

ORIGINAL RESEARCH ARTICLE

Budget of ⁹⁰Sr in the Gulf of Gdańsk (southern Baltic Sea)

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KEYWORDS ⁹⁰Sr; Budget; Gulf of Gdańsk **Summary** In the period from 2005 to 2011 the major source of ⁹⁰Sr to the Gulf of Gdańsk was the Vistula river. Its contribution was 99.7% of the total load. The main processes responsible for the decrease in ⁹⁰Sr activity in the Gulf of Gdańsk were: radioactive decay (87%) and sediment deposition (13%). Average increase in the activity of ⁹⁰Sr in the Gulf of Gdańsk during the study period was 5.0% (114 GBq), which was almost 2 times higher than the loss of ⁹⁰Sr due to radioactive decay. In the years 1997–2015, the effective half-life of ¹³⁷Cs was 9.1 years and that of ⁹⁰Sr was 50.3 years. Assuming a further decrease in ¹³⁷Cs and maintaining ⁹⁰Sr concentrations at present level, it is expected that ⁹⁰Sr will become the major anthropogenic isotope having impact on the level of radioactivity in the Gulf of Gdańsk.

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1. Introduction

The Baltic Sea is an inland sea which is practically closed since the only connection with the North Sea and the Atlantic Ocean through the Danish Straits (Skagerrak and Kattegat between southern Sweden and the Danish islands) is relatively narrow. As a consequence, the water exchange between the Baltic Sea and North Sea is limited and amounts to 0.05% per year (Wängberg et al., 2001). This makes the Baltic Sea very sensitive to contamination with different pollutants (HELCOM, 2010), including radionuclides (IAEA, 2005). The Baltic Sea is still considered as one of the water bodies that is most polluted with ⁹⁰Sr and ¹³⁷Cs in the world

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(HELCOM, 2009; IAEA, 2005). The major sources of radionuclides inputs into the Baltic Sea were: atmospheric testing of nuclear weapons carried out during the late 1950s and early 1960s, the Chernobyl accident in 1986, discharges from nuclear reprocessing plants located outside the Baltic Sea (Sellafield and La Hague) and discharges from nuclear facilities in the Baltic Sea drainage area (Baklanov and Sorensen, 2001; HELCOM, 2009; Nielsen et al., 1999; Nies et al., 1995). It has been estimated that the total load of ⁹⁰Sr and ¹³⁷Cs introduced into the Baltic Sea from all sources amounts to 621 TBq and 5752 TBq, respectively.

Among many pollutants introduced into the Baltic Sea, the ⁹⁰Sr isotope is considered to be particularly dangerous, due to its specific nature and relatively long half-life (28.8 years) (Kryshev, 2006). Its chemical similarity to calcium is the reason why strontium is quite easily taken up and accumulated in a body, especially in bone tissues. However, data on the activity levels of $^{90}\mathrm{Sr}$ in various compartments of the marine environment and biota is largely limited. At the same time knowledge on the ⁹⁰Sr levels in particular elements of the marine environment and knowledge on factors controlling its temporal and spatial distribution, are crucial for determining the degree of environmental contamination, especially in incidental situations. Information on the concentration factors in organisms is of special importance as allows to determine the exposure of organisms to the radioactivity related to ⁹⁰Sr presence.

Long-term observations of the variability in concentrations of ⁹⁰Sr and ¹³⁷Cs showed that the decrease of ⁹⁰Sr levels is not as significant as in the case of ¹³⁷Cs. Moreover, the decrease is smaller than expected from the radioactive decay. Therefore, based on the analysis of data on radionuclide concentrations in various components of the marine environment (abiotic - seawater and sediment, and biotic fish and macrophytobenthic plants), obtained in the period of 2005-2011, the study, which results would become the basis for future scenarios concerning ⁹⁰Sr levels in the Baltic Sea, was undertaken. The assessment of the present level of ⁹⁰Sr pollution in the Baltic Sea was carried out in relation to current sources - riverine and atmospheric inputs of this isotope, based on the results of ⁹⁰Sr concentrations in the Vistula and atmospheric deposition, obtained for the same period as mentioned above. Finally, the main factors controlling distribution of the ⁹⁰Sr in the marine ecosystem were indicated, also in relation to long-term changes observed after potential introduction of significant ⁹⁰Sr loads into the Baltic Sea.

