BOREAL ENVIRONMENT RESEARCH 11: 95–107 ISSN 1239-6095
Helsinki 24 April 2006

Estimation of recent sediment accumulation rates in the Baltic Sea using artificial radionuclides ^{137}Cs and $^{239,240}Pu$ as time markers

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Received 10 Mar. 2005, accepted 28 June 2005 (Editor in charge of this article: Johanna Mattila)

Mattila, J., Kankaanpää, H. & Ilus, E. 2006: Estimation of recent sediment accumulation rates in the Baltic Sea using artificial radionuclides ^{137}Cs and $^{239,240}Pu$ as time markers. *Boreal Env. Res.* 11: 95–107.

This study reviews sediment data obtained from the Baltic Sea from 1995–2003. Recent sediment accumulation rates (SAR) were estimated at 69 stations in the Baltic Sea from 99 sediment cores using concentration peaks of ^{137}Cs and $^{239,240}Pu$ as time markers of the years 1986 and 1963, respectively. SAR values varied widely between 60–6160 g m^{-2} yr^{-1}. The highest SAR values were observed at stations in the northern part of the Bothnian Sea, river estuaries and the eastern part of the Gulf of Finland. Generally, the SAR (median) of stations in the Bothnian Sea was two times higher than values in the Gulf of Finland or the Bothnian Bay, and about seven times higher than in the Baltic Proper. A strong correlation was detected between the SAR and total ^{137}Cs activities. The usability of estimation methods has been considered and, e.g., unstable sedimentation could limit the use of radionuclides in sediment dating and estimation of the SAR at stations in the Baltic Sea.

Introduction

The sediment accumulation rate (SAR) or net sedimentation is an essential parameter in contamination or monitoring studies and in budget calculations (Wulff *et al*. 1993, Kankaanpää *et al*. 1997, Vallius and Leivuori 1999, Emeis *et al*. 2000, Isosaari *et al*. 2002). We dated sediment cores from the Baltic Sea in several projects dealing with the estimation of sediment accumulation rates (Ilus *et al*. 2001), organic pollutants (Isosaari *et al*. 2002), the history of cyanobacterial blooms (Poutanen and Nikkilä 2001), catchment nutrient flows and coastal retention (SEGUE project coordinated by the Finnish Environment Institute), factors influencing phosphorus and silicon binding in the sediment (SEGUE project coordinated by the Finnish Institute of Marine Research) and several other smaller co-projects carried out with the Finnish Institute of Marine Research and the Finnish Environment Institute.

The vertical distribution of artificial radionuclides ^{137}Cs and $^{239,240}Pu$ and the naturally occurring radionuclide ^{210}Pb in sediments can be used in sediment dating and in SAR estimation. Such methods were used for nearly three decades in several earlier studies in the Baltic Sea (Simola *et al*. 1979, Niemistö and Voipio 1981, Kankaanpää *et al*. 1997). These radionuclides have high concentration factors for sediments in the brackish water environment of the Baltic Sea (IAEA 1985, HELCOM 2003, Ikäheimonen 2003), and the use of the highest concentration as a time marker is also supported by time series available from the Gulf of Finland (HELCOM 2003). ^{137}Cs in Baltic Sea sediments mostly originates from the fallout following the Chernobyl accident in 1986, while the origin of ^{239,240}Pu is mainly the fallout from global weapons testing in the 1960s (HELCOM 2003). The origin and general circulation of naturally occurring ^{210}Pb is well known and various models have been developed for using this nuclide in sediment dating (Robbins 1978, Appleby and Oldfield 1992). ^{210}Pb can be used over a time scale of about 100–150 years.

Due to the heterogeneity of soft sediment deposits, variation in the SAR inside sedimentation basins and between closely-situated positions can be large. Sediment layers formed over thousands of years may vary considerably depending on the bottom topography (Winterhalter 1972, 1992, 2001). The thicknesses of more recent soft sediment layers inside sedimentation basins may also indicate large differences in accumulation (Kankaanpää *et al*. 1997, Vallius and Leivuori 1999, Perttilä *et al*. 2003). On a small spatial scale, variation in the SAR could have serious effects on monitoring results, such as on the total ^{137}Cs activities recorded in sediments (HELCOM 2003). Sediment sampling methods and equipment may also noticeably affect the results (Blomqvist 1985, Crusius and Anderson 1991, HELCOM 2000).

This study reviewed the SAR estimates from our sediment investigations conducted from 1995–2003. The data obtained were also used to estimate internal variation in the SAR in some sedimentation basins and around some permanent monitoring stations. In addition, the repeatability of sediment sampling was tested. The SAR estimates were based on time markers, and the values were generally compared with those based on ^{210}Pb models. Annually accumulated sediment layers (AAL) were also counted on the basis of the SAR results and sediment data. Additionally, results obtained with two sediment corers were compared. Summarizing these results enables the mapping of SARs in the study area and evaluation of the applicability of SAR estimation methods.

Material and methods

Sediment samples were mainly taken during cruises by the Finnish Institute of Marine Research (FIMR) on r/v *Aranda*. Most of the samples were taken with GEMINI or GEMAX twin corers that give two parallel cores of 80 mm or 90 mm in diameter, respectively. To study the smearing effect, outer layers of the second GEMAX core were peeled to produce a core with a diameter of 80 mm. All the samples had undisturbed surface layers. Most of the sediment cores were sliced into 1-cm subsamples to a depth of 10–20 cm, and in most cases the rest of the core below this was sliced at 2-cm intervals as deeply as possible, generally to a depth of 30 cm. All the sediment samples were frozen (–20 °C) immediately after sampling. In the laboratory they were freeze dried, homogenized and weighed before and after drying to determine the dry matter contents. The amount of salt in the sediment was calculated using water content of the sediment slice and salinity in the near-bottom water.

