.39	26.6 ± 1.12	154±11.1
	17 – 19 0 – 19	2.15 ± 0.624	17.0 ± 5.45 2170 ± 81.7	<mda< td=""><td>_ 2460±86.1</td><td>14.7 ± 1.01</td><td>109 ± 10.8 3660 ± 88.8</td></mda<>	_ 2460±86.1	14.7 ± 1.01	109 ± 10.8 3660 ± 88.8
P1	0 - 1	349 + 12.2	279+265	321+8.68	379+32.9	368+12.2	241+22.4
••	1 - 2	299 ± 13.2	387 ± 40.3	279 ± 11.2	440 ± 44.0	246 ± 10.8	357±37.1
	2 - 3	275 ± 12.9	409 ± 43.8	249 ± 76.96	427±37.6	185 ± 6.12	332 ± 30.9
	3 - 4	304 ± 12.8	471 ± 48.1	273 ± 11.2	506 ± 51.1	118 ± 5.48	258 ± 27.4
	4 - 5	244 ± 11.2	453 ± 48.0	234 ± 10.6	484 ± 50.8	56.6 ± 3.81	133 ± 17.0
	3 - 7 7 - 9	76+3.38	407 ± 30.9 323 ± 23.9	103 ± 3.70 130 + 7.42	539+46.9	11.2 ± 1.15	59.4 + 7.89
	9 – 11	46.8 ± 2.62	227 ± 19.5	93.4 ± 3.18	408 ± 26.1	3.94 ± 0.81	23.5 ± 5.53
	11 – 13	32.5 ± 2.37	172 ± 17.7	40.6 ± 2.40	183 ± 16.3	2.30 ± 0.66	13.6 ± 3.89
	13 – 15	11.0 ± 1.41	62.7±10.0	15.7 ± 1.58	78.6 ± 10.3	2.78 ± 0.66	17.2 ± 4.10
	15 - 17	5.15 ± 0.70	28.0 ± 4.67	6.86 ± 0.92	34.9 ± 5.72	<mda< td=""><td>-</td></mda<>	-
	17 – 19 0 – 19	3.01±0.32	19.0 ± 3.39 3300 ± 108	7.13±1.05	38.0 ± 6.73 4144±66	< MDA	1627 ± 30
P5	0 - 1	118 ± 5.86^{b}	$166 \pm 18.2^{\circ}$) 122±5.23	196 ± 20.2	90.2±6.13	103 ± 13.1
	1 - 2	85.6 ± 6.93	171 ± 24.1	107 ± 5.43	177 ± 19.7	92.2 ± 6.63	145 ± 17.5
	2 - 3 3 - 4	70.4 ± 0.04 79.0 ± 6.88	$1/1\pm 23.0$ 102 ± 28.3	91.4 ± 4.57 86.8 + 5.64	$1/3 \pm 19.1$ 195 ± 24.4	74.3 ± 4.61 82.8 + 5.05	105 ± 20.2 184 ± 22.3
	4 - 5	53.7 ± 3.11	130 ± 15.4	67.0 ± 3.28	155 ± 24.4 156 ± 17.0	72.6 ± 4.79	104 ± 22.0 182 ± 23.0
	5 - 7	64.6 ± 4.52	352±35.3	50.2 ± 2.21	245 ± 18.1	68.9 ± 3.17	297±22.6
	7-9	57.8 ± 2.83	300 ± 23.7	23.8 ± 2.02	126 ± 14.5	45.5 ± 2.91	222 ± 20.9
	9 – 11	58.3 ± 2.86	293 ± 23.1	16.6 ± 1.49	97.4±11.7	30.9 ± 2.04	161 ± 15.5
	11 - 13 13 - 15	54.0 ± 2.97 34.1 ± 2.15	$2/1 \pm 23.2$ 184 + 17 1	$1/.1 \pm 1.54$ 13.0 ± 1.30	88.0 ± 10.6	11.9 ± 0.869 8.06 ± 0.604	12.2 ± 1.44 48.9 ± 5.13
	15 - 15 15 - 17	16.6 ± 1.11	104 ± 17.1 104 ± 10.1	6.09 ± 0.938	32.9 ± 6.05	2.51 ± 0.388	14.0 ± 2.58
	17 – 19	16.0 ± 0.98	101±9.20	5.32 ± 1.04	29.8 ± 6.74	<mda< td=""><td>-</td></mda<>	-
	0 – 19		2440±76.6 ^d)	1590 ± 54.7		1600 ± 56.4
P39	0 - 1	114 ± 5.69	163 ± 17.9			120 ± 6.51	177 ± 20.2
	1 - 2 2 - 3	120±9.96 123+8 73	229 ± 31.9 263 ± 34.5			129±6.69 115+6.42	290±33.1 290+33.6
	$\frac{2}{3} - 4$	114 ± 8.70	277 ± 37.7			87.6 ± 4.82	275 ± 31.6
	4 - 5	111 ± 9.06	312±44.3			84.0 ± 3.91	276 ± 29.4
	5 - 7	76.8 ± 6.14	388 ± 42.7			46.4 ± 2.60	266 ± 22.8
	7 - 9	55.5 ± 2.61	277 ± 21.3			27.6 ± 1.71	155 ± 14.3
	9 – 11 11. 12	49.0 ± 3.59 26.8 + 2.10	190 ± 23.2 142 ± 16.0			$13./\pm1.32$ 10.0+1.10	75.4 ± 9.50
	11 - 13 13 - 15	13.8 ± 1.63	72.4 ± 10.0			6.90 ± 0.690	41.1 ± 5.35
	15 – 17	4.42 ± 1.10	24.3±6.79			4.23 ± 0.583	24.9±4.19
	17 – 19	2.84 ± 0.440	14.9 ± 2.76			3.57 ± 0.603	24.1 ± 4.80
	0 – 19		2350 ± 95.7				1960 ± 73.8