2. Material and methods

2.1. Study area

The Gulf of Gdańsk is located in the southeastern part of the Baltic Sea. Its northern boundary is the straight line connecting Cape Rozewie ($54^{\circ}50'N$, $18^{\circ}20'E$) with Cape Taran ($54^{\circ}58'N$, $19^{\circ}59'E$). The area of the Gulf of Gdańsk is 4940 km² (Łukawska-Matuszewska and Bolałek, 2008), while the volume of water is estimated at 291.2 km³ (Majewski, 1990). The Gulf of Gdańsk has an average depth of about 50 m, and a maximum of 118 m.

2.2. Methods of budget calculations

In order to balance the loads of 90 Sr in the Gulf of Gdańsk, it was assumed that the main sources of this isotope are: atmospheric deposition and Vistula river waters, while the factors having impact on the decrease of 90 Sr concentration in seawater are: radioactive decay, bioaccumulation and sedimentation processes (Fig. 1). The bioaccumulation took into account in the calculations was related only to marine plants and fish. The loads of 90 Sr were calculated based on literature data (mainly from own research) concerning concentrations of this isotope in particular components of the marine environment, measured in the period of 2005–2011, adopted for the estimation of the 90 Sr budget in the Gulf of Gdańsk.

Data on ⁹⁰Sr loads introduced into the Gulf of Gdańsk with atmospheric deposition and riverine runoff was obtained from the study by Saniewski and Zalewska (2016).

The concentrations of ⁹⁰Sr in seawater were measured in samples collected between 2005 and 2011 at five stations located in the Gulf of Gdańsk. The samples were obtained from the sea surface, from the bottom, and additionally along vertical profiles (every 20 m) at two stations (Fig. 1, Table 1). The analysis of ⁹⁰Sr distribution in seawater of the Gulf of Gdańsk, for the abovementioned study period, was presented in the work by Saniewski (2013).

Mean activity of 90 Sr in sediments was calculated based on literature data (Zalewska and Suplińska, 2013) and unpublished own data. The bottom areas with intensive sedimentation processes, associated with the transportation type of bottom (LOI – loss on ignition, values of 4–10%) and the accumulation bottom (with LOI values >10%) (Håkanson et al., 2003) account for respectively 1426 km² and 1840 km² of the Gulf of Gdańsk (Carman and Cederwall, 2001). Therefore, it was assumed that the 90 Sr deposition into sediments is most intensive in the area of 3266 km² (Fig. 1).

Since ⁹⁰Sr is bioaccumulated and biomagnified in the trophic chain, the concentrations of ⁹⁰Sr in macrophytobentic plants and selected fish species specific to the Gulf of Gdańsk were taken into account in the budget calculations. To estimate the load of ⁹⁰Sr removed with the caught fish, the average activity values of ⁹⁰Sr in fish species (Zalewska et al., 2016) and the mass of fish caught in 2005–2011 in the study area (Szostak et al., 2006, 2007, 2008, 2009, 2010, 2011, 2012) were used.

The estimation of ⁹⁰Sr accumulated in marine plants was carried out using data on ⁹⁰Sr concentrations in selected species of macrophytobenthic plants (Zalewska, 2015). The amount of biomass having the potential for bioaccumulation was assessed on the basis of data collected during macrophytobenthos monitoring campaigns carried out in two locations: Orłowo Cliff and Kuźnica Hollow (Brzeska and Saniewski, 2012). The sampling for monitoring purposes took place in June, i.e. in the period of intensive primary production and rapid growth of both macroalgae and vascular plants. Samples were taken by a diver, along transects, from a depth of 1 m to a maximum depth of plant occurrence. The plant material was collected from the area determined by a randomly placed frame $(0.5 \text{ m} \times 0.5 \text{ m})$. The frame was placed three times at each depth. The collected material was analyzed macroscopically and microscopically to sepa-



Figure 1 Location of sampling stations in the Gulf of Gdańsk.