^{137}Cs profiles were analysed from each core. Activities of ^{239,240}Pu could only be analysed from 17 stations, because of the high costs of analysis, while ^{210}Pb profiles were analysed from 56 cores. ^{137}Cs and ^{210}Pb activities were determined by gamma-ray spectrometry, taking into account the self-absorption of gamma rays in samples of varying density and height (Klemola *et al*. 1996). When the concentrations of excess ^{210}Pb were calculated, the supported ^{210}Pb concentration (assumed to be equal to ^{226}Ra concentration) was subtracted from the total ^{210}Pb activity measured. ^{226}Ra activities were obtained using the weighted average activity of ^{214}Bi (609.3 keV), ^{214}Pb (295.2 keV) and ^{214}Pb (351.9 keV) activities. ^{239,240}Pu analyses were performed according to the method of Taipale and Tuomainen (1985). Total ^{137}Cs activities (Bq m^{-2}) were calculated from the ^{137}Cs activity profiles by multiplying ^{137}Cs concentrations (Bq g^{-1}) by dry matter weights (g m^{-2}) and summing the results for each sediment slice in the sediment core.

In the estimation of the SAR by means of ^{137}Cs or ^{239,240}Pu, the highest measured radionuclide concentration was used as a time marker in the sediment profile (reference years 1986 for ^{137}Cs and 1963 for ^{239,240}Pu). Depths of time markers were identified from the profiles as objectively as possible by using concentrations and their measurement errors. In clear cases, time markers could be bounded to one sediment slice, but in many cases the time marker was assumed to be situated in a broader sediment layer (two or more sediment slices). Calculations were based on the equation:

$$\mathrm{SAR} \pm \Delta\mathrm{SAR} = \frac{m \pm \Delta m}{t} \quad (1)$$

where SAR is the average bulk sediment accumulation rate between the year of sampling and the time marker (g m^{-2} yr^{-1}), ΔSAR is the uncertainty in the SAR, m is the average cumulative dry matter weight (mass depth, g m^{-2}) at the highest concentration, Δm is the difference between the average cumulative dry matter weight and the lower or upper edge of the sediment layer in which the highest concentration exists, and t is the time (years) between sampling and the reference year. This general equation could be used with other time markers in estimating the SAR. The salt-free dry matter weights of the sediments were also used in estimation of the SAR values to consider the magnitude of the possible salt corrections.

Two sediment dating models based on ^{210}Pb were also used. One was the Cf:Cs (constant flux:constant sedimentation rate) model and the other the CRS (Constant Rate of Supply) model (Robbins 1978, Appleby and Oldfield 1992, Crusius and Anderson 1995) In the Cf:Cs model the SAR is assumed to be constant, while in the CRS model it can be variable. Excess ^{210}Pb fluxes were calculated by multiplying the total excess ^{210}Pb activity of the core by the ^{210}Pb decay constant.

The thickness of the annually accumulated layer (AAL, mm yr^{-1}) comparable to the estimated SAR was calculated by using the SAR and porosities of the sediment (Crusius and Anderson 1995). Dry matter density was assumed to be 2.5 g cm^{-3} (Niemistö *et al.* 1978). AAL could also be graphically resolved by using the cumulative weight of dry matter (g m^{-2}) curve against depths of the sediment slices (cm), assuming minor effects of the sediment sampler on sediment core shortening. With both methods AAL could be estimated to the surface or to deeper layers of the sediment.

Results

The average bulk SAR values were between 60–6160 g m^{-2} yr^{-1} at 69 stations and in 99 cores (Table 1). The highest SAR values were observed in the northern part of the Bothnian Sea (stations F18, US2, US5b), near the mouth of river Paimionjoki (stations Paila 14, As5, Paila 10), at one station in the sedimentation basin south of Åland (station F69) and in the eastern Gulf of Finland (stations LL3a, SL11b) (Table 1). The highest total ^{137}Cs activities were found in the northern part of the Bothnian Sea. At stations within 0–10 km from the mouth of the Paimionjoki the average SAR was about 4900 g m^{-2} yr^{-1} (stations Paila 10, Paila 14 and AS5), while at stations 20 to 40 km distant from the river mouth the SAR was about 900 g m^{-2} yr^{-1} (stations AS2 and AS3). At several stations in the eastern part of the Gulf of Finland the SAR values were clearly over 1000 g m^{-2} yr^{-1}, with maximum values of nearly 3000 g m^{-2} yr^{-1}. At stations about 10 km from the eastern mouth of the Kymijoki (stations KAS8 and K15) the SAR was about 1200 g m^{-2} yr^{-1}, while at stations about 20 km from the river mouth (K19, K20 and K27) the SAR was about 900 g m^{-2} yr^{-1}. At stations F69 and F15 the SAR and total ^{137}Cs activities were substantially higher than typical levels in the represented sea areas of the Baltic Proper and the Bothnian Bay, respectively (Tables 1 and 2, Fig. 1). Because of the relatively low salinities of the water, the salt free SAR values were clearly smaller than the average bulk SAR values only at some stations of the Baltic Proper (Table 1). At the other stations the effects of salt corrections were considered to be insignificant.

At the Bothnian Sea stations the median SAR values were two, three and seven times higher than at the stations of the Bothnian Bay, Gulf of Finland and Baltic Proper, respectively (Table 2). Time-corrected total ^{137}Cs activities were four times higher in the Bothnian Sea than in the Gulf of Finland or the Bothnian Bay and nearly 12 times higher than in the Baltic Proper. Correlation coefficients between SAR values and total ^{137}Cs activities were high, especially at the stations of the Bothnian Sea, Bothnian Bay and Gulf of Finland (Table 2). The average dry matter contents in surface sediments (0–2 cm) were

Table 1. Sampling stations of the study, samplings dates (dd.mm.yy), depths (*d*, m), positions latitude (Lat.) and longitude (Long.) in degrees in the WGS-84 coordinate system, SAR (g m^{-2} yr^{-1}), SAR estimation method (m), estimated ranges of SAR in percentages (±), annually accumulated layer AAL (mm yr^{-1}) to surface (AALs) sediment layers (0–2 cm) and to deeper (AALd) sediment layers (about 10 cm) and total ^{137}Cs activities (Bq m^{-2}). Symbols (S): – = clear peculiarities in sediment profiles or uncertain results, v = study of internal variation in sediment accumulation area, s = comparison of sediment samplers,! = parallel cores, a = Gemax Ø 90 mm, b = peeled Gemax Ø 80 mm, c = Gemini Ø 80 mm. The SAR values are not salt corrected. Correction terms in percentages (corr) higher than 10% for the salt-free SAR values are given (how many percent smaller is the salt-free value as compared with presented SAR value).

