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ACCUMULATION OF METALS IN THE SEDIMENTS IN THE GULF OF BOTHNIA AND IN THE GULF OF FINLAND

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ABSTRACT

Concentrations of heavy metals in the sediments of the Gulf of Bothnia and in the Gulf of Finland have been studied in the several sediment studies during 1991 - 1993. Mainly metal concentrations in the surface sediments are processed and some background concentrations are stated from the sediments deeper downcore. Studied metals are mercury, lead, cadmium, zinc, copper, iron, lithium, vanadium, titanium, arsenic, chromium and aluminium. The metal concentrations in the sediments in the different parts of the Gulf of Bothnia (the Bothnian Bay and the Bothnian Sea) are presented and compared with the concentrations in the Gulf of Finland. Some reasons for metal concentrations and differences in the concentrations are introduced and a few distribution patterns of metals are presented.

Keywords: metals, sediments, the Bothnian Bay, the Bothnian Sea, the Gulf of Bothnia, the Gulf of Finland.

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INTRODUCTION

Several studies about trace metals content in the sediment in the sea areas around Finland have been carried out in the last years (Tervo and Niemistö 1989, Ingri and Pontér 1986, Brüggmann and Lange 1989, Jankovski and Pöder 1980, Ott and Jankovski 1980, Borg and Jonsson 1996, Jonsson and Blomkvist 1992, Leivuori and Niemistö 1993, Leivuori and Niemistö 1995). In the latest sediment studies have been tried to clarify the metals distributions in the Gulfs of Bothnia and Finland. During studies 1991 - 1992 in the Gulf of Bothnia sediment samples were collected in a few places ranging from near coastal areas to the open sea sedimentation basins (Leivuori and Niemistö 1995). For the first time in 1992 the international sediment studies were allowed in the Russian territorial waters. During the sediment Baseline Study 1993 sediment stations in the Baltic Sea were sampled and some new sedimentation areas in the Estonia side of the Gulf of Finland were located. The purpose of the present study is to give a view of the metal concentrations in the sediments in the different parts of the Gulf of Bothnia and in the Gulf of Finland and discuss reasons for the metal distributions.

MATERIALS AND METHODS

Sediment samplings were performed during years 1991- 1993 in the Gulf of Bothnia and years 1992-1993 in the Gulf of Finland. The sampling stations are presented in Fig. 1. In the Gulf of Bothnia were 62 and in the Gulf of Finland 20 stations. The sediments were sampled with different samplers, a gravity corer for soft sediments (Niemistö 1974) and Gemini twin corer, on board R/V Aranda. In the Gulf of Bothnia were used also Box Corers and different corers used by the local water authorities in the coastal areas. Immediately after sampling the cores were dissected into 1 cm subsample slices. These slices were sealed in plastic petri dishes and stored frozen until freeze drying and analysing them. In the Gulf of Bothnia generally the five uppermost subsamples and in the Gulf of Finland the ten uppermost subsamples were analysed. Sediment cores taken during the Sediment Baseline Study were analysed into 20 / 25 cm depth.

The freeze dried and homogenated subsamples were digested by modified method of Loring and Rantala 1992. The sediment samples from the Gulf of Bothnia 1991-1992 were digested with aqua regia and hydrofluoric acid in special teflon bombs with an ordinary kitchen microwave oven (Leivuori and Niemistö 1994). To get more effective sample digestion procedures and more safety laboratory working a laboratory microwave oven CEM-250 was purchased in 1992. The sediment subsamples (0.4 g) were digested with 4 ml aqua regia (1:3 mixture of HCl and HNO₃), 2 ml hydrofluoric acid (40 %) and 5 ml H₂O (MQ-quality) in teflon bombs with the microwave oven CEM-250. The samples were heated with pressure regulation at 120 psi for 20 minutes. After cooling 12 ml boric acid (3 % m/v) added into the bombs and heated the samples with the microwave oven pressure controlled at 40 psi 10 minutes. The amples were cooled, diluted up to mark 50 ml in volumetric plastic flask, transferred into plastic store-bottles and stored in a room temperature until analysing them. For mercury analyses the samples were digested in all studies using same digestion procedure (Nordforsk 1975). The samples (1 g) were digested with 10 ml nitric acid (65 %) in glass bottles with an autoclave (30 min). After cooling added 40 ml H₂O (MQ-quality) into the samples