Station	Sampling depth [m]	Activity con	Activity concentration of ⁹⁰ Sr [Bq m ⁻³]								
		2005	2006	2007	2008	2009	2010	2011			
ZN2	0	$\textbf{5.6} \pm \textbf{0.5}$	$\textbf{9.2}\pm\textbf{0.5}$	$\textbf{5.6} \pm \textbf{0.5}$	$\textbf{4.2}\pm\textbf{0.4}$	$\textbf{3.8}\pm\textbf{0.4}$	$\textbf{6.4} \pm \textbf{0.5}$	7.6 ± 0.5			
	12	$\textbf{8.7}\pm\textbf{0.6}$	$\textbf{8.8} \pm \textbf{0.5}$	$\textbf{6.7} \pm \textbf{0.5}$	$\textbf{5.4} \pm \textbf{0.6}$	$\textbf{6.3} \pm \textbf{0.4}$	$\textbf{9.8} \pm \textbf{0.6}$	$\textbf{4.6} \pm \textbf{0.4}$			
P1	0	$\textbf{8.6} \pm \textbf{0.5}$	$\textbf{9.9}\pm\textbf{0.6}$	$\textbf{8.3} \pm \textbf{0.5}$	$\textbf{7.6} \pm \textbf{0.9}$	$\textbf{7.7} \pm \textbf{0.6}$	$\textbf{8.8} \pm \textbf{0.5}$	$\textbf{5.9} \pm \textbf{0.5}$			
	20	$\textbf{8.2}\pm\textbf{0.5}$	$\textbf{9.8}\pm\textbf{0.7}$	$\textbf{8.5}\pm\textbf{0.6}$	$\textbf{7.9} \pm \textbf{0.5}$	$\textbf{7.1} \pm \textbf{0.4}$	$\textbf{8.4} \pm \textbf{0.4}$	$\textbf{4.3} \pm \textbf{0.6}$			
	40	$\textbf{9.5}\pm\textbf{0.6}$	$\textbf{8.8}\pm\textbf{0.7}$	$\textbf{7.5} \pm \textbf{0.4}$	$\textbf{7.8} \pm \textbf{0.4}$	$\textbf{7.4} \pm \textbf{0.4}$	$\textbf{7.6} \pm \textbf{0.5}$	$\textbf{9.3}\pm\textbf{0.9}$			
	60	$\textbf{9.2}\pm\textbf{0.6}$	$\textbf{6.9} \pm \textbf{0.6}$	$\textbf{9.9}\pm\textbf{0.5}$	$\textbf{7.2} \pm \textbf{0.6}$	$\textbf{8.2}\pm\textbf{0.4}$	$\textbf{8.1}\pm\textbf{0.5}$	$\textbf{7.9} \pm \textbf{0.5}$			
	80	$\textbf{8.7}\pm\textbf{0.6}$	$\textbf{8.6} \pm \textbf{1.2}$	$\textbf{6.6} \pm \textbf{0.6}$	$\textbf{8.2}\pm\textbf{0.7}$	$\textbf{9.2}\pm\textbf{0.4}$	$\textbf{8.0} \pm \textbf{0.6}$	$\textbf{7.9} \pm \textbf{0.4}$			
	105	$\textbf{7.8} \pm \textbf{0.5}$	$\textbf{9.8} \pm \textbf{0.6}$	$\textbf{6.9} \pm \textbf{0.4}$	$\textbf{5.4} \pm \textbf{0.6}$	$\textbf{7.6} \pm \textbf{0.4}$	$\textbf{6.9} \pm \textbf{0.5}$	$\textbf{6.9} \pm \textbf{0.4}$			
P116	0	$\textbf{7.4} \pm \textbf{0.6}$	$\textbf{8.9}\pm\textbf{0.5}$	$\textbf{6.9} \pm \textbf{0.5}$	$\textbf{7.6} \pm \textbf{0.8}$	$\textbf{8.7} \pm \textbf{0.4}$	$\textbf{8.6} \pm \textbf{0.4}$	$\textbf{5.0} \pm \textbf{0.4}$			
	85	$\textbf{8.4}\pm\textbf{0.5}$	$\textbf{10.0} \pm \textbf{0.8}$	7.7 ± 0.5	$\textbf{6.4} \pm \textbf{0.5}$	$\textbf{6.5} \pm \textbf{0.4}$	$\textbf{8.2}\pm\textbf{0.4}$	$\textbf{9.3}\pm\textbf{0.5}$			
P110	0	$\textbf{7.7} \pm \textbf{0.7}$	$\textbf{8.1}\pm\textbf{0.8}$	$\textbf{8.5}\pm\textbf{0.4}$	$\textbf{6.5} \pm \textbf{0.5}$	$\textbf{7.5} \pm \textbf{0.5}$	$\textbf{7.0} \pm \textbf{0.5}$	$\textbf{7.0} \pm \textbf{0.5}$			
	69	$\textbf{8.6} \pm \textbf{0.6}$	$\textbf{9.4} \pm \textbf{1.0}$	$\textbf{7.8} \pm \textbf{0.4}$	$\textbf{4.5} \pm \textbf{0.5}$	$\textbf{7.1} \pm \textbf{0.5}$	$\textbf{8.6} \pm \textbf{0.4}$	$\textbf{6.6} \pm \textbf{0.5}$			
ZN4	0	$\textbf{7.7} \pm \textbf{0.5}$	$\textbf{9.9}\pm\textbf{0.5}$	$\textbf{9.3} \pm \textbf{1.1}$	$\textbf{5.9} \pm \textbf{0.4}$	$\textbf{8.6} \pm \textbf{0.4}$	$\textbf{6.7} \pm \textbf{0.5}$	$\textbf{9.6}\pm\textbf{0.5}$			
	20	$\textbf{8.1}\pm\textbf{0.8}$	$\textbf{10.1} \pm \textbf{0.6}$	$\textbf{9.9}\pm\textbf{0.8}$	$\textbf{6.4} \pm \textbf{0.5}$	$\textbf{8.5}\pm\textbf{0.6}$	$\textbf{6.5} \pm \textbf{0.5}$	$\textbf{9.4}\pm\textbf{0.5}$			
	40	$\textbf{8.7}\pm\textbf{0.8}$	$\textbf{10.6} \pm \textbf{0.5}$	$\textbf{10.3} \pm \textbf{0.8}$	$\textbf{5.5} \pm \textbf{0.4}$	$\textbf{7.9} \pm \textbf{0.6}$	$\textbf{9.5}\pm\textbf{0.5}$	$\textbf{8.8}\pm\textbf{0.5}$			
	60	$\textbf{10.0} \pm \textbf{0.7}$	$\textbf{8.9}\pm\textbf{0.7}$	$\textbf{8.0}\pm\textbf{0.7}$	$\textbf{5.2} \pm \textbf{0.6}$	$\textbf{8.7} \pm \textbf{0.5}$	$\textbf{8.9}\pm\textbf{0.5}$	7.7 ± 0.5			
	69	$\textbf{11.7} \pm \textbf{0.7}$	$\textbf{9.0} \pm \textbf{0.9}$	$\textbf{9.1}\pm\textbf{0.8}$	$\textbf{4.4} \pm \textbf{0.5}$	$\textbf{6.5} \pm \textbf{0.6}$	$\textbf{8.1} \pm \textbf{0.4}$	$\textbf{8.6} \pm \textbf{0.5}$			
Average c	concentration [Bq m $^{-3}$]	8.5	9.2	8.1	6.2	7.5	8.0	7.4			