Station	Date	*d*	Lat.	Long.	SAR	m	±	AALs	AALd	Cs_{tot}	S	corr
Bothnian Bay												
F15	16.06.96	48	63.5169	21.5137	2270	Cs	10	16.4	4.8	98350		
	18.06.99	47	63.5168	21.5130	1530	Cs	9	13.5	3.1	90900		
SM3	18.06.96	78	63.9836	21.7625	120	Cs	100	0.5	0.2	7400	–	
	18.06.99	81	63.9943	21.7730	680	Cs	15	4.4	2.9	13550		
BO3	17.06.96	109	64.3006	22.3470	910	Cs	13	6.2	3.7	17490		
F9	18.06.96	122	64.7003	22.0628	320	Cs	25	2.7	1.0	9830		
					280	Pu	12	2.4	0.9			
RR6	17.06.96	85	64.8001	23.4793	280	Cs	100	1.8	0.7	5660	–	
RR3	17.06.96	94	64.9336	22.3460	1070	Cs	29	5.8	4.0	9700	–	
CV	17.06.96	87	65.0002	23.2459	220	Cs	100	1.1	0.5	6200	–	
CVI	17.06.96	68	65.2340	23.5636	290	Cs	100	1.3	0.6	4330	–	
					350	Pu	17	1.6	0.7			
Bothnian Sea												
AS2	23.09.03	47	60.0813	22.2647	900	Cs	24	6.3	3.4	15580		
F64	16.06.99	286	60.1890	19.1427	320	Cs	100	2.1	0.7	13410	–	
AS3	26.09.03	19	60.2212	22.4330	840	Cs	21	5.2	2.4	11610		
AS5	25.09.03	33	60.3078	22.5005	4650	Cs	4	34.5	14.7	54740		
Paila14	24.09.03	29	60.3250	22.5225	5540	Cs		38.3	16.6	> 61530	–	
Paila10	24.09.03	12	60.3690	22.5802	4580	Cs	8	24.6	13.0	58480		
SR6	19.06.96	102	61.0509	20.2634	950	Cs	100	4.5	2.2	29170	–	
SR7	17.06.99	78	61.0837	20.5968	270	Cs	100	0.8	0.4	9480	–	
SR5	15.06.96	125	61.0839	19.5798	1160	Cs	81	6.2	3.2	31490		
					1090	Pu	6	5.8	3.0			
OLK12	28.06.95	16	61.2318	21.3799	1810	Cs	22	12.5	6.5	36950		
SR1a	16.06.99	64	61.2327	17.6622	930	Cs	37	4.9	0.9	39090	–	
EB3	21.06.99	130	61.5002	20.3297	1190	Cs	10	6.9	3.9	31940		
F26	21.06.99	138	61.9835	20.0628	1200	Cs	20	5.8	3.3	27670		
US5b	18.06.96	209	62.5865	19.9666	4090	Cs	32	22.6	14.9	100180		
US6b	18.06.96	82	62.6000	20.2626	150	Cs	100	0.5	0.3	7800	–	
					110	Pu	100	0.4	0.2		–	
US2	20.06.99	201	62.8453	18.8885	5250	Cs	20	24.1	16.0	121770	–	
F18	16.06.96	105	63.3170	20.2800	6160	Cs	13	27.7	15.6	115910		
	18.06.99	104	63.3142	20.2727	5710	Cs	8	26.9	14.6	113340		
Gulf of Finland												
JML	24.04.96	80	59.5821	23.6267	350	Cs	60	2.4	1.1	3770	v!	
					430	Cs	25	3.2	1.6	3760	v!	
	24.04.96	80	59.5813	23.6242	370	Cs	67	2.5	1.4	4250	v	
	24.04.96	80	59.5818	23.6255	360	Cs	66	2.6	1.5	4300	v	
	24.04.96	80	59.5818	23.6265	440	Cs	22	3.1	1.6	4110	v	
	29.04.95	80	59.5818	23.6268	190	Cs	100	1.1	0.7	4110	–	
	25.05.99	80	59.5822	23.6302	360	Cs	82	2.5	1.5	4940		
					470	Pu	13	3.3	1.9			
	27.09.03	80	59.5818	23.6267	170	Cs	76	1.7	0.6	4850		

Continued

Table 1. Continued.