^{a)} content of ¹³⁷Cs was calculated as activity of radionuclide in sample divided by sampling surface.

b) activity concentration \pm uncertainty due to counting of sample at the 95% confidence level.

 $^{\rm c)}$ content \pm uncertainty due to counting of sample and sampling at the 95% confidence level.

^{d)} total deposition \pm combined uncertainty due to counting of sample and sampling of all layers at the 95% confidence level. Table 4a. Plutonium-239,240 in layers of bottom sediments from the Gdansk Basin, content of this radionuclide in particular layers and its total deposition.

	^{239,240} Pu							
-	P110/1998		P110/2000		P116/1998		P116/1999	
Layer (cm)	Activity (Bq kg ⁻¹ d.w.)	Content ^{a)} (Bq m ⁻²)	Activity (Bq kg ⁻¹ d.w.)	Content (Bq m ⁻²)	Activity (Bq kg ⁻¹ d.w.)	Content ^{a)} (Bq m ⁻²)	Activity (Bq kg ⁻¹ d.w.)	Content (Bq m ⁻²)
0 - 1	1.29 ± 0.22	1.72 ± 0.39	2.85 ± 0.36	3.25 ± 0.60	3.73±0.47	2.70 ± 0.50	3.44 ± 0.49	5.23 ± 1.06
1 - 2	1.56 ± 0.25	3.39 ± 0.74	2.51 ± 0.35	6.87 ± 1.37	4.35 ± 0.47	5.14 ± 0.87	3.13 ± 0.66	3.94 ± 1.07
2 - 3	2.32 ± 0.30	6.40 ± 1.22	3.21 ± 0.36	11.0 ± 1.87	4.89 ± 0.57	6.87 ± 1.21	3.71 ± 0.33	6.16 ± 0.91
3 - 4	2.96 ± 0.30	8.29 ± 1.34	3.41 ± 0.39	13.3 ± 2.32	5.23 ± 0.60	9.53 ± 1.66	4.01 ± 0.33	6.81 ± 0.97
4 - 5	3.83 ± 0.70	11.8 ± 2.87	3.75 ± 0.40	13.5 ± 2.24	5.27 ± 0.62	11.1 ± 1.98	5.46 ± 0.59	10.8 ± 1.81
5 - 7	5.90 ± 0.73	32.8 ± 5.02	6.07 ± 0.66	43.5 ± 5.97	4.42 ± 0.46	18.4 ± 2.46	3.33 ± 0.33	13.5 ± 1.74
7 - 9	7.18 ± 1.15	47.1 ± 9.00	5.23 ± 0.54	46.4 ± 6.17	3.13 ± 0.35	15.8 ± 2.24	2.06 ± 0.22	10.6 ± 1.44
9 – 11	5.32 ± 0.50	41.6 ± 5.16	1.52 ± 0.20	23.0 ± 3.69	1.36 ± 0.17	8.65 ± 1.32	1.90 ± 0.21	8.53 ± 1.21
11 – 13	2.06 ± 0.35	18.5 ± 3.74	0.07 ± 0.03	0.75 ± 0.40	0.18 ± 0.04	1.08 ± 0.29	0.75 ± 0.13	4.01 ± 0.79
13 – 15	0.23 ± 0.06	2.43 ± 0.68	MDA	-	0.05 ± 0.02	0.38 ± 0.16	0.05 ± 0.03	0.32 ± 0.21
0 - 15		174±12.6		162 ± 10.2		79.7±4.70		69.7±3.81

^{a)} content of ^{239,240}Pu was calculated as activity of radionuclide in sample divided by sampling surface.