Table 1	Activity concentrations	of ⁹⁰ Sr in seawater	of the Gulf of	Gdańsk in 2005–2011.
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rate individual species which biomass was measured and converted into 1 m^2 of bottom area. In order to determine the biomass in the Gulf of Gdańsk, the total surface area of the occurrence of macrophytobenthic plants in the investi-

gated research areas was estimated and multiply by biomass specific to 1 m^2 (Fig. 1).

It was assumed that the exchange of ⁹⁰Sr in seawater at the northern boundary of the Gulf of Gdańsk is negligible as

⁹⁰Sr concentrations in seawater are uniform in different areas of the southern Baltic Sea.

The load of ⁹⁰Sr in seawater of the Gulf of Gdańsk determined for each year served as "a starting point" for calculations. The inflowing loads were given the positive values, while the loads removed from the environment as a result of the discussed processes were given the negative values.

3. Results and discussion

3.1. Load of ⁹⁰Sr in seawater

In the period from 2005 to 2011, the concentrations of ⁹⁰Sr in seawater of the Gulf of Gdańsk were at the similar levels (Table 1). The mean concentrations varied in a very narrow range, from 7.4 Bq m⁻³ in 2011 to 9.2 Bq m⁻³ in 2006. The exceptional year was 2008, when the average activity was 6.2 Bg m⁻³, and this was due to the fact that concentrations in that year ranged from 4.2 Bg m⁻³ to 8.2 Bg m⁻³. The lowest value was reported at the mouth of the Vistula. This is a common feature, also observed in other years, resulting from a slightly diluting influence of the Vistula waters. In 2009, the concentration at the mouth of the Vistula river was only 3.8 Bq m^{-3} , however, in 2006 the value in that area reached 9.2 Bq m^{-3} . Such variability can be associated with the wind pattern preceding the sampling period. During the study period, the maximum concentrations of ⁹⁰Sr, at the level of 10 Bq m⁻³ and higher were observed in the vicinity of the Hel Peninsula (Table 1). For the ⁹⁰S loads calculations in seawater, there were taken the average values of ⁹⁰Sr activities in seawater determined for particular years and the volume of seawater in the Gulf of Gdańsk. Calculated loads varied from 1817 GBg in 2008 to 2684 GBg in 2006 (Table 2). The standard deviation of the loads deposited in the Gulf of Gdańsk over the period of 2005-2011 did not exceed 15% (Saniewski, 2013).