making total volume to 50 ml. The samples were stored in a room temperature in the glass digestion bottles. The used digestion methods represent repeatable, intercalibrated, and interrelated results, rather than to distinguish between the environmental effects of different biologically available fractions of metals, which also means that it is very difficult to point out differences between the anthropogenic load and the natural mineralogical background.

An ICP-AES instrument (TJA-25, Thermo Jarrell Ash) was used in the analyses of Cu, Ni, Ti, V, Zn, Mn, Al, Fe, Cr and Li. An AAS instrument Perkin Elmer 4000 (PE 4000) with a graphite furnace HGA-400 for the sediment samples of the Gulf of Bothnia 1991-92 and Perkin Elmer 5100 ZL for the other samples were used in the analyses of Pb, Cd and As. These elements were determined by a flameless AAS with the matrix modification and the platform techniques. Mercury was determined from the sediment samples of the Gulf of Bothnia with PE 4000 AAS by the cool vapour technique and from the other samples by the hybride technique with PE 5100 AAS combined with FIAS-400 and an amalgamation system. Carbon was analysed with a carbon-nitrogen analyzer (Heraeus CHN-O-rapid, Leivuori and Niemistö 1993) and by loss of ignition (SFS 1990, LOI = 2 x C).

Commercial sediment reference material SRM 2704 (NIST) and MESS-1 (NRCC) were used to control the analytical reliability. For studied elements nearly 100 % recoveries (84 - 110) are obtained for both reference materials. For vanadium the digestion appears to be somewhat incomplete for SRM 2704 (recovery 84 %), but for MESS-1 recovery is better (92 %). In case of lithium MESS-1 shows better recovery (107 %) than SRM 2704 (152 %), which mainly caused by the different matrices of the reference materials. Especially for titanium the new digestion procedure is much more better (recovery 97 %) than the one used in the earlier study (recovery 40 %, Leivuori and Niemistö 1994). By used two tail T-test founded differences for Al (P= 0), Li (P=0), As (P=0.0001), Ti (P=0), Hg (P=0.005) and Ni (P=0.003) in SRM 2704 between used different digestion and analytical methods. T-test result from Mess-1 confirm results from SRM 2704. These differences are mainly expected, because the later used procedure is more effective for silicate lattices associated metals, as Al, Fe, Li and Ti, and difficult dissolved chromium species.

RESULT AND DISCUSSION

The mean values of mercury, arsenic, zinc, and iron are higher in the surface sediments in the Bothnian Bay than in the Bothnian Sea and the Gulf of Finland (Table 1). In the Gulf of Finland are the highest mean values of cadmium and lead in the surface sediments. In the Bothnian Bay the mean concentrations of mercury and arsenic are even 3-4 times higher than in the Bothnian Sea. In the surface sediments in the Gulf of Finland the mean concentration of mercury is 2 times higher than in the Bothnian Sea, but lower than in the Bothnian Bay. The past loads of the Rönnskärsverken factory (Jonsson and Blomkvist 1992, Borg and Jonsson 1996) probably are the explanation for arsenic in the Bothnian Bay, but for mercury the reason can not be in the differences in the total load (2.0 tonnes year⁻¹ in the Bothnian Bay and 2.5 tonnes year⁻¹ in the Bothnian Sea, Enckell-Sarkola et. al. 1989, > 28 tonnes year⁻¹ in the Gulf of Finland, Helcom 1993). In case of mercury the reason might be in the higher content of organic material (Table 1) or the less abundant benthic population in the Bothnian

