^{137}Cs activity distribution in the Lithuanian coastal waters of the Baltic Sea*

OCEANOLOGIA, 49 (1), 2007.
pp. 71–90.

KEYWORDS
Radiocaesium
Activity concentration
Specific activity
Baltic Sea
Curonian Lagoon
Hydrodynamic model

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Received 15 September 2006, revised 6 November 2006, accepted 4 December 2006.

Abstract

The main ^{137}Cs accumulation zone in the study area was found to be located at depths below the 50-metre isobath, i.e. below the layer of hydrodynamic activity. In coastal waters not influenced by the fresh water discharge from the Curonian Lagoon, ^{137}Cs occurs mostly in soluble form. The particulate ^{137}Cs activity concentration in the marine area affected by Curonian Lagoon water can make up 10% of the total ^{137}Cs activity concentration. The circulation model was developed to assess the distribution of artificial radionuclides in Lithuanian territorial waters. The model was validated on the basis of data acquired during the measurement

* This research was partly supported by the IAEA projects 'Assessment of radionuclide migration in the Lithuanian part of the Baltic Sea environment', No LIT, 2/002 (1999 –2000), and 'Assessment of radionuclide migration in the Lithuanian part of the Baltic Sea and Curonian Lagoon', No LIT, 7/002 (2001–2002), and also by the Lithuanian State Science and Studies Foundation T-04181.

campaign in the Lithuanian part of the Baltic Sea and the Curonian Lagoon in the years 1999–2001. The model enables the ^{137}Cs activity concentration to be simulated as a passive admixture (error within c. 15%).

1. Introduction

There are several reasons for the particular interest in ^{137}Cs activity concentration in the Baltic Sea off the Lithuanian coast. Firstly, measurements of ^{137}Cs activity concentration in these waters have revealed an inhomogeneous distribution and remarkable fluctuations of concentration depending on the meteorological conditions (Styro et al. 2001). This was also noticed in Polish territorial waters (Špirkauskaitė et al. 1994, Bojanowski et al. 1995, Szefer 2002). Nevertheless, no correlation between ^{137}Cs and salinity was found in the surface water (Styro et al. 2001). Secondly, given the presence of the operating nuclear power reactor at Ignalina, Lithuania, which is a Chernobyl-type RBMK reactor, measuring the activity concentration of ^{137}Cs would offer the possibility of forecasting its distribution as a result of radioactive substances being transported in Nemunas river water in the aftermath of a serious accident involving the reactor.

It is important to note that the River Nemunas (Niemen) is the third largest river entering the Baltic Sea and its drainage basin covers nearly the whole of Lithuania. The Nemunas first discharges into the Curonian Lagoon and then flows through the Klaipeda Strait into the Baltic Sea. The effect of the water exchange between the Curonian Lagoon and the Baltic Sea on the long-term activity distribution of the artificial radionuclide ^{137}Cs in Lithuanian coastal waters is not yet known.

Of all the seas in the world, the Baltic is the most contaminated with the artificial radionuclide ^{137}Cs (NATO report 1998, Osvath et al. 2001). The origin of this contamination is diverse: global fallout as a result of nuclear weapons testing, release from reprocessing plants, input from the nine nuclear power plants that use sea water for reactor cooling in countries surrounding the Baltic, river run-off, and the fallout immediately after the Chernobyl Nuclear Power Plant (NPP) accident in 1986 (Nielsen 1997).

For the reasons given above, the circulation model of the North and Baltic Seas, known as BSHcmod, has been applied with a greater resolution to the south-eastern Baltic off the Lithuanian coast (Kleine 1994, Dick et al. 2001, Davuliene et al. 2002). This is a three-dimensional model of Lithuanian territorial waters, covering both the Lithuanian part of the Baltic Sea and the Curonian Lagoon. The applicability of this model to describe the distribution of radiocaesium activity concentration, which is found in both soluble and particulate forms in Lithuanian waters, is one of the issues that we are focusing on. If successfully validated, the model could be used to analyse the heterogeneous distribution of ^{137}Cs activity concentration in Lithuanian waters, as well as to assess the influence of the River Nemunas on Lithuanian territorial waters.

We also discuss recent projects assessing the spread of ^{137}Cs in the Lithuanian part of the Baltic Sea and in the Curonian Lagoon. The results of the ^{137}Cs activity concentration measurement campaign in 1999–2001 were used to validate the hydrodynamic model of the Lithuanian sea waters by considering the ^{137}Cs activity as a passive tracer.

2. Study area, material and methods

2.1. Study area

The region under consideration is the coastal zone of the Lithuanian part of the Baltic Sea and the Curonian Lagoon. A transitional water body, collecting the runoff waters from the Nemunas (98%) and other, minor rivers, the Curonian Lagoon is a shallow freshwater body separated from the Baltic Sea by the Curonian Spit. Annually, some 24 km^3 of water are transported from the lagoon to the sea. The lagoon is connected to the Baltic Sea via the narrow Klaipeda Strait, which has been artificially deepened to 14 m. Saline water is recorded mostly in the northern part of the lagoon. On rare occasions, however, it may penetrate 40 km into central part of the lagoon (Dubra et al. 1998).

The sea water mixes with the fresh water of the Curonian Lagoon in its upper layer, which is less than 5–7 metres deep. Fresh water is carried mostly in the NNW direction and can be traced as far as 7–10 nautical miles (n.m.) with salinity decreasing to 3 PSU in the Klaipeda Strait (Dubra et al. 1998). In the deeper waters (> 60 m) of the study area, i.e. in the open sea, the salinity in the water column above the thermocline is nearly uniform and reaches 7.5 PSU.

The open sea part of the study area is quite shallow, with an average depth of 30 m. The topography and structure of the sediments are strongly affected by wave action. The sediments on the Lithuanian coast consist mainly of particulate matter transported from the Curonian Lagoon, of erosion products from the Sambian peninsula carried by the longshore current, and of submerged moraine deposit outcrops accumulated mainly to the north of the Klaipeda Strait (Gavrilov et al. 1990). Coarse sand prevails near the coast. Moraine loam, silt and alevrites prevail in deeper places and between boulders, in canyons and pits. The bottom profile of the shallow coastal areas varies considerably as a result of storms. The transfer of resuspended sediments offshore takes place, resulting in the formation of radionuclide accumulation zones in the bottom sediments of the deeper sea areas.