Station	Date	*d*	Lat.	Long.	SAR	m	±	AALs	AALd	Cs_{tot}	S	corr
LL11	14.06.96	68	59.5833	23.2982	320	Cs	34	1.7	0.5	3640	–	
	25.05.99	67	59.5851	23.2961	140	Cs	100	0.3	0.2	930	–	
	25.05.99	67	59.5872	23.2965	140	Cs	53	0.9	0.3	5970	–,s,a,!	
					460	Cs	72	2.5	0.9	6500	–,s,b,!	
	25.05.99	67	59.5872	23.2965	200	Cs	42	1.1	0.3	3720	–,s,c	
GF1	28.04.95	84	59.7051	24.6821	1250	Cs	12	5.7	4.9	7640		
	28.09.03	83	59.7052	24.6820	350	Cs	94	3.6	1.4	5690	–	
GF2	27.04.95	84	59.8385	25.8569	380	Cs	100	2.3	1.3	6230	–	
	26.04.96	84	59.8414	25.8716	960	Cs	6	6.5	4.2	7850		
Hanko Bay	17.08.95	74	59.8434	22.8998							–	
LL6a	27.05.96	72	59.9148	25.0330	750	Cs	52	4.6	3.3	8600		
LL4a	13.06.96	66	60.0162	26.0719	260	Cs	64	2.0	0.7	4200		
	18.05.01	64	60.0158	26.0742	240	Cs	63	1.5	0.5	7590		
SL2	14.05.96	28	60.0589	29.1961	1430	Cs	22	7.0	4.4	21860		
	12.12.97	29	60.0584	29.1998	1090	Cs	12	7.3	3.0	17730		
					750	Pu	21	5.0	2.1			
LL3a	12.06.96	68	60.0677	26.3466	2940	Cs	5	24.9	15.7	37460		
	02.06.98	66	60.0668	26.3466	2440	Cs	3	24.9	13.1	37100		
					1490	Pu	4	15.2	8.0			
F40S	12.12.97	37	60.1004	28.7728	460	Cs	61	2.7	1.6	8160	–	
F41	25.04.95	51	60.1176	28.0613	1290	Cs	9	12.5	4.5	24090		
					570	Pu	16	5.6	2.0			
SL6	15.05.96	39	60.1328	28.6442	1270	Cs	15	10.0	4.0	14330		
F42	13.06.96	63	60.1336	27.4641	1170	Cs	25	9.3	2.2	19430	–	
	22.05.99	63	60.1351	27.4664	410	Cs	100	3.2	1.1	13160	–,s,a,!	
	22.05.99	63	60.1351	27.4664	440	Cs	79	2.9	1.1	13620	–,s,b,!	
	24.05.99	64	60.1352	27.4666	690	Cs	27	4.5	2.0		–	
SL11b	17.05.96	50	60.2334	27.7185	2530	Cs	5	31.1	13.5	58930		
SL8	16.05.96	38	60.2418	28.3885	300	Cs	28	2.8	1.0	8370		
XV1	26.04.95	64	60.2502	27.2475	240	Cs	54	2.4	1.6	10820		
	02.06.98	57	60.2441	27.2459	880	Cs	16	17.5	5.1	26280		
					860	Pu	8	17.3	5.0			
	22.05.99	64	60.2500	27.2473	350	Cs	15	5.5	2.3	13280	s,a,!	
	22.05.99	64	60.2500	27.2473	340	Cs	18	5.6	1.9	18430	s,b,!	
	22.05.99	64	60.2500	27.2473	410	Cs	15	6.0	2.9	17800	s,c	
K19	04.06.98	38	60.3239	26.8788	1190	Cs	7	36.1	9.6	48250		
K20	04.06.98	37	60.3286	27.0167	660	Cs	36	14.2	3.0	24100		
					400	Pu	22	8.5	1.8			
K27	04.06.98	30	60.3332	26.5823	870	Cs	40	9.9	5.5	34880		
LOV3	08.06.95	18	60.3702	26.3819	880	Cs	9	10.9	5.3	40340		
					650	Pu	4	8.0	3.9			
K15	11.12.97	20	60.4011	26.9535	1240	Cs	5	15.0	7.0	49760		
					1310	Pu	4	15.9	7.4			
A1	15.02.96	8	60.4177	26.4921	840	Cs	21	6.8	3.5	33640		
					530	Pu	22	4.2	2.2			
KAS8	12.08.97	20	60.4676	27.0726	1090	Cs	13	14.9	6.6	74420		
Baltic Proper												
BY2	23.05.01	48	55.0000	14.0835						2690	–	
BCSIII10	01.06.96	90	55.5497	18.3969	440	Cs	100	2.5	1.4	3600	–	
HL6	31.05.96	102	56.4832	19.9636	80	Cs	100	0.5	0.1	2180	–	
HL4	31.05.96	144	56.4833	19.5636	60	Cs	100	0.5	0.3	1760	–	11

Continued

Table 1. Continued.

Station	Date	*d*	Lat.	Long.	SAR	m	±	AALs	AALd	Cs_{tot}	S	corr
HL1	01.06.96	99	56.4917	18.8307	80	Cs	100	0.5	0.2	2010	–	
HA1	30.05.96	88	56.9367	18.8237	130	Cs	100	0.5	0.3	1140	–	
HA5	30.05.96	178	56.9382	19.8069	160	Cs	25	3.0	0.8	1 040		21
					120	Pu	11	2.3	0.6			18
HA8	31.05.96	133	56.9499	20.2636	90	Cs	100	0.8	0.3	2040	–	10
HA9	31.05.96	86	56.9501	20.4069	230	Cs	100	1.1	0.3	3040	–	
BCSIVb2-4	27.05.99	240	57.2815	20.0964	120	Cs	32	2.3	2.0	990		23
					250	Pu	5	4.9	4.4		–	20
BCSIVb6	29.05.96	115	57.3001	20.5302	250	Cs	60	2.6	1.5	4370		10
BCSIVb4	29.05.96	237	57.3166	20.0303	140	Cs	21	3.3	0.7	710		26
					110	Pu	15	2.5	0.6			21
LF2	29.05.96	81	57.9833	21.0802	680	Cs	73	3.8	2.3	10230	–	
LF4	28.05.96	101	57.9916	20.4136	100	Cs	47	1.4	0.3	1310		15
F80	28.05.96	195	58.0101	19.8968	170	Cs	25	3.3	1.9	1590		20
IBSV10	20.05.01	78	58.3500	20.2472	90	Cs	100	0.2	0.1	4200	–	
LL17	23.04.96	173	59.0360	21.0775	160	Cs	60	2.0	0.5	1570	v	16
	23.04.96	180	59.0360	21.0825	410	Cs	27	5.3	4.4	3210	–,v	15
	23.04.96	180	59.0360	21.0830	520	Cs	13	7.6	6.0	3290	–,v	15
	23.04.96	180	59.0362	21.0836	480	Cs	17	6.5	5.0	3030	–,v	14
LL15	02.06.96	130	59.1834	21.7470	290	Cs	76	10.0	1.9	7430	–	13
AS7	22.09.03	71	59.4667	21.9417	840	Cs	33	6.2	2.3	9940		
LL12	28.05.96	82	59.4834	22.8968	310	Cs	60	2.1	1.0	4110		
F69	14.06.96	189	59.7820	19.9303	3410	Cs	32	15.4	10.8	38500	–	

about 16% at the stations of the Bothnian Bay and Bothnian Sea and about 12% at the stations of the Gulf of Finland and Baltic Proper, where dry matter contents at a depth of around 10 cm were approximately 32% and 25%, respectively.

Table 2. Median values of the SAR (g m^{-2} yr^{-1}), AAL (mm yr^{-1}) to surface (AALs) sediment layers (0–2 cm) and to deeper (AALd) sediment layers (about 10 cm) and total ^{137}Cs activities (Cs_{tot}, Bq m^{-2}) for stations of the different sea areas. Spearman rank correlation coefficients (r_s) were calculated between the SAR values and the total ^{137}Cs activities. To calculate median values, the total ^{137}Cs activities at each station were time-corrected to the year 2000. In cases where several sediment cores were taken at the same station on the same sampling date, average values of observations were used to calculate the SAR and AAL. The medium SAR value of the Baltic Proper would be 20% smaller if salt free values are used in the calculations.