3.2. Load of ⁹⁰Sr from atmospheric deposition and riverine input

The total loads of ⁹⁰Sr and ¹³⁷Cs from both sources: atmospheric deposition and riverine input, reaching the Gulf of Gdańsk between 2005 and 2011, were 1238 GBg and 450 GBg, respectively (Saniewski and Zalewska, 2016). The major source of both radionuclides to the Gulf of Gdańsk was the Vistula river; its contribution reached 99.7% in the case of 90 Sr and 95.8% as regarding 137 Cs. In the years 2006–2010, the riverine load of 137 Cs (431 GBq) was nearly three times smaller than that of 90 Sr (1234 GBq), although the average activity of ¹³⁷Cs in the surface soil laver in the Vistula river drainage area was about three times higher than the activity of ⁹⁰Sr (Solecki, 2006). The main reason for this fact was the migration of both isotopes in the terrestrial environment. The Sr^{2+} cations are mobile and more easily washed out by water to the rivers than the Cs⁺ ions which are strongly absorbed on soil particles limiting its movement by chemical and biological processes (Ritchie and McHenry, 1990). The environmental mobility of ⁹⁰Sr is approximately one order of magnitude greater than that of ¹³⁷Cs and the runoff coefficient (transfer of radioactivity from catchments to the sea surface) for ⁹⁰Sr is much greater than that for ¹³⁷Cs (Cross et al., 2002). It was estimated that after the Chernobyl power plant disaster, the maximum deposition of ⁹⁰Sr in Poland was 1 kBg m^{-2} , and after the nuclear weapons testing it amounted to, on average, 3.2 kBq m^{-2} . As a result, in 1986 the deposition still remained at a level of approximately 1.5 kBg m⁻² (UNSCEAR, 1982). In 1999, the average activity of ⁹⁰Sr in soil of the middle part of the Vistula drainage basin was 26.15 Bq kg⁻¹, the values ranged from 5.3 Bq kg⁻¹ to 85.3 Bg kg⁻¹ (Solecki and Chibowski, 2001). Taking into account published results (Solecki, 2006; Solecki and Chibowski, 2001) and the surface area of the Vistula drainage basin, it was calculated that in 1999 about 0.67 PBg of ⁹⁰Sr was deposited in the 0-10 cm soil layer, which could have

Table 2	Budget of	⁹⁰ Sr in the	e Gulf of	Gdańsk in	years 2005-2011.
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Years	2005	2006	2007	2008	2009	2010	2011
Total loads in seawater in the Gulf of Gdańsk [GBq]	2477	2684	2355	1817	2181	2331	2165
Loads from Vistula River [GBq]	+122	+137	+137	+118	+175	+316	+228
Loads from atmospheric deposition [GB]	+0.58	+0.53	+0.49	+0.64	+0.41	+0.49	+0.87
Radioactive decay [GBq]	-59	-64	-56	-43	-52	-55	-52
Loads deposited in sediments [GBq]	-7.45	-8.55	-7.48	-8.29	арр. —8.00	арр. —8.00	арр. —8.00
Loads bioaccumulated in	app.	app.	app.	app.	app.	app.	-0.027
macrophytobentic plants [GBq]	-0.027	-0.027	-0.027	-0.027	-0.027	-0.027	
Loads in caught fish [GBq]	-0.0029	-0.0028	-0.0023	-0.0019	-0.0032	-0.0030	-0.0028
Increase of loads in seawater in the Gulf of Gdańsk [GBq]	56.4 (2.3%)	65.1 (2.4%)	74.3 (3.2%)	67.2 (3.7%)	115.1 (5.3%)	253.4 (10.9%)	169.7 (7.8%)

acted as a potential source of this isotope washed out by water to the river.

The lowest 90 Sr load of 118 GBq was introduced into the Gulf of Gdańsk with waters of the Vistula in 2008 (Table 2). In the period between 2005 and 2007, the load was in the range from 122 to 137 GBq, while in 2011 it reached 228 GBq. However, the highest load of 316 GBq was introduced in 2010, due to the flood event in the Vistula river drainage basin.

In the case of atmospheric deposition, the highest load of 90 Sr (0.87 GBq) was introduced to the Gulf of Gdańsk in 2011, as a result of the Fukushima Dai-ichi nuclear power plant disaster, which took place in March that year. The contaminated air masses were transported towards the area of Europe within about a month (Table 2). In other years, the loads remained in the range from 0.41 GBq in 2009 to 0.64 GBq in 2008. The total load of 90 Sr introduced to the Gulf of Gdańsk in the period of 2005–2011 amounted to 4 GBq, and was almost five times lower than the load of 137 Cs (Saniewski and Zalewska, 2016).