The Curonian Lagoon is a shallow water body with a mean depth of 3.8 m. Therefore, sediments are periodically resuspended and mixed as a result of wind-induced wave action and water currents. The ^{137}Cs activity concentration is highly variable and site-dependent in the whole Curonian Lagoon, owing to differences in sediment composition and resuspension intensity (Tarasiuk et al. 1995). The ^{137}Cs activity concentration in the whole lagoon depends on the seasonal and annual variations of the Nemunas runoff, the water exchange between the lagoon and the Baltic Sea, and the meteorological conditions.

Several ^{137}Cs accumulation zones can be delineated in Lithuanian territorial waters. Anthropogenic ^{137}Cs accumulation zones on the Lithuanian coast of the Baltic have developed near deep-water pipe-lines. Here, accumulation is enhanced by coagulation because of the enforced interaction of the radionuclides in the sea water with underground fresh water or

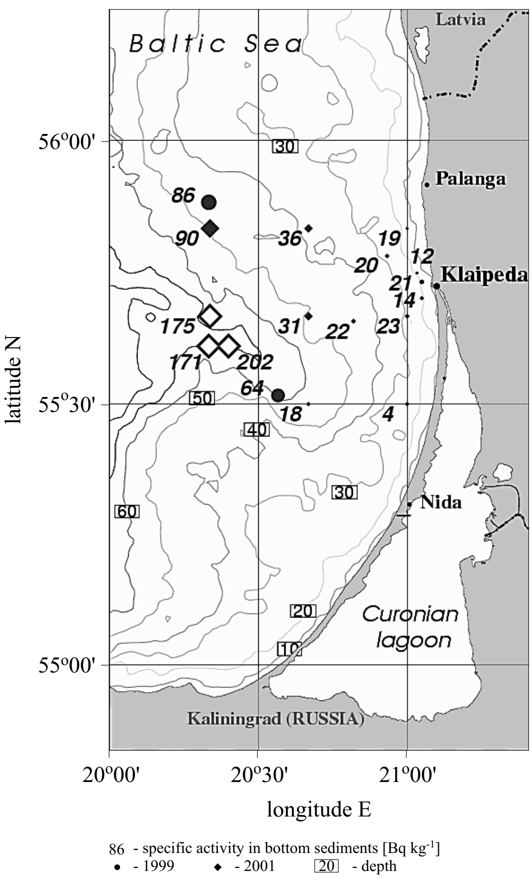

Fig. 1. Topography of the modelling area of the Baltic Sea and the measured ^{137}Cs specific activity in sediments

organic-rich industrial effluents (Tarasiuk & Spirkauskaite 1994). There are further anthropogenic ^{137}Cs accumulation zones in the Klaipeda Strait and in the Curonian Lagoon (Tarasiuk et al. 1995, Lujaniene et al. 2005).

The measurements indicate that the Klaipeda Strait can also be regarded as an important ^{137}Cs accumulation zone. Constant dredging means that the sediment distribution in the strait is continually changing. The dredged sediment is taken to the dumping site in the vicinity of station 20a in the Baltic Sea (Fig. 1). Detailed studies of the sediment transport were carried out by Galkus & Jokšas (1997).

In the marine environment ^{137}Cs is found in both soluble and particulate forms. The ability of suspended matter to bind the radionuclide changes on its way from the Nemunas river mouth to the Curonian Lagoon and in the coastal zone of the Baltic Sea. The first zone of serious changes is the Curonian Lagoon, which contains large amounts of organics (phytoplankton, algae, fish products, etc.) capable of accumulating ^{137}Cs. The second one is the fresh and saline sea water-mixing area in the Baltic Sea coastal zone.

2.2. Material and methods

Investigations of the distribution of ^{137}Cs activity in the Lithuanian part of the Baltic Sea were carried out in 1999–2001 (Tables 1–3). Water and bottom sediment samples were collected at the national and Helsinki Commission (HELCOM) monitoring stations in the Baltic Sea and in the Curonian Lagoon. Measurements were carried out during the campaigns at the background stations at Preila and Juodkrantė and on board the research vessel 'Vėjas' (at sea) and the cutter 'Gintaras' (in the lagoon). The stations visited in 2001 were located on a $20' \times 10'$ grid (Fig. 2). Some samples were also collected from the beach at Preila and Juodkrantė.

The volume of water samples varied from 22 to 150 litres depending on the lower detection limit of radiocaesium. They were collected from different horizons using 10–30 L bathometers.

Along with the water, soft sediment samples were taken using an Ekman-Birge type bottom sampler (effective sampling area of 225 cm^2), which enabled silt sediment cores from the Curonian Lagoon to be taken from depths down to 15 cm. In the Baltic Sea the van Veen grab sampler (surface area – 992 cm^2) available on r/v 'Vėjas' was used. Sediment cores were wet-sliced into layers (0.5–3.2 cm), dried at room temperature and analysed for vertical profiles of ^{137}Cs specific activity. For separate measurement of the ^{137}Cs activity concentration in the dissolved and suspended forms in sea and fresh water, samples were filtered through 0.2 μm Nuclepore (Dubna) and Nylon (Estonia) membranes (Filtrak-89 type filters were used as prefilters). Initially, filters were dried in desiccators to constant weight. After filtration,

Table 1. ^{137}Cs activities in the water and bottom sediments in the Lithuanian part of the Baltic Sea (1999–2000)