Stations of the	SAR	AALs	AALd	Cs_{tot}	r_s
Bothnian Bay	500	3.5	1.9	9000	0.81
Bothnian Sea	1200	6.2	3.7	32400	0.93
Gulf of Finland	690	5.6	2.4	13200	0.73
Baltic Proper	180	2.6	0.8	2600	0.64

In the sedimentation basin of station JML (western Gulf of Finland), within a distance of about 170 metres, the coefficients of variation for mean SAR values and total ^{137}Cs activities in 1996 were 11% and 6%, respectively (Table 1). In the vicinity of station LL17 (Baltic Proper), the highest estimated SAR values (average) were about three times higher than the lowest, although the distances between sampling positions were only about 350 metres (Table 1). Variable activity and SAR results were observed in repeated samplings at certain stations in different years. Variability between SAR estimates and total ^{137}Cs activities seemed to be highest at stations SM3 (1300 m), LL11 (450 m), GF2 (880 m), XV1 (690 m) and F42 (230 m) (distances between sampling locations in parentheses) (Table 1). At stations F15 (40 m), F18 (490 m), JML (170 m), LL4a (130 m) and LL3a (100 m) variation was less, especially when the ranges of estimation errors of SAR were taken into account (Table 1).

Calculated error ranges of the SAR estimates based on the ^{137}Cs method were below 25% in 40% of the cases. In about one fifth of the cases

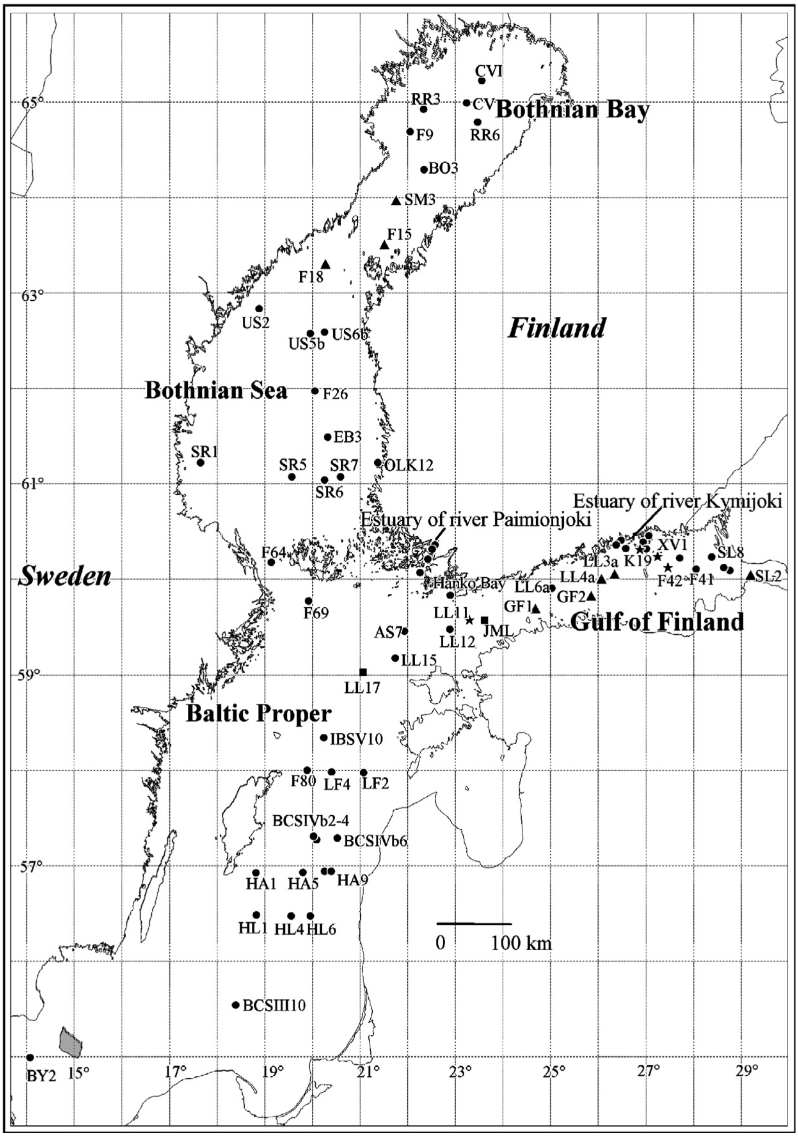

Fig. 1. Locations of sediment sampling stations of the present study (1995–2003). Stations visited once (dots), more than once (triangles), sedimentation basins for variation studies (squares) and stations where sediment sampling was also studied (stars) are indicated.

the error range was 100%. With the exception of one station, the ^{239,240}Pu method produced error ranges of below 25%. When taking into account the ranges of these estimates, the ^{137}Cs and ^{239,240}Pu methods provided comparable results for 10 stations out of 17 and at three stations the estimates were clearly different (Table 1). At station BCSIVb2-4, the Pu method gave SAR values that were twice as high as those given by the Cs method, but at stations F41 and LL3a the situation was the opposite. There was generally a positive correlation between estimates based on the ^{137}Cs method and the ^{210}Pb models. The difference between average SAR estimates based on the two ^{210}Pb models and the ^{137}Cs method were under 25% at nearly half the stations and over 100% at about one fifth of the stations. The mean SAR estimates based on Cf:Cs and CRS models were clearly divergent (difference over 50%) at about 15% of the stations.

Various radionuclide activity and dry matter content profiles were measured, but reasonable categorisation of these profiles was not possible. In some sediment profiles there were excellent time markers (Fig. 2a), but in others, as at station BY2, clear disturbances were observed in activ-