3.3. Radioactive decay

The correction for radioactive decay of 90 Sr was calculated based on the total load in seawater estimated for each year, taking into account the half-life value of 28.8 years. The obtained results ranged from 43 GBq in 2008 to 64 GBq in 2006 (Table 2).

3.4. Load of ⁹⁰Sr in sediments

The load of ⁹⁰Sr deposited in bottom sediments of the Gulf of Gdańsk between 2005 and 2008 was determined based on literature data concerning concentrations of ⁹⁰Sr in sediments (Zalewska and Suplińska, 2013; unpublished data) and taking into account the estimated area of sediments where undisturbed sedimentation occurs. Average concentrations of ⁹⁰Sr in 19-cm profiles of bottom sediments collected at stations P110, P116 and P1 varied in a relatively narrow range, from about 2-4 Bq kg⁻¹ d.w. The average values of ⁹⁰Sr concentration determined for each year (Table 3) were used to calculate the loads deposited in bottom sediments during one year. It was assumed that the average linear sedimentation rate specific to the Gulf of Gdańsk area is c.a.2 mm per year (Zalewska et al., 2015). The average mass of a 1-cm thick sediment layer taken from the area of 19.63 cm² amounts to 7 g (unpublished data). The load of ⁹⁰Sr deposited every year in the topmost sediment

Table 3 Average activity concentrations of 90 Sr in sediments in the Gulf of Gdańsk in years 2005–2008, based on Zalewska and Suplińska (2013).

Year	Activity of	⁹⁰ Sr [Bq kg	Mean activity of ⁹⁰ Sr	
	P110	P116 P1		$[Bq kg^{-1} d.w.]$
2005	2.73	3.49	3.38	3.20
2006	2.4	4.25	4.36	3.67
2007	4.25	2.29	3.1	3.21
2008	3.49	4.03	3.16	3.56

layer of the average thickness of 2 mm was determined on the basis of the following equation:

$$\mathsf{TLS} = \mathsf{A} * \mathsf{LAR} * \frac{\mathsf{W}}{\mathsf{S}_{\mathsf{N}}} * \mathsf{S},$$

where TLS – total load in sediment [Bq], A – mean activity of ⁹⁰Sr in sediments [Bq kg⁻¹], LAR – linear accumulation rate [cm], W – average mass of a single 1-cm sediment layer collected from the surface area determined by the Niemistö corer [kg], S_N – surface area determined by the Niemistö corer [m²], S – surface of sediment accumulation in the Gulf of Gdańsk [m²].

Since the concentrations of 90 Sr in sediment layers collected in 2005–2008 were very similar (range: 3.20–3.61 Bq kg⁻¹ d.w.), the values of 90 Sr loads were also comparable, from 7.45 GBq in 2005 to 8.55 in the following year (Table 2). Based on the results from the period of 2009–2011, the average value which we adopted for further calculations was 8 GBq.

3.5. Load of ⁹⁰Sr in macrophytobentic plants

The calculations of ⁹⁰Sr loads accumulated in macrophytobenthic plants were carried out based on the research results from 2011 (Saniewski and Zalewska, 2017; Zalewska, 2015). Samples of macrophytobenthic plants were obtained from two locations. In the area of the Orlowo Cliff, 13 species were recognized, including 12 taxa of macroalgae and 1 species of a vascular plant (Zostera marina). Macroalgae were most commonly represented by green algae -5 taxa, and red algae -5 taxa; only 2 species were identified as brown algae. Along the transect in the Kuźnica Hollow, 14 species of macrophytobenthos were identified in 2011, including 4 taxa of green algae, 2 taxa of brown algae and also 2 taxa of red algae, as well as 6 species of vascular plants. Based on the analysis of ⁹⁰Sr in particular species, the average concentrations for individual groups collected in two investigated locations were determined. The lowest concentrations of 90 Sr were found in green algae (1.36 Bq kg⁻¹ d.w.), while the highest were measured in the case of red algae (6.58 Bq kg⁻¹ d.w.) (Table 4). For each group of the macrophytobenthic plants, the biomass per unit area (1 m²) was determined. Then, taking into account the bottom areas where macrophytobenthic plants commonly apear (Fig. 1), namely the Puck Bay within a depth range of 0-4 m (98.9541 km²) and the Orlowo Cliff area, depth range of 0-7 m (0.98 km²), the average biomass of macrophytobenthos in the Gulf of Gdańsk was estimated. On this basis, the load of ⁹⁰Sr bioaccumulated in plant tissues of the particular groups was calculated. The lowest ⁹⁰Sr load (7.5 e-07 GBg) was attributed to the vascular plants from the Orlowo Cliff, while the highest load (1.8 e-02 GBg) was calculated for the brown algae in the Kuźnica Hollow. The estimated total load of ⁹⁰Sr bioaccumulated in all the macrophytobenthic plants in 2011 amounted to 0.027 GBq.