Date	Station No	Water depth [m]	Coordinates N E	Sampling depth [m]	PSU	Activity concentration[1] [Bq m^{-3}] total	dissolved	particulate	Particulate loading[1] [mg dm^{-3}]	Specific activity in bottom sediments[1] [Bq kg^{-1}]
1999.05.17	43	154	56°42′ 19°52′	surface	7.1	79±6	79±6	< 0.1	0.08	
1999.05.17	46a	73	56°03′ 19°24′	surface	7.1	76±7	76±7	< 0.1	0.06	
				70	9.3	77±6	77±6	< 0.1	-	-
1999.05.15	1b	28	56°02′ 20°50′	surface	6.7	62±6	62±6	< 0.1	0.18	
				25	7.0	69±8	69±8	< 0.1	0.07	-
1999.05.15	65	47	55°53′ 20°20′	surface	7.2	73±2	73±2	< 0.1	0.13	
				44	6.9	67±6	67±6	< 0.1	0.06	86±5
1999.05.15	64	34	55°46′ 20°54′	surface	6.4	63±6	63±6	< 0.1	0.1	
				30	7.0	72±6	72±6	< 0.1	0.05	-
1999.05.15	16	17	55°45′ 21°02′	surface	5.0	44±4	44±4	< 0.1	0.39	
				14	6.7	59±6	59±6	< 0.1	0.12	12±1
1999.05.15	4	14	55°44′ 21°03′	surface	3.9	41±3	41±3	< 0.1	0.55	
				12	6.6	52±5	52±5	< 0.1	0.17	21±2
1999.05.16	6b	65	55°31′ 20°34′	surface	6.6	62±3	62±3	0.2±0.1	-	
				62	8.5	73±5	73±5	< 0.1	-	64±3
1999.05.11	Preila	1.5	55°21′ 21°01′	surface[2]	6.7	77±6	71±6	6.4±0.7	1.61	
1999.05.13	Preila	1.5	55°21′ 21°01′	surface[2]	7.0	98±8	88±8	9.7±1.1	2.17	
1999.05.15	Preila	1.5	55°21′ 21°01′	surface[2]	6.9	85±5	84±5	0.9±0.3	3.21	
1999.05.17	Preila	1.5	55°21′ 21°01′	surface[2]	6.8	82±8	78±8	3.6±0.7	1.14	
1999.05.19	Preila	1.5	55°21′ 21°01′	surface[2]	6.7	75±7	73±7	1.8±0.4	1.12	
1999.05.21	Preila	1.5	55°21′ 21°01′	surface[2]	6.8	84±6	79±6	4.9±0.6	3.89	
1999.05.18	7	1.3	55°19′ 20°57′	surface	6.9	80±6	80±6	< 0.1	0.1	
1999.10.27	Preila	1.5	55°21′ 21°01′	surface[2]	6.9	89±6	81±5	8.0±1.2	22.6	
2000.06.12	Preila	1.5	55°21′ 21°01′	surface[2]	5.9	58±3	56±3	2.0±0.2	13.8	
2000.06.12	Preila	1.5	55°21′ 21°01′	surface[2]	5.9	59±3	57±3	2.0±0.2	15.2	
2000.06.16	Juodkrantė	1.3	55°32′ 21°06′	surface[2]	6.6	69±3	67±3	2.0±0.2	-	
2000.06.16	Juodkrantė	1.3	55°32′ 21°06′	surface[2]	6.5	63±3	61±3	2.0±0.2	-	

[1] Radiochemical analyses and estimation of ^{137}Cs activity concentration by dr. Galina Lujaniene (Institute of Physics, Lithuania); [2] from the beach.

Table 2. ^{137}Cs activities in the water and bottom sediments in the Lithuanian part of the Baltic Sea on 16 October 2001

Station No	Water depth [m]	Coordinates N E	Sampling depth [m]	PSU	Activity concentration (soluble fraction) [Bq m^{-3}]	^{137}Cs activity in surficial sediments	
						layer [cm]	activity [Bq kg^{-1}]
4	21	55°50′ 21°00′	surface	6.1	54±2		
			18	6.6	60±3	0.8	19±1
5	40	55°50′ 20°40′	surface	7.0	61±3		
			37	7.0	60±3	0.9	36±1
6	48	55°50′ 20°20′	surface	7.0	66±3		
			20	7.0	62±3		
			45	7.2	61±3	0.9	90±1
5I	29	55°46′ 20°56′	29	6.8	-	0.8	20±1
4a	18	55°44′ 21°03′	surface	5.6	39±2		
			15	6.0	55±2	0.9	14±1
7	29	55°40′ 21°00′	surface	7.0	63±4		
			26	7.0	62±3	0.9	23±1
8	48	55°40′ 20°40′	surface	7.0	62±3		
			20	7.0	61±3		
			45	7.1	59±4	0.9	31±1
9	58	55°40′ 20°20′	surface	7.0	64±3		
			20	7.0	67±3		
			40	7.0	62±3		
			55	7.3	59±3	2.4	175±2
20a	45	55°39′ 20°44′	surface	7.0	60±4		
			20	7.0	62±3		
			42	7.1	64±3	0.9	22±1
10	28	55°30′ 21°00′	surface	7.0	60±3		
			25	7.0	62±3	0.9	4.0±0.2
11	62	55°29′ 20°40′	surface	7.0	60±3		
			59	7.4	64±3	0.5	18±1
12	50	55°30′ 20°20′	surface	7.0	61±3		
			20	7.0	66±3		
			47	7.2	65±3	-	-
12a (I)	74	55°37′ 20°20′	74	8.5	-	2.6	171±5
12a(II)	74	55°37′ 20°24′	74	8.5	-	1.3	202±5

Table 3. ^{137}Cs activities in the water and sediments in the Curonian Lagoon (1999–2000)*