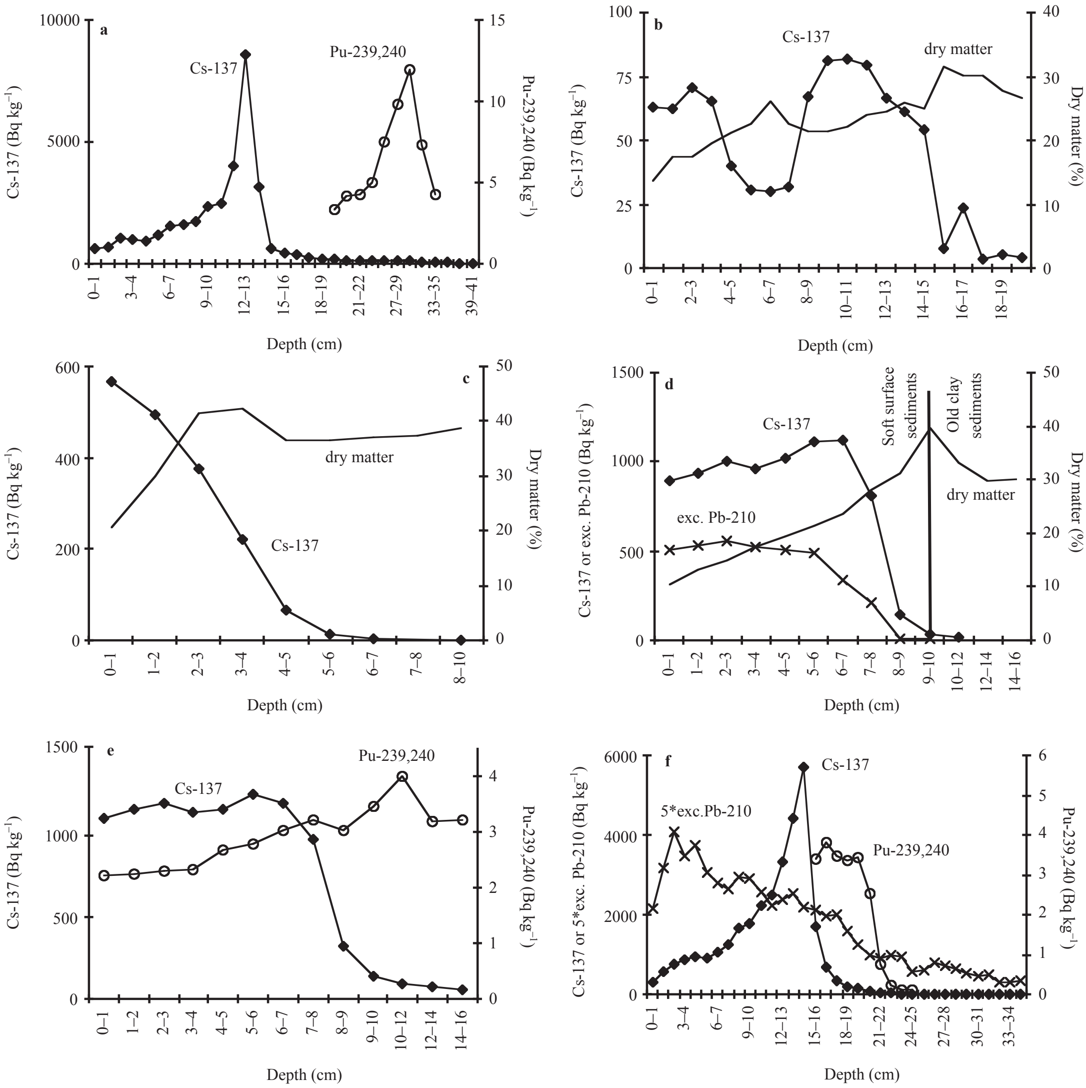

Fig. 2. Examples of various activity and dry matter profiles in sediments. ^{137}Cs, ^{239,240}Pu, excess ^{210}Pb (exc.^{210}Pb) activity concentrations and/or dry matter content profiles at stations (**a**) K15, (**b**) BY2, (**c**) US6b, (**d**) F42, (**e**) SR5 and (**f**) K19. At station K19, a coefficient of five was used in scaling exc.^{210}Pb activities.

ity profiles due to episodic accumulation of different kinds of sediments (Fig. 2b). Because of the same type of effect within the sedimentation basin around station LL17, completely different activity profiles and SAR values were recorded. Profiles of the type observed at station BY2 could lead to clear overestimation of the SAR for particular time periods. Estimation of the SAR was consequently avoided at station BY2, and also at the Hanko Bay station due to a thick sand layer in the surface layers of the core. The highest radionuclide concentrations could also be present in the top or topmost slices, as at station US6b (Fig. 2c). At such stations the estimated SAR was relatively low and the SAR range was 100%, or close to it. At station F42 (eastern Gulf of Finland) there were surface layers of recent soft sediments followed by layers of old clay, which were clearly also reflected in the radionuclide and dry matter content profiles (Fig. 2d). At station SR5 a broad layer of high ^{137}Cs concentrations was observed, although the ^{239,240}Pu activities peaked quite clearly at a depth of 10–11 cm (Fig. 2e). In most of the seventeen ^{239,240}Pu

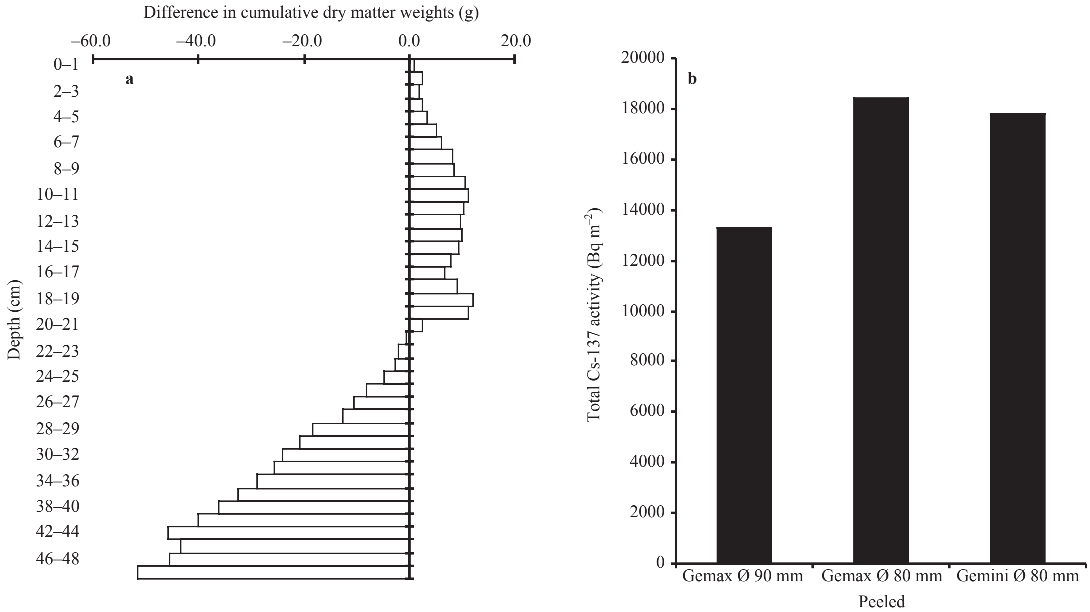

Fig. 3. — **a**: Difference in cumulative dry matter weights (g) as a function of slicing depth (cm) in two simultaneously collected parallel cores (Gemini twin corer; station K19) sliced by two persons at the same time with similar slicing equipment. — **b**: Total ^{137}Cs activities of three parallel sediment cores taken with the Gemax and Gemini twin corers at station XV1. The diameters of the Gemax cores were 90 mm without peeling (Gemax Ø 90 mm) or 80 mm with peeling of the outermost margin of the core (Gemax Ø 80 mm peeled). The diameter of the Gemini core was 80 mm (Gemini Ø 80 mm).