3.6. Load of ⁹⁰Sr in caught fish

The average activity value of ⁹⁰Sr in fish caught in the southern Baltic did not reveal any decreasing trend over the study period, and remained at relatively similar level, from

Table 4	Average activity of S	⁰ Sr and calculated	loads ac	cumulated in the ma	crophytobenthic	plants based on data	a from 2011.
	Phylum		Riomass	Average activity	Surface [km ²]	Total biomass [kg]	Load [GBa]

	Phylum	Biomass [kg m ²]	Average activity of ⁹⁰ Sr [Bq kg ⁻¹]	Surface [km²]	Total biomass [kg]	Load [GBq]
Puck Bay	Green algae (Chlorophyta)	0.000668	1.36	98.95	6.61 E+04	9.0 E-05
	Brown algae (<i>Phaeophyta</i>)	0.04186	4.37	98.95	4.14 E+06	1.8 E-02
	Red algae (Rhodophyta)	0.005391	5.47	98.95	5.33 E+05	2.9 E-03
	Vascular plant (Spermatophyta)	0.012497	4.56	98.95	1.24 E+06	5.6 E-03
Orłowo Cliff	Green algae (Chlorophyta)	0.03092	1.36	0.98	3.03 E+04	4.1 E-05
	Brown algae (<i>Phaeophyta</i>)	0.00769	6.58	0.98	7.54 E+03	5.0 E-05
	Red algae (<i>Rhodophyta</i>)	0.02156	5.32	0.98	2.11 E+04	1.1 E-04
	Vascular plant (Spermatophyta)	0.00019	4.06	0.98	1.86 E+02	7.6 E-07

Table 5 Average activity concentration of 90 Sr and load accumulated in caught fish in the Gulf of Gdańsk in 2005–2011, based on Zalewska et al. (2016).

	Average activity of ⁹⁰ Sr [Bq kg ⁻¹ w.w.] ^a	Load [GBq]					
		2005	2006	2007	2008	2009	2010	2011
(Clupea harengus)	$\textbf{0.027} \pm \textbf{0.004}$	1.2 E-04	8.3 E-05	1.3 E-04	1.2 E-04	1.5 E-04	1.8 E-04	2.1 E-04
Flounder (<i>Platichtys flesus</i>)	$\textbf{0.163} \pm \textbf{0.017}$	2.9 E-04	2.7 E-04	3.0 E-04	2.4 E-04	2.4 E-04	2.8 E-04	2.4 E-04
Cod (Gadus morhua)	$\textbf{0.186} \pm \textbf{0.020}$	7.5 E-04	1.0 E-03	5.2 E-04	6.5 E-04	7.5 E-04	9.6 E-04	8.8 E-04
Sprat (Spratus spratus)	$\textbf{0.037} \pm \textbf{0.005}$	1.8 E-03	1.4 E-03	1.4 E-03	9.3 E-04	2.1 E-03	1.6 E-03	1.4 E-03
^a Data from Zalewska et	t al. (2016).							

 0.027 Bg kg^{-1} w.w. for herring to 0.186 Bg kg^{-1} w.w. in the case of cod (Table 5) (Zalewska et al., 2016). From among over 25 species of fish caught in the area of the Gulf of Gdańsk, the dominant in terms of quantity were: sprat, herring, cod and flounder. In the period between 2005 and 2011, the average contribution of these species was 98.1% of 52 796 tons of fish caught annually (Szostak et al., 2006, 2007, 2008, 2009, 2010, 2011, 2012). On average, in the years 2005–2011, the contribution of individual fish species was: 76.6% for the sprat, 10.0% for the herring, 8.2% for the cod and 3.2% for the flounder. On the basis of data on the mass of fish species caught in particular years and the values of ⁹⁰Sr concentrations in particular fish species, the total loads of the examined isotope removed from the Gulf of Gdańsk as a result of fishery activities were estimated. The total load of ⁹⁰Sr for all fish species ranged from 0.0019 GBg in 2008 to 0.0032 GBg in 2009 (Table 2).