Date	Sampling station No	Water depth [m]	Coordinates N E	Sampling depth [m]	PSU	Activity concentration[1] [Bq m^{-3}]			Particulate loading[1] [mg dm^{-3}]	^{137}Cs activity in surficial sediments** [Bq kg^{-1}]
						total	dissolved	particulate		
1999.10.27	Nemunas river mouth, 12	1.5	55°20′ 21°17′	surface	0.0	8±1	7±1	0.7±0.1	4.6	-
1999.10.27	Nemunas river mouth, 12	1.5	55°20′ 21°17′	1.5	-	-	-	-	-	0.7±0.1
1999.10.27	Nida	3.5	55°19′ 21°01′	surface	0.0	23±2	16±1	7±1	32.9	-
1999.10.27	Nida	3.5	55°19′ 21°01′	3.5	-	-	-	-	-	58±6
1999.10.27	8	2	55°25′ 21°08′	surface	0.0	15±2	11±1	4±1	32.6	-
1999.10.27	8	2	55°25′ 21°08′	2.0	-	-	-	-	-	47±3
1999.10.27	Klaipeda port, 3	2	55°39′ 21°09′	surface	6.9	74±7	67±6	7±1	31.6	-
1999.10.27	Klaipeda port, 3	2	55°39′ 21°09′	2.0	-	-	-	-	-	22±1
1999.10.27	Klaipeda port, 2	13	55°42′ 21°08′	13.0	-	-	-	-	-	67±6
1999.10.27	Klaipeda port, 1	18	55°44′ 21°06′	surface	4.9	52±5	46±4	6±1	48.1	-
1999.10.27	Klaipeda port, 1	18	55°44′ 21°06′	12.0	5.4	74±7	54±5	20±2	102.0	-
1999.10.27	Klaipeda port, 1	18	55°44′ 21°06′	18.0	-	-	-	-	-	30±3
2000.06.13	Nida	4	55°19′ 21°01′	surface	0.0	2.3±0.4	0.3±0.1	2.0±0.3	37.9	-
2000.06.13	Nida	4	55°19′ 21°01′	4.0	-	-	-	-	-	90±6
2000.06.14	Preila	4	55°21′ 21°04′	surface	0.0	1.1±0.2	0.5±0.1	0.6±0.1	36.7	-
2000.06.14	Preila	4	55°21′ 21°04′	4.0	-	-	-		-	71±7
2000.06.18	Klaipeda port, 3	8	55°44′ 21°09′	surface	5.2	53±2	52±2	1.0±0.2	9.00	-
2000.06.18	Klaipeda port, 3	8	55°44′ 21°09′	8.0	-	-	-	-	-	150±11

*Radiochemical analyses and estimation of ^{137}Cs activity concentration by dr. Galina Lujaniene; ** averaged over the upper 20 cm layer of the sediment column.

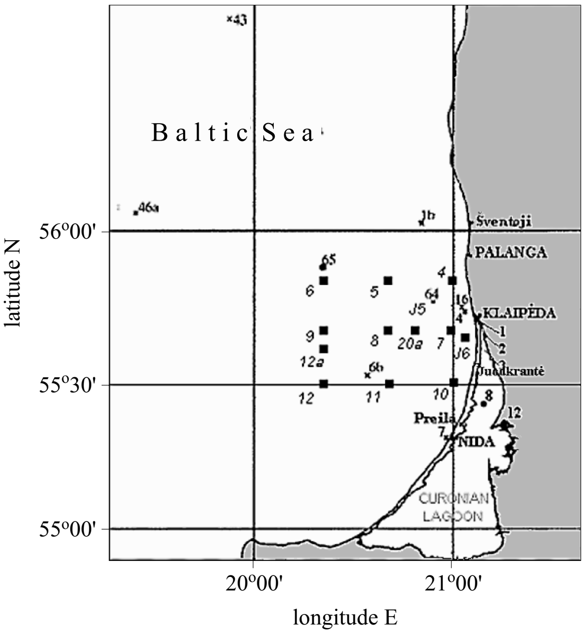

Fig. 2. Location of the sampling stations in the Baltic Sea and the Curonian Lagoon during investigations in 1999–2000 (asterisk) and 2001 (rectangles)

the filters and prefilters were dried at room temperature in the desiccator and weighed once more to determine the amount of suspended matter.

As a rule, sediments in the accumulation zones of the Curonian Lagoon are completely mixed owing to the permanent wave action and the shallowness of the water. Therefore, specific activities of the sediment samples taken in the Curonian Lagoon are shown averaged over the upper 20 cm layer of the sediment column.

To calculate the ^{137}Cs activity concentration in water (dissolved form), the water filtrate was treated radiochemically using the ferrocyanide precipitation method (Borisenko et al. 1986). ^{134}Cs was used to monitor the yield of the chemical procedure.

The water and sediment samples were analysed for ^{137}Cs using the SILENA and ORTEC γ-spectrometric systems with an HPGe detector, and also using a γ-spectrometer in conjunction with a Ge(Li) semiconductor detector. The γ-spectrometric calibration was carried out using radioactive sources of different density (1.0 and 1.45 g cm^{-3}) and the radionuclide mixture (^{152}Eu+^{137}Cs) prepared by the Russian Scientific Research Institute of Physico-Technical and Radiometric Measurements (Moscow, Russia). Measurement errors of the radionuclide concentrations for the γ-spectrometer with the Ge(Li) detector were calculated manually.

The measurement errors for the γ-spectrometric systems with the HPGe detector were calculated by the SILENA and the ORTEC software programs GAMMAPLUS and GAMMA VISION. Estimated errors were mostly less than 30% at the 95% confidence level (1.96σ).

For long-term measurements (350 000 s) using the SILENA and ORTEC γ-spectrometric systems the respective detection limits of the ^{137}Cs activity concentration in 100 L water samples and the sediment samples were 0.1 Bq m^{-3} and 0.2 Bq kg^{-1}. The errors indicated in Tables 1–3 are due to counting statistics. These errors usually comprise less than 7% at the 95% confidence level ($\pm 1.96\sigma$).

3. Results

Field data on the ^{137}Cs activity concentration in the marine sediments showed few zones of ^{137}Cs accumulation in the eastern part of the Baltic Sea. The sea bottom was covered mainly with sand of grain size varying from coarse to alevritic (densities varied from 1.73 to 1.37 g cm^{-3}) or with a silt-size admixture (station 6, depth 45 m, density 0.91 g cm^{-3}). In the latter case, the ^{137}Cs specific activity in the surface sediments was rather high, up to 90 Bq kg^{-1} dry weight (d.w.) (Table 2). The lowest ^{137}Cs specific activity (8–21 Bq kg^{-1}) on the Lithuanian coast of the Baltic Sea was recorded in the sandy sediments of shallow waters down to the 5-metre isobath. Since the percentage of the fine fraction of suspended particles in sediments increases with depth, the ^{137}Cs specific activity in sediments is also expected to do so. Generally, the ^{137}Cs specific activity in the bottom sediments varied from 4 to 90 Bq kg^{-1} d.w. (Tables 1 and 2). The main accumulation zone of ^{137}Cs in the area under investigation was found in the region known as the old bed of the River Nemunas in the Baltic Sea (stations 9, 12a) (Table 2).