profiles measured an observable subsurface peak formed the time marker. However, at station K19 there was no clear ^{239,240}Pu peak at all, although the ^{137}Cs peak was well defined and the excess ^{210}Pb profile was reasonably exponential, indicating continuous accumulation of recent sediments (Fig. 2f). In the most deeply located cases, the ^{137}Cs peak was situated at around or below a depth of 20 cm (in the Bothnian Sea stations F18, US2, Paila 10 and AS5, in the Gulf of Finland station LL3a). The most deeply located ^{239,240}Pu peaks were found as deep as about 30 cm (in the Gulf of Finland stations LL3a and K15).

At station K19, possible errors caused by sediment slicing were examined in parallel GEMINI cores (Fig. 3a). The difference in the cumulative weight of dry matter as a function of depth (mass depth) between the parallel cores changed from positive to negative. In the deepest layers (48–50 cm) the difference was about 50 g, which corresponds to a difference of about 9 years (based on the average SAR estimate in calculation). The effects of the sediment sampler and sampling procedures on cumulative dry matter weights and total ^{137}Cs activities were examined using cores from stations XV-1 (very soft sediment), F42 (soft surface layers followed by deeper layers of clay) and LL11 (a thin surface layer of soft sediment followed by relatively compact clay-rich sediments). At station XV-1 there was a clear difference in total ^{137}Cs activities between the two corers (Fig. 3b). Because of differences in sediment compaction the total ^{137}Cs activities in (Bq m^{-2}) were nearly 40% greater after peeling of soft sediments. The total ^{137}Cs activities obtained with the GEMINI corer were over 30% greater than those with the GEMAX corer without peeling. At other stations peeling seemed to have a much smaller effect on the results, and at station LL11, with compacted sediments, the results obtained with the GEMAX corer were greater than those with the GEMINI corer.

Discussion

The large variation in SAR values was due to several factors, although generally the most important factors affecting sediment accumulation in these relatively shallow sea areas are water depth and bottom topography (Winterhalter 1972, 1992, 2001, Brydsten 1993). Intensive accumulation could be promoted by the sheltered locations or, as in the river estuaries and in the eastern part of the Gulf of Finland, an estuarine-type current system and flocculation of particulate material (Morris *et al.* 1988, Winterhalter 1992, Brydsten 1993, Pitkänen 1994). In the northern Bothnian Sea and southern Bothnian Bay, there appear to be areas with effective accumulation due to abundant sources of accumulating material and dynamic features such as bottom topography (Winterhalter 1992, Brydsten 1993). In deeper areas south of Åland, in the northern Baltic Proper, effective accumulation of recent sediments quite probably occurs also due to the same type factors as, for example, in the Bothnian Sea.

There were clear differences between the studied sea areas, such as in the surface areas of accumulation bottoms, the influence of transportation and erosion processes on bottom areas, the loading from rivers and the amount of primary production (Voipio 1981, HELCOM 2002, 2003, 2004). These partly explain the differences in results between the separate sea areas studied. For example, in the Gulf of Bothnia over 50% of the bottoms are influenced by transportation and erosion processes (Winterhalter 1972, Brydsten 1993), providing considerable sedimentary material from shallower areas to areas of accumulation. Large amounts of eroded sediment and large river loads have affected the composition of accumulating material (clays and silt), and average dry matter contents of surface sediments were higher in the Bothnian Bay and Bothnian Sea, while sediments in the accumulation bottoms of the Gulf of Finland and Baltic Proper contained more organic material with lower dry matter weights (HELCOM 2002).

The strong correlations between the SAR and the total ^{137}Cs activities (Table 2) and excess ^{210}Pb fluxes are mostly due to the high sediment/water distribution coefficients (K_d) of these metals in the brackish water environment of the Baltic Sea (Duursma and Gross 1971, IAEA 1985, HELCOM 2003). On a larger scale, the variation in total ^{137}Cs activities is primarily due to the distribution of Chernobyl fallout (HELCOM 2003). Nonetheless, based on the present results, it seems that the correlation between the SAR and total ^{137}Cs activities could in certain cases be used to confirm the validity of SAR estimations. This correlation could offer a valuable tool for SAR estimation within smaller areas with a similar deposition history, and also around monitoring stations. Based on the variation studies, estimation of total ^{137}Cs activities was also more accurate than the SAR estimation.

In previous studies, estimates were based on various methods such as the load of solid substances, the thicknesses of Litorina stage sediments, nutrient budgets, sediment laminates, sedimentation traps, mercury concentrations and radionuclides (Ignatius 1958, Ignatius and Niemistö 1971, Winterhalter 1972, Tulkki 1977, Niemistö *et al.* 1978, Niemistö 1982, Niemistö 1986, Pitkänen 1994, Salo *et al.* 1986, Tuomainen *et al.* 1986, Jonsson *et al.* 1990, Perttilä *et al.* 1995, Kankaanpää *et al.* 1997, Perttilä *et al.* 2003). All these methods have different time scales and the resulting AAL values (mm yr^{-1}) depend on how the compaction of the sediments was taken into account in the estimations, or whether it was considered at all. However, the previous general AAL estimates of 0.6–1.3 mm yr^{-1} for the Bothnian Bay (Tulkki 1977, Niemistö 1986), 0.9–2.4 mm yr^{-1} for the Bothnian Sea (Niemistö 1986), 1.5–4 mm yr^{-1} or about 400 g m^{-2} yr^{-1} for the Gulf of Finland (Salo *et al.* 1986, Perttilä *et al.* 1995, Kankaanpää *et al.* 1997), 0.5–2.3 mm yr^{-1} for areas in the Baltic Proper (Ignatius 1958, Niemistö 1986, Jonsson *et al.* 1990) or 1200 g m^{-2} yr^{-1} for the estuary of the Kymijoki (Pitkänen 1994) were reasonably or highly comparable with the results of this study. Nonetheless, it should be kept in mind that reliable mean values for sea areas require a large amount of data and the median values of the present study could therefore only be described as tentative values.