^{b)} activity concentration \pm uncertainty due to counting of sample at the 95% confidence level.

^{c)} content \pm uncertainty due to counting of sample and sampling at the 95% confidence level.

^{d)} total deposition ± combined uncertainty due to counting of sample and sampling of all layers at the 95% confidence level.

Table 4b. Plutonium-239,240 in layers of bottom sediments from the Gdansk Deep and Bornholm Deep, content of this radionuclide in particular layers and its total deposition.

	^{239,240} Pu							
Layer (cm)	P1/1999		P5/2	1998	P5/1999			
	Activity (Bq kg ⁻¹ d.w.)	Content ^{a)} (Bq m ⁻²)	Activity (Bq kg ⁻¹ d.w.)	Content (Bq m ⁻²)	Activity (Bq kg ⁻¹ d.w.)	Content (Bq m ⁻²)		
0 - 1	2.89±0.35 ^{b)}	4.11 ± 0.74^{c}	0.785 ± 0.17	1.12 ± 0.30	1.15 ± 0.17	2.25 ± 0.46		
1 - 2	2.76 ± 0.37	5.25 ± 1.02	0.905 ± 0.14	1.82 ± 0.40	1.32 ± 0.19	2.66 ± 0.55		
2 - 3	3.79 ± 0.43	7.85 ± 1.36	0.919 ± 0.49	2.05 ± 0.49	1.16 ± 0.13	2.65 ± 0.46		
3 - 4	3.57 ± 0.42	7.99 ± 1.43	1.01 ± 0.20	2.48 ± 0.64	1.15 ± 0.16	3.13 ± 0.65		
4 - 5	3.76 ± 0.42	9.38 ± 1.61	0.96 ± 0.24	2.35 ± 0.73	1.17 ± 0.13	3.28 ± 0.65		
5 - 7	3.23 ± 0.44	15.0 ± 2.48	0.86 ± 0.09	4.72 ± 0.66	0.82 ± 0.07	4.82 ± 0.89		
7 - 9	2.94 ± 0.35	14.7 ± 2.18	0.79 ± 0.13	4.13 ± 0.79	0.72 ± 0.10	4.62 ± 0.60		
9 – 11	2.93 ± 0.34	13.6 ± 1.97	0.81 ± 0.12	4.06 ± 0.72	0.60 ± 0.05	4.27 ± 0.85		
11 – 13	1.76 ± 0.22	9.57 ± 1.48	0.63 ± 0.11	3.17 ± 0.65	0.30 ± 0.04	1.85 ± 0.38		
13 – 15	1.23 ± 0.22	7.43 ± 1.56	0.40 ± 0.08	2.19 ± 0.51	0.14 ± 0.02	0.89 ± 0.26		
0 – 15		94.90 ± 5.24^{d}		28.10 ± 1.92		30.40 ± 1.91		

 a) content of ^{239,240}Pu was calculated as activity of radionuclide in sample divided by sampling surface.

^{b)} activity concentration \pm uncertainty due to counting of sample at the 95% confidence level. ^{c)} content \pm uncertainty due to counting of sample and sampling at the 95% confidence level. ^{d)} total deposition \pm combined uncertainty due to counting of sample and sampling of all layers at the 95% confidence level.



Fig. 3. Vertical distribution of ²²⁶Ra in bottom sediments from the Gdansk Basin.