The ^{137}Cs specific activities in the bottom sediments sampled in the Curonian Lagoon varied from 0.7 to 90 Bq kg^{-1}. A high ^{137}Cs specific activity (150 Bq kg^{-1} d.w.) was characteristic of black silt sediments found in the port of Klaipeda (station 2a) and at the Klaipeda municipal sewage outfall (Fig. 2, Table 3), but ^{137}Cs activities in the sediments of the Klaipeda Strait (Klaipeda port, station 3) were low (22 Bq kg^{-1} d.w.). A comparatively high ^{137}Cs specific activity in the bottom sediments measured near human settlements in the central part of the Lagoon (at Preila – up to 71 Bq kg^{-1} d.w.) could be related to local zones of anthropogenic organic pollution. The thick layer of black silty bottom sediments was sampled in the vicinity of Nida; the ^{137}Cs specific activity in them varied from 58 to 90 Bq kg^{-1} d.w. (Table 3).

According to the field data obtained in May 1999, the ^{137}Cs activity concentration in open sea surface water was almost uniform at a level of 73 –79 Bq m^{-3}. In October 2001 it decreased by about 20% to 60–67 Bq m^{-3} (Fig. 3). The lowest ^{137}Cs activity concentration in the sea (39–53 Bq m^{-3}) was found in the samples collected at stations 4, 4a and 16, which are located in the close vicinity of the Klaipeda Strait, where substantial mixing of fresh and sea water takes place. The highest ^{137}Cs activity concentration (75–98 Bq m^{-3}) was observed at the Preila background station in May 1999. No increase was found in October 2001, however (Tables 1 and 2).

The ^{137}Cs activity concentrations in the Baltic Sea were linearly correlated with the salinity of the water (Fig. 4). The increase in salinity

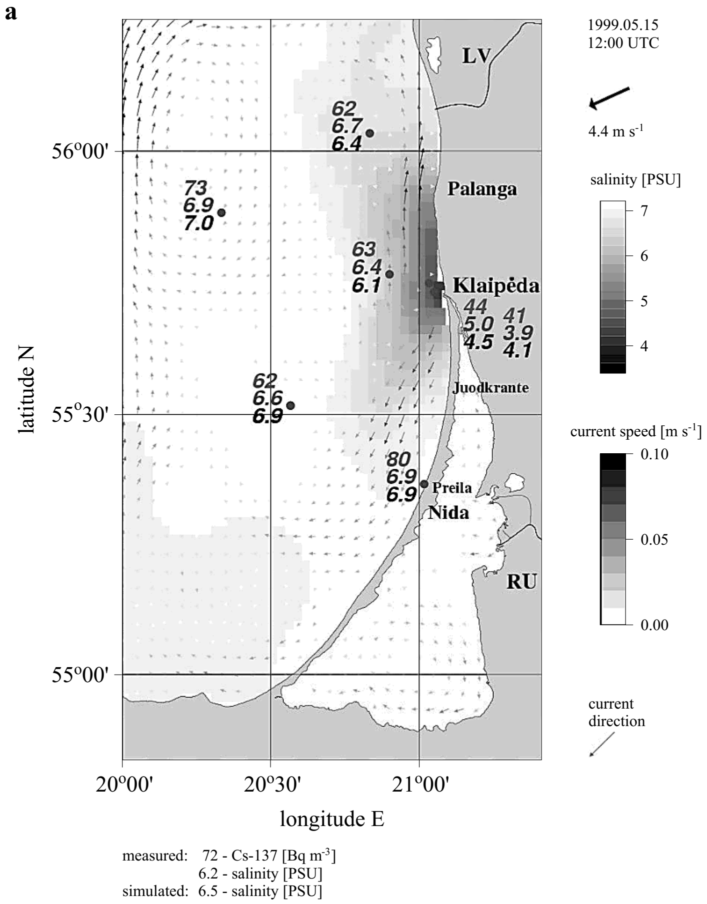

Fig. 3. Measured ^{137}Cs activity concentration and salinity together with the simulated salinity distribution on 15–16 May 1999 (a) and on 16–17 October 2001 (b). (Salinity in the Curonian Lagoon is < 3.5 PSU)

b

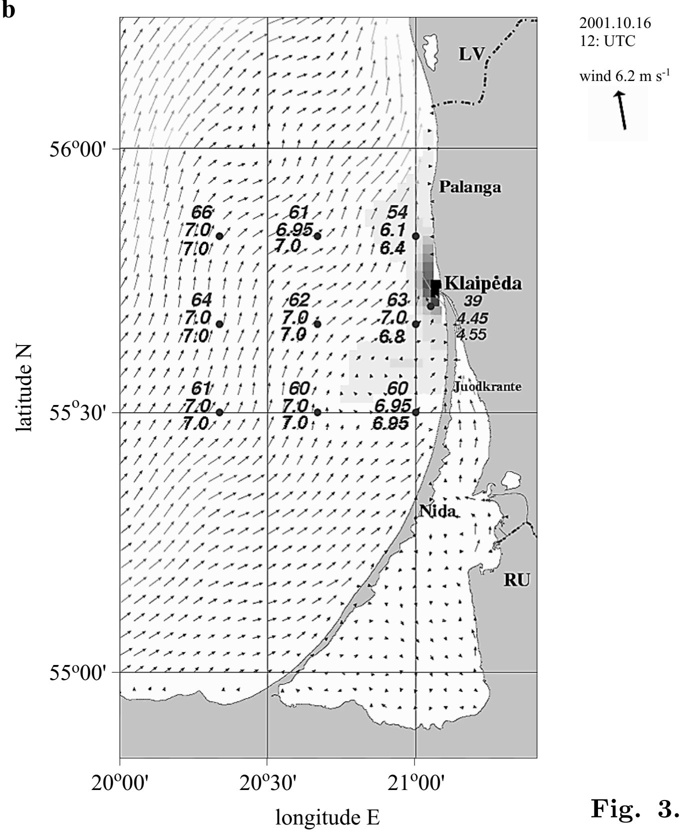

Fig. 3. (*continued*)