3.7. Budget of ⁹⁰Sr in the Gulf of Gdańsk

The basis for the 90 Sr budget calculation was estimation of the loads of this isotope in seawater, for particular years in the period from 2005 to 2008. Then, the loads of 90 Sr related to atmospheric deposition and riverine input, responsible for the increase in the amount of this isotope in seawater were analyzed. Subsequently the loads accumulated in various elements of the marine environment were estimated (Table 2). During the entire study period, the processes such as radioactive decay of 90 Sr, sediment deposition, and bioaccumulation in macrophytobenthic plants and fish species, taking place in the Gulf of Gdańsk, were compensated by a significantly higher input of 90 Sr with the Vistula waters

(Table 2). Due to the fact that the load of ⁹⁰Sr bioaccumulated in fish and plant tissues is very small, and additionally some amount of this isotope may return from plant tissues to the environment as a consequence of plant decomposition (the scale of this process is unknown), and also having in mind the fact that in the case of fish, the removed load of ⁹⁰Sr only applied to the caught fish and not to the fish resources present in the Gulf of Gdańsk, it was decided to exclude these values from the budget.

In the period 2005-2011, a theoretical increase in the load of ⁹⁰Sr in seawater of the Gulf of Gdańsk was demonstrated. The smallest increase of 56.4 GBq, which accounted for 2.3%, was calculated for 2005. A comparable increase of approximately 2.4% (65.1 GBq) occurred in the following year. It the years 2007 and 2008, it remained at 74.3 GBq and 67.2 GBg, which accounted for 3.2% and 3.7%, respectively. The largest increase in the 90Sr load, linked to the increased riverine water input caused by the flood event, was observed in 2010. It amounted to 253.4 GBq. The average increase in the activity of ⁹⁰Sr in the Gulf of Gdańsk in the period from 2005 to 2011 was 5.0% (114 GBg), which was almost 2 times higher than the loss of ⁹⁰Sr due to radioactive decay. In the forecasting of further variations in ⁹⁰Sr concentration in seawater of the Gulf of Gdańsk, the equalization of concentrations between the study area and open sea waters should be assumed.

The obtained results pointed to the cause of the lack of a clear decreasing trend related to ⁹⁰Sr concentration. Assuming that such a set of factors as discussed above, influencing the increase and decrease of ⁹⁰Sr concentrations in the Gulf of Gdańsk was maintained, it is expected that ⁹⁰Sr will become the major anthropogenic isotope determining the



Figure 2 Average activity of ⁹⁰Sr and ¹³⁷Cs in the Gulf of Gdańsk in 1984–2015.

level of radioactivity. As observed based on the analysis of average ⁹⁰Sr concentration in seawater of the southern Baltic, the concentration of this isotope reached a maximum value of 23 Bg m^{-3} after the Chernobyl power plant accident (Fig. 2). In the period from 1988 to 1996 the concentration of 90 Sr ranged from 14.2 to 18.9 Bq m⁻³, whereas a significant decrease to the value of 9.2 Bq m⁻³ occurred in 1996 and 1997. Between 1998 and 2016 the concentration was stabilized, and varied within a range from 5.6 to 8.7 Bg m $^{-3}$. In the similar period, since 1991 until now, there has been an apparent decrease in ¹³⁷Cs concentration. This direction of changes was also visible based on the ¹³⁷Cs to ⁹⁰Sr ratio in seawater of the Gulf of Gdańsk and the southern Baltic. Before the accident in the Chernobyl nuclear power plant in 1986, this ratio had been 0.8 (Majewski, 1990). Just after the accident it increased to 3.7 and further growth was observed in subsequent years (Majewski, 1990), mainly as a result of the increase in ¹³⁷Cs due to the inflow of more contaminated waters from the northern Baltic Sea and also due to a significant input with riverine waters. In 1991, the concentration of ¹³⁷Cs reached its maximum value of 101 Bg m^{-3} , which was eight times higher than concentration of ⁹⁰Sr. In the period covered by this study, the abovementioned ratio of both isotopes decreased slightly from 5.1 to 3.7 and it is currently at a similar level. However, assuming a further decrease of ¹³⁷Cs and maintaining ⁹⁰Sr concentrations at present level, the alignment f activity levels is expected in 2022 (Fig. 2). In addition, there were determined the effective half-lifes for these two isotopes in seawater of the southern Baltic Sea. Based on data form the period 1997-2015, they amounted to only 9.1 years in the case of ¹³⁷Cs and 50.3 years in the case of ⁹⁰Sr, which is almost twice longer than the theoretical half-life of ⁹⁰Sr. It could be assumed with high probability that approximately 35 years after the Chernobyl disaster, the activity of ⁹⁰Sr will have exceeded that of ¹³⁷Cs, and the ⁹⁰Sr will have become the main isotope having substantial impact on the anthropogenic radioactivity in the southern part of the Baltic Sea.

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