	²¹⁰ Pb (total)	²¹⁰ Pb (unsu	pported)	²¹⁰ Pb (total)	²¹⁰ Pb (unsup	ported)
Layer (cm)	Activity (Bq kg ⁻¹ d.w.)	Activity (Bq kg ⁻¹ d.w.)	Content ^{a)} (Bq m ⁻²)	Activity (Bq kg ⁻¹ d.w.)	Activity (Bq kg ⁻¹ d.w.)	Content (Bq m ⁻²)
	P110/1998				P110/2000	
$ \begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	$434 \pm 15.5^{\text{b}}$ 413 ± 15.6 216 ± 12.8	406 ± 15.7^{c}) 385 ± 15.8 205 ± 14.0	537 ± 53.0^{d} 839 ± 84.7	338 ± 12.1 289 ± 8.28 227 ± 7.22	310±12.5 264±8.59	357 ± 44.7 721 ± 66.7
2 - 3 3 - 4 4 - 5	310 ± 13.8 390 ± 14.4 323 ± 13.6	293 ± 14.0 362 ± 14.6 294 ± 13.8	812 ± 87.3 987 ± 100 906 ± 96.9	227 ± 7.33 206 ± 7.09 198 ± 6.96	202 ± 7.68 182 ± 7.48 173 ± 7.36	692 ± 67.8 709 ± 71.9 623 ± 63.8
5 – 7 7 – 9 9 – 11	267 ± 12.7 260 ± 12.6 162 ± 6.76	240±12.9 234±12.8 133±7.22	1330 ± 112 1530 ± 130 1040 ± 87.5	211 ± 7.16 141 ± 6.08 92.3 ± 5.07	185±7.59 117±6.51 69.0±5.56	1330 ± 94.2 1030 ± 88.7 1050 ± 116
11 – 13 13 – 15 15 – 17	108 ± 5.90 90.1 ± 5.43 65.8 ± 4.93	81.3±6.36 64.1±6.00 38.8±5.55	728 ± 78.8 665 ± 82.1 428 ± 74.1	56.7 ± 4.28 50.6 ± 4.15 44.5 ± 74.1	30.4 ± 4.91 24.8 ± 4.85 18.7 ± 4.65	316 ± 60.5 249 ± 56.1 188 ± 52.4
17 – 19 0 – 19	46.2±4.57	15.7±5.23	154 ± 56.2 9960 ± 310^{e}	40.3±3.94	14.2±4.67	139 ± 50.0 740 ± 249
	P116/1999				P1/1999	
$ \begin{array}{ccc} 0 - 1 \\ 1 - 2 \end{array} $	397 ± 13.3 467 ± 15.2	369 ± 13.5 440 ± 15.4	567 ± 54.7 559 + 53 1	349 ± 9.38 314 ± 8.69	312±9.85	443 ± 40.6 528+49.2
2 - 3 3 - 4	403 ± 14.4 300 ± 12.0	375 ± 14.6 270 ± 12.0	628 ± 62.0 458 ± 48.3	270 ± 8.12 284 ± 8.30	230 ± 8.70 246 ± 8.81	477±46.6 551±52.8
4 - 5 5 - 7 7 - 9	287 ± 12.4 215 ± 10.6 131 ± 5.66	257 ± 12.4 185 ± 10.6 101 ± 5.66	511 ± 55.8 756 ± 67.1 522 ± 47.8	255 ± 7.95 248 ± 7.89 218 ± 7.46	215 ± 8.55 211 ± 8.44 177 ± 8.09	537±53.5 975±68.4 888+67.1
9 – 11 11 – 13	118±5.57 94.1±5.32	88.8 ± 5.57 65.4 ± 5.23	398 ± 39.4 349 ± 41.1	210 ± 7.40 203 ± 7.32 160 ± 5.61	160 ± 8.03 119 ± 6.42	843±67.6 646±54.3
13 - 15 15 - 17 17 - 19	81.6±5.01 70.8±4.67 56.4±4.64	52.9 ± 5.01 41.9 ± 4.67 26.8 ± 4.64	317 ± 43.0 253 ± 39.5 162 ± 36.8	113 ± 5.80 102 ± 5.71 $85 3 \pm 5 39$	73.9 ± 6.55 62.4 ± 6.50 46.2 ± 6.20	448±53.2 383±51.4 297±48.8
0 – 19	JU.4±4.04	20.0 - 4.04	5480 ± 173	65.52	40.2±0.20	7020 ± 191

Table 5. Lead-210 (total and unsupported) in layers of bottom sediments from the Gdansk Basin, content of unsupported ²¹⁰Pb in particular layers and its total deposition.

^{a)} content of ²¹⁰Pb was calculated as activity of radionuclide in sample divided by sampling surface.

^{b)} activity concentration \pm uncertainty due to counting of sample at the 95% confidence level.

c) activity concentration \pm combined uncertainty due to counting of sample at the 95% confidence level.

^{d)} content \pm uncertainty due to counting of sample and sampling at the 95% confidence level.

e) total deposition ± combined uncertainty due to counting of sample and sampling of all layers at the 95% confidence level.



Fig. 4. Vertical distribution of ¹³⁷Cs in bottom sediments from the Gulf of Gdansk (P116).