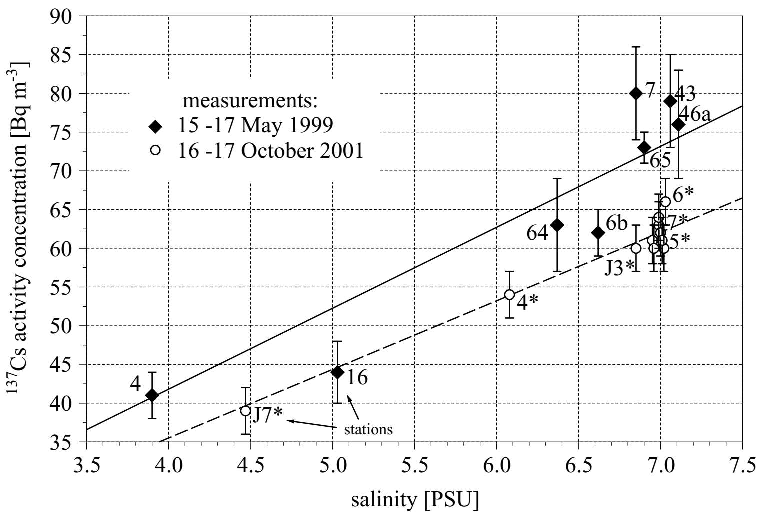

Fig. 4. Relationship between salinity and ^{137}Cs activity concentration

up to the mean value for the open sea surface water (about 7 PSU) was followed by an increase in the ^{137}Cs activity concentration to 82 Bq m^{-3} in 1999 and to 66 Bq m^{-3} in 2001. The correlation coefficients between the measured radionuclide activity concentration and the salinity for the two periods were 0.94 and 0.90, with respective standard deviations of 5.8 and 1.7 Bq m^{-3}.

During the May 1999 field work, the ^{137}Cs total dissolved and suspended activity concentration was measured. The activity concentration of ^{137}Cs bound to suspended matter varied from 0.9 to 9.7 Bq m^{-3} in the coastal zone of the Baltic Sea (Table 1), making up to 10% of the total ^{137}Cs activity. The ^{137}Cs specific activity in samples of suspended matter collected in the open waters of the Baltic Sea was below detection limits (Tables 1 and 2).

Large variations in the ^{137}Cs activity concentration were observed in the northern and central parts of the Curonian Lagoon, including the Klaipeda Strait, during the storm of October 1999. The total ^{137}Cs activity concentration varied from 8 to 74 Bq m^{-3}, the highest values being recorded in the Klaipeda Strait and decreasing values towards the central part of the Curonian Lagoon (Table 3). A W and NW wind of 12 m s^{-1} prevailed during the field work and sea water flowed into the Curonian Lagoon. During this period, the activities of ^{137}Cs in its water-soluble and particulate forms in the Curonian Lagoon varied considerably, from 7 to 67 Bq m^{-3} for water-soluble and from 0.7 to 20 Bq m^{-3} for particulate caesium (Table 3). The percentage of particulate ^{137}Cs in the Curonian Lagoon water samples varied from 4 to 27% of the total activity.

During the field work in the Curonian Lagoon on 13–18 June 2000 the meteorological conditions were rather calm with a dominant SE wind of 5 m s^{-1}. The total ^{137}Cs activity concentration measured in the central part of the Curonian Lagoon ranged from 1.1 to 2.3 Bq m^{-3}. This range was due mainly to considerable variations in particulate ^{137}Cs activities, which reached 87% of the total activity (Table 3, Nida station).

The particulate loading showed only a slight correlation with the particulate ^{137}Cs activity concentration measured in the Curonian Lagoon. During the stormy and calm weather periods, the particulate loading in the northern and central part of the Curonian Lagoon was about 35 mg dm^{-3}. This is a considerably higher value than the measured particulate loading in the coastal zone of the sea (22.6 mg dm^{-3}, Preila station) and in the open waters of the Baltic Sea (< 0.1 mg dm^{-3}, stations 43, 46a) (Tables 1 and 3). The particulate loading in the Lithuanian coastal zone varied from 0.06 to 0.55 mg dm^{-3} owing to the influence of the fresh water outflow from the Curonian Lagoon. It is worth noting that the particulate loading in the Klaipeda Strait reached 102.2 mg dm^{-3} in the near-bottom water layer during the stormy period in October 1999.

4. Model

4.1. Model implementation

The three-dimensional circulation model of the BSH (Bundesamt für Seeschiffahrt und Hydrographie) for the North and Baltic Seas (BSHcmod) was adapted for the Lithuanian coastal area of the Baltic Sea. The model was validated in the Curonian Lagoon and the south-eastern part of the Baltic Sea off the Lithuanian coast using the 1999–2001 temperature and salinity measurement data (Davuliene et al. 2002). A detailed model description is given by Kleine (1994) and Dick et al. (2001) and will not be discussed here. The model enables us to obtain information about the water level, water temperature and salinity as well as about the dispersion of passive substances in a real time period. The advection-diffusion equation in spherical coordinates λ, ϕ, z was applied for a passive substance C as follows:

$$\frac{\partial C}{\partial t} + \frac{1}{R\cos\phi}\frac{\partial (uC)}{\partial \lambda} + \frac{1}{R\cos\phi}\frac{\partial (v\cos\phi\, C)}{\partial \phi} + \frac{\partial (wC)}{\partial z} = \tag{1}$$

$$= \frac{1}{R\cos\phi}\frac{\partial}{\partial\lambda}\left(\frac{K_h}{R\cos\phi}\frac{\partial C}{\partial\lambda}\right) + \frac{1}{R\cos\phi}\frac{\partial}{\partial\phi}\left(\frac{\cos\phi\, K_h}{R}\frac{\partial C}{\partial\lambda}\right) + \frac{\partial}{\partial z}\left(K_v\frac{\partial C}{\partial z}\right),$$

where u, v, w are the velocity components of flow, and K_h and K_v are the coefficients of the horizontal and vertical diffusion of mass. The definitions of these coefficients are briefly described by Kleine (1994).