Variation in the SAR estimates and total ^{137}Cs activities can be substantial within sedimentation basins and cores collected from the same stations in different years. This variation has also previously been discussed (Niemistö and Voipio 1974, Winterhalter 1992, Kankaanpää *et al.* 1997, Vallius 1999, Perttilä 2003). Uneven sediment accumulation within sedimentation basins, irregular accumulation and transportation or even erosion of accumulated material are mostly due to unstable sedimentation processes in the Baltic Sea (Winterhalter 1972, 1992, Tulkki 1977, Emelyanov 1988, Brydsten 1993, Perttilä *et al.* 2003). Occasional accumulation or transportation events could occur even in the deepest areas of the Baltic Sea, as results of the present study also clearly illustrated (Winterhalter 1992, Perttilä *et al.* 2003). Within the sedimentation basin of station LL17 (northern Baltic Proper), accumulation of relatively thick layers of sediments with high water contents at only slightly deeper locations reflected the influence of bottom topography on recent sediment accumulation. At station BCSIVb2-4 the presence of sediment layers with a high water content ("fluffy layers"), which has been observed in the deep bottoms of the Baltic Proper (Perttilä *et al.* 2003), also caused large variation in the SAR values estimated by two independent time markers.

The selection of suitable stations or sedimentation areas for monitoring work or for sediment sampling in general is a highly important quality factor, but also difficult in bottom areas of the Baltic Sea (Perttilä *et al.* 2003). There have been good considerations of the suitability of stations in the Baltic for the monitoring work (Larsen and Jensen 1989). Nonetheless, as the results of the present study also indicate, we consider it very important to empirically test the repeatability of sediment sampling around monitoring stations and correctly check differences with repeated samplings before long-term monitoring is undertaken. However, it could be difficult to find a reliable parameter for reliably testing variation. SAR estimation and sediment sampling methods, and even the persons performing sediment sampling could cause variation in the results. Based on the results of this study, the total ^{137}Cs activities could provide a straightforward means to study the internal variation in sediment accumulation within certain sediment basins if standardized methods are used in sediment sampling. Results of the present study also emphasize the need for accurate positions in monitoring studies.

Based on the present results and comparison of different methods, the combination of several estimation methods is recommended in sediment dating and in estimation of the SAR, especially in areas with substantial heterogeneity. Where ^{210}Pb-based methods are used there is a need for time markers to validate the dating results (Smith 2001). The vertical distribution of radionuclides in sediment cores could be affected by several biological, chemical and physical processes such as mixing processes (Duursma and Gross 1971, Håkansson *et al.* 1996, HELCOM 2000). These, together with unstable sedimentation, could limit the use of time markers and ^{210}Pb models in SAR estimation in the Baltic Sea. In most of the cases in this study the SAR could be quite reasonably estimated with time markers. Nevertheless, in many cases the time marker in the ^{137}Cs profile was broad or situated near to the sediment surface, causing estimates to be highly approximate, whereas more deeply situated time markers, such as ^{239,240}Pu, provided better results. In general, the application of the ^{239,240}Pu method is limited by its high costs and time-consuming analysis. The fulfilment of the model assumptions should be considered when using time markers and models based on ^{210}Pb. Dating results and SAR estimation could be erroneous in activity profiles where heavier material has occasionally accumulated or sediment layers are missing because of resedimentation.

Interpretation of radionuclide, dry matter and other available stratigraphies in sediment columns and comparison of the SAR results obtained using different methods are also tools for evaluating the quality and reliability of SAR estimates. It is relatively easy to distinguish radionuclide profiles and SAR results of high or poor quality. Profiles with minor disturbances or sediment cores, where periodical accumulation or transportation of sediment layers has occurred, are more difficult to identify. For example, the combined use of several methods confirmed that station F42 (eastern Gulf of Finland) represented a bottom with accumulation and transportation events, although based on the soft surface sediments it clearly represents an area of sediment accumulation. Furthermore, at station SR5 (Bothnian Sea), the use of ^{239,240}Pu allowed for good estimation of the SAR, but at the same time indicated that factors other than the mixing of surface sediments could also affect the formation of broad time markers in ^{137}Cs profiles. In the Bothnian Sea these factors could be related to accumulating material and sources of it. Standard methods are also needed to avoid subjectivity in profile reading, especially when time markers are not very distinct.

Sediment corers and sampling procedures, such as slicing, significantly affect the estimation of the SAR and the total ^{137}Cs activities. The corer diameter influences the compaction of the sediment core, especially in soft sediments (Blomqvist 1985, Crusius and Anderson 1991, HELCOM 2000). Additional treatment of wet sediment cores, such as peeling off the outermost margin, could compress the core. Moreover, corer dimensions and core slicing could affect the bulk density of the sediment core (Blomqvist 1985, Joshi 1989) and hence the SAR estimates and total ^{137}Cs activities obtained. All these facts together emphasize that there is a need for standardized sediment sampling and subsampling methods, and this is especially important in monitoring work, where time trends need to be reliably evaluated from soft sediment bottoms.

Acknowledgements: The authors wish to thank all the collaborators. Especially we are grateful to Dr. Seppo Klemola and Dr. Tarja K. Ikäheimonen from the STUK, Dr. Jouni Lehtoranta from the Finnish Environment Institute, Dr. Mirja Leivuori from the Finnish Institute of Marine Research, Professor Terttu Vartiainen and Dr. Pirjo Isosaari from the National Public Health Institute (Department of Environmental Health) and Dr. Henry Vallius from the Geological Survey of Finland. For financial support we are grateful to Nordic Nuclear Safety Research, the Ministry of the Environment, the Academy of Finland and the Maj and Thor Nessling Foundation. We also thank the scientific staff and the crews of r/v *Aranda* and laboratory personnel of the NPP Environmental Laboratory at STUK.

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