Discussion

Decrease of ¹³⁷Cs concentration in bottom sediment proceeds very slowly. The ¹³⁷Cs concentrations are similar to these observed in 1990 (258–395 Bq kg⁻¹ d.w. in 0–3 cm layer). The patterns of ¹³⁷Cs vertical distribution in P116 core samples (Gulf of Gdansk) (Fig. 4) show that the maximum observed in the layer 0–1 cm in 1993 was moved to the 1–2 cm layer in 1999 and 2000 years. Concentrations of ¹³⁷Cs in bottom sediments from the Gdansk Basin decrease exponentially with depth and in the deeper layers of the cores the influence of global fallout contamination was still observed.

The main source of plutonium in the Southern Baltic Sea was global fallout. The highest concentrations of plutonium were observed always in deeper layers of the sediment cores. The ratios of ²³⁸Pu to ^{239,240}Pu in the majority of samples examined since 1991 ranged 0.03-0.05 [10, 11], being similar to the ratios found for the cumulative deposit from global fallout after the nuclear weapons tests [3]. The maximum of plutonium in core sample P110/2000 (Table 5) was found in 5-7 cm layer, however, in the year 1998 and the previous ones (1991-1997) this maximum was most frequently observed in the 7-9 cm layer. The sedimentation rate in this region, calculated from plutonium distribution and max. global fallout dated to 1963, ranged from 1.6 to 2.2 mm year⁻¹. The concentrations of ^{239,240}Pu in the core samples P116 and P1 in the layer 0-7 cm ranged from 3.13 to 5.46 Bq kg⁻¹ d.w. and 2.74 to 3.79 Bq kg⁻¹ d.w., respectively. Because the maxima of plutonium were not well marked, the sedimentation rate based on plutonium distribution was not calculated.

Dating by ²¹⁰Pb is another method, particularly in the environment with uniform sediment accumulation rates. In the aquatic environment ²¹⁰Pb originated mainly from atmospheric input and also *in situ* from decay of ²²⁶Ra [5]. Geochronology of sediments is based on the decay of the unsupported ²¹⁰Pb. The ²¹⁰Pb analysis in sediments has been carried out in the layers, which represent several years of deposition. Under the assumption of steady deposition of ²¹⁰Pb from atmosphere and an exponential decrease of

unsupported ²¹⁰Pb, the sedimentation rate was evaluated to be 1.8–1.9 mm year⁻¹ in P110 area. The unsupported ²¹⁰Pb concentrations were calculated as a difference between ²¹⁰Pb and ²²⁶Ra concentrations determined along the profiles. Estimated sedimentation rate in the P116 area was equal to 2.2 mm year⁻¹ and in the Gdansk Deep (P1) – 2.3 mm year⁻¹. These values are in good agreement with sedimentation rates, equal to 1.86–2.25 mm year⁻¹, calculated by Pempkowiak *et al.* [8].

In general, the patterns of vertical distributions of unsupported 210 Pb were similar to 137 Cs distributions (Table 3 and 5) and (Fig. 5). It is suggested that a continued sedimentation of 137 Cs and/or 210 Pb – containing matter can be responsible for such a distribution of these radionuclides in the bottom sediments. From the data of [13, 14] it can be seen that the Baltic Sea water still contains large amount of 137 Cs from the Chernobyl accident. In the patterns of vertical distribution of 239,240 Pu a maximum concentration was observed at a deeper layer of 5–7 cm (Fig. 5). The loss of soft surface sediments and slicing errors during sampling may significantly affect the result of sedimentation rate if the calculation is based on 239,240 Pu peaks. This could be the reason of wide range of sedimentation rate (1.6–2.2 mm year⁻¹) calculated from 239,240 Pu peaks in sediments from station P110.

Conclusions

- The highest concentrations of ¹³⁷Cs, originated from the fallout after the Chernobyl accident, were observed in upper 0–3 cm layer of sediment. The patterns of ¹³⁷Cs vertical distributions suggested that ¹³⁷Cs containing sedimentation matter are still in new-formed layers of the bottom sediments.
- The maxima of ^{239,240}Pu concentrations observed in deeper layers of sediments indicate that the global fallout was the main source of plutonium contamination.
- The sedimentation rates in the Gdansk Basin based on vertical distribution of ²¹⁰Pb and ^{239,240}Pu, ranged from 1.6 to 2.3 mm year⁻¹.



Fig. 5. Vertical distribution of ¹³⁷Cs, ²¹⁰Pb and ^{239,240}Pu concentrations in bottom sediments from the Gulf of Gdansk (P110) in 2000.

Acknowledgments Thanks are due to Professor Zofia Pietrzak-Flis from the Central Laboratory for Radiological Protection for remarks and comments on the manuscript. I also wish to thank Mr A. Adamczyk and Mr E. Chrzanowski for their competent technical assistance.

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