The area selected for modelling (with the upper left corner at 56°20′45″N, 19°55′25″E) covered the Lithuanian part of the Baltic Sea including the Curonian Lagoon, and part of the Latvian and Kaliningrad (Russia) coasts (Fig. 1). A 1 nm grid (or $1' \times 1'40''$) was chosen and the vertical water column was divided into five layers. Only one river input (the Nemunas splitting into two branches – the Atmata and the Skirvytė) was taken into account. For the western and northern open boundaries the BSHcmod simulation data were used.

The applicability of the circulation model to simulate the ^{137}Cs activity concentration distribution as a passive tracer in the Baltic Sea near the Lithuanian coast was also examined before the model was applied to our coastal configuration.

4.2. Hydrodynamic model of the Lithuanian part of the Baltic Sea: validation for ^{137}Cs modelling

As already discussed in the experimental part, the measurement data show a linear relation between salinity and ^{137}Cs activity concentration (Fig. 4). The salinity and passive tracer distributions for the cruise periods were simulated in order to assess the applicability of the circulation model for simulating the distribution of ^{137}Cs activity concentration in Lithuanian territorial waters (Fig. 3). The respective standard deviations between simulated and measured salinity for the two periods in 1999 and 2001 were 0.34 and 0.19 PSU. This is about 10% of the salinity variation in the surface water of the area. Using the linear relation, the ^{137}Cs activity concentration can be calculated from measured or simulated salinity data.

The correlations between the simulated distribution of the tracer and measured ^{137}Cs activity concentration during the field campaigns were 0.93 and 0.96, with respective mean standard errors of about 6 and 2 Bq m^{-3} (Fig. 5). This indicates that the hydrodynamic model of Lithuanian marine waters for simulating the ^{137}Cs activity concentration is applicable within an error gap of about 15%.

4.3. Modelling results

The mean ^{137}Cs activity concentration in Nemunas river mouth water is less than 2 Bq m^{-3} (Tarasiuk et al. 1999). Water from the Nemunas can therefore be regarded as a minor factor contributing to the ^{137}Cs activity concentration in the south-eastern part of the Baltic Sea coastal zone. The observed fluctuation in ^{137}Cs activity concentration can thus be attributed unequivocally to the distribution of fresh water along the Lithuanian coast (Fig. 3). The fresh water flow in Lithuanian coastal waters is readily detected from the salinity decrease. In the reduced salinity zone the correlation between the ^{137}Cs activity concentration and the salinity is most useful.

The data sets of the ^{137}Cs activity concentration measured during the two field campaigns show different data scattering. As follows from the simulations of these two periods the variation in ^{137}Cs activity concentration could also be explained by the fresh water distribution in the coastal zone. Several days before the field work on 11–14 May 1999 E and SE winds were dominant. Fresh water from the Curonian Lagoon was spreading along the Lithuanian coast in a northerly direction from the Klaipeda Strait. During the field work, however, the wind backed N and NW. As a result, the fresh water spreading from the Curonian Lagoon was now moving south-west from the Klaipeda Strait. This caused a decrease in salinity followed by a considerable decrease in the ^{137}Cs activity concentration in a large area of

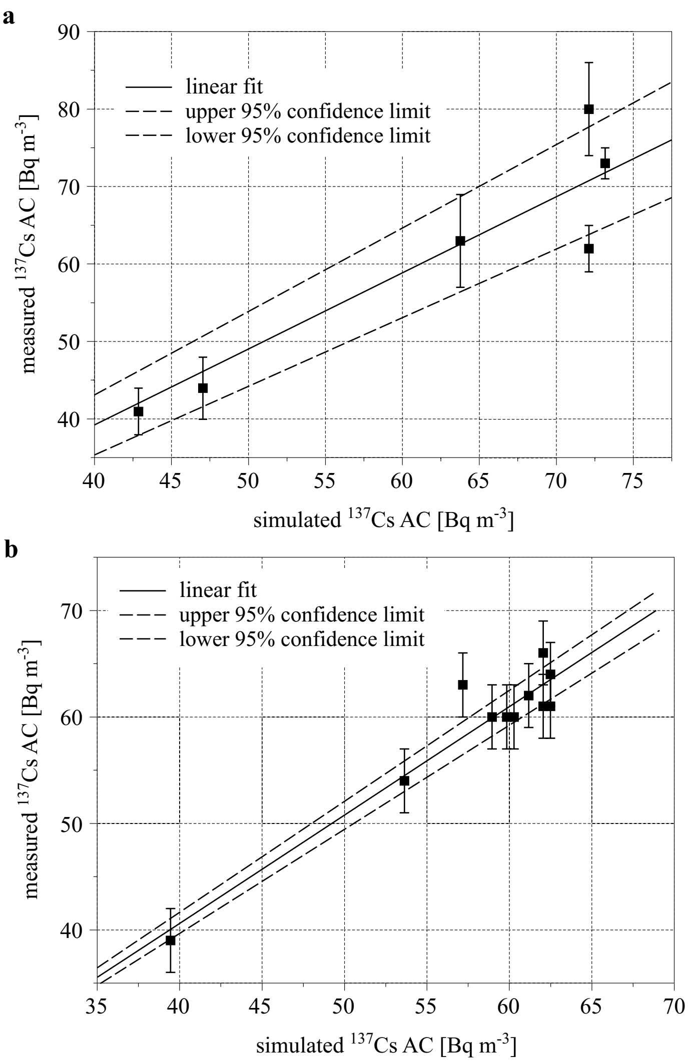

Fig. 5. Measured and simulated ^{137}Cs activity concentration (AC) on 15–16 May 1999 (a) and on 16–17 October 2001 (b) together with error bars and 95% confidence limits

the Baltic Sea off the Lithuanian coast during the period 15–16 May 1999 (Fig. 3a). But this was not the case during the field campaign of 16–17 October 2001. As a result of the strong (up to 15 m s^{-1}) onshore W wind dominating from 11 October 2001, the sea level off Lithuanian coast rose and the water outflow through the Klaipeda Strait was all but stemmed. Later, the wind backed SE and water outflow through the strait resumed; the measured salinity as well as variations in the ^{137}Cs activity concentration at the Lithuanian coast were rather weak (Fig. 3b).

The simulations of the salinity distribution for 1999–2001 have shown that the fresh water flow from the Curonian Lagoon modifies the salinity in the Baltic Sea off the Lithuanian coast mostly in an area 15 km from the shore (Davuliene et al. 2002a). Because of the dominant N and SE winds on the Lithuanian coast, the average simulated salinity distribution generally displays the lowest salinities along the shore to the north of the Klaipeda Strait. The simulated salinity variability was found to be negligible in the area approximately 30 km offshore. In the open sea the measured salinity and ^{137}Cs activity concentration can be regarded as constant; the salinity in this area has a constant value down to the halocline. The measured ^{137}Cs activity concentration in the water column down to the halocline fluctuates around the mean ^{137}Cs activity concentration within the range of the measurement error, indicating no ascending or descending trends of decrease or increase. Therefore, the values determined in this area could be used for model calibration purposes as background values typical of the south-eastern Baltic Sea.

5. Discussion and conclusions

A comprehensive study of the spread of ^{137}Cs was carried out in the south-eastern Baltic Sea off the Lithuanian coast and in the Curonian Lagoon. During this study two field campaigns during 1999–2001 were organised in Lithuanian territorial waters. The total activity concentration of ^{137}Cs in the Baltic Sea remains at a high level compared to the World Ocean (NATO report 1998). At present this value in the south-eastern Baltic is 60–70 Bq m^{-3}. The measured ^{137}Cs total activity concentration in open Baltic waters is determined by the ongoing process of self-cleaning.

Our measurements revealed a linear relation between the ^{137}Cs activity concentration and the salinity in the mixing zone of fresh and saline waters. The proportionality coefficient between the ^{137}Cs activity concentration and the salinity depends on the average ^{137}Cs activity concentration in the Baltic Sea. The applied hydrodynamic model enabled us to simulate the salinity distribution in Lithuanian sea waters and to assess the extent of the mixing zone.

Measurements carried out in the Lithuanian part of the Baltic Sea and in the Curonian Lagoon in different seasons indicate that the ^{137}Cs activity concentrations in the water depend on the hydrological situation as well as on the water exchange between the sea and the Curonian Lagoon. According to the relation found between salinity and ^{137}Cs activity, the observed fluctuation of the ^{137}Cs activity concentration in the Baltic Sea off the Lithuanian coast can be clearly attributed to the distribution of fresh water along the Lithuanian coast (reliability error within c. 15%). Along with the applied hydrodynamic model of Lithuanian sea waters, this relation may help to indicate the possible contribution of ^{137}Cs activity sources not taken into consideration when estimating the total ^{137}Cs activity concentration.

The measured ^{137}Cs activity concentration in the surface water in the central Baltic (stations 46a and 43) in spring 1999 was on average 5 Bq m^{-3} higher than that off the Lithuanian coast (offshore station 65). In the open sea and in coastal waters far from the fresh and saline sea water mixing zone radiocaesium was found mostly in the soluble form. The particulate ^{137}Cs activity concentration in the coastal water was $< 10\%$ of the ^{137}Cs total activity concentration.

According to the measurement data, the main ^{137}Cs accumulation zone in the area under consideration is located in the region that happens to be the old bed of the River Nemunas (below the 50 metre isobath), which is in the reduced hydrodynamic activity zone below the thermocline (Fig. 1). The ^{137}Cs specific activity of the sediments here is about 10 times higher than the value measured on the coast.

According to the measurements, the activity concentration of particulate ^{137}Cs in the northern and central part of the Curonian Lagoon can be as high as 90% during calm weather. However, as the total ^{137}Cs activity concentration in the fresh water of the Curonian Lagoon is $< 5\%$ of the total ^{137}Cs activity in the open sea water, this fact is of minor importance for modelling the ^{137}Cs activity concentration distribution in Lithuanian territorial waters.

During storms with dominant NW and W winds, saline intrusions into the Curonian Lagoon occur; this occurs more often during the autumn (Dubra & Dubra 1998). The influence of this process on the salinity and on the ^{137}Cs activity concentration distributions in the Curonian Lagoon has not yet been studied in detail. During the study period in October 1999 the ^{137}Cs activity concentration in the northern part of the Lagoon was measured, thus highlighting the effect of the saline water intrusion. During this event, the total ^{137}Cs activity concentration in the northern part of the Curonian Lagoon increased markedly and varied over a wide range. The largest values characteristic of the open sea were found in the Klaipeda Strait. The particulate ^{137}Cs activity concentration measured in the surface water of the Curonian Lagoon reached about 6 Bq m^{-3} and was nearly 5 times higher than the ^{137}Cs total activity concentration measured during calm weather.

It can be concluded that the dispersion of the ^{137}Cs activity concentration in the Curonian Lagoon as a passive tracer may be subject to certain limitations. Resuspension may be of crucial importance to the total ^{137}Cs activity concentration during stormy weather. In general, the contribution of all ^{137}Cs accumulation zones located in Lithuanian territorial waters to the total ^{137}Cs activity concentration depending on the meteorological situation has not been studied yet.

The high correlation between simulated and measured ^{137}Cs activity concentrations off the Lithuanian coast shows the hydrodynamic model for Lithuanian territorial waters to be suitable for assessing spatial variations of ^{137}Cs activity concentration (error within c. 15%).

The variability in salinity was found to be negligible in an area approximately 30 km offshore. For model calibration purposes, therefore, the ^{137}Cs activity concentration measured at this distance could be used as the background value typical of the south-eastern part of the Baltic Sea.

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