Bay and in the Gulf of Finland (Andersin et. al. 1977, Andersin and Sandler 1991). Also the recent artificial release of mercury into the Bothnian Bay by river transport from the huge inland reservoirs for hydroelectric power constructed in 1970' s (Leivuori and Niemistö 1993) might be the reason. In the Gulf of Finland also the high estimated total load influence the mean concentration of mercury. In the Bothnian Sea high concentrations of the specific contaminants titanium $4.91 \text{ mg g}^{-1} \text{ dw.}$ and vanadium $130 \text{ mg g}^{-1} \text{ dw.}$ are found in the sea area off Pori (Fig. 1), due to the Vuorikemia titanium dioxide factory.

In the Bothnian Bay the mean concentration of cadmium is two times higher than in the Bothnian Sea, both in the surface (0-1 cm, Table 1) and in the background (preindustrial, Table 2) layers. It has been suggested that cadmium is concentrated in the water phase in cold (Antarctic) waters (Niemistö and Perttilä 1995) by low primary production and inhibited sedimentation of organic material (Frew and Hunter 1992). In the Bothnian Bay the humic substances coming into the Bay via rivers might be a replacing factor for the strongly seasonal and relatively scarce primary production of organic material in this area. In the Gulf of Finland the mean concentration of cadmium is almost twice as in the surface sediments in the Bothnian Bay. In the deeper downcore 24-25 cm the concentrations of cadmium (Table 2) varied not so much in the different sea areas. Reasons might be the same humic substances as described above and also the higher estimated total annual load of cadmium into the Gulf of Finland (> 45 tonnes in the Gulf of Finland Helcom 1993 vs. 5.7 tonnes in the Bothnian Bay and 10 tonnes in the Bothnian Sea Enckell-Sarkola et. al. 1989). One reason might be also the correlation of the smaller abundance of benthic fauna with the higher content of organic matter and the correlation between organic matter and trace metals, including cadmium and mercury in marine sediments, e.g. Niemistö and Voipio 1981.

The distribution of mercury, lead and cadmium presented in Fig. 2. a.- c. show the higher contents of mercury in the surface sediments in the Bothnian Bay and lead and cadmium in the Gulf of Finland. For most the other studied elements there are no significant differences between the distribution of the contents in the Gulf of Bothnia. In the Gulf of Finland for some studied elements (e.g. Hg, Pb, Cd, Cr, Cu, Zn and Ni) a decreasing distribution was found in the direction of east to west along the Gulf. Arsenic and lead seem to accumulate in the deepest part of both sea areas of the Gulf of Bothnia as pointed out by Leivuori and Niemistö (1993). The correlation for accumulation effects are in the Bothnian Bay $r_{\text{Pb},35} = 0.880$, $r_{\text{As},31} = 0.817$, in the Bothnian Sea $r_{\text{Pb},28} = 0.739$, $r_{\text{As},14} = 0.739$ and in the Gulf of Finland $r_{\text{Pb},20} = -0.177$, $r_{\text{As},20} = 0.317$.

The vertical distribution of elements are dissimilar in the sediment cores in the different sea areas, e.g. in the station BO-3 in the Bothnian Bay for Pb, Hg, and Cd (Fig. 3). Also the maximum concentration of elements situated in the different depths of the sediment cores depending the sea areas. In the Gulf of Bothnia the maximum value situated between depths 5-9 cm and in the Gulf of Finland between 12 - 18 cm. These are partly due to the different rate of sedimentation: BO-3 325, EB-1 1.5, GF-2 1.2 and GF-6 $1.9 \text{ g m}^{-2} \text{ yr}^{-1}$ (Jensen 1995). The background levels are similar in the different sea areas, but for mercury levels are high in the eastern Gulf of Finland expressing high past

anthropogenic loads. For zinc the distributions varied markedly indicating large earlier loads into the Gulf of Finland. Also the natural mineralogical background influence the concentration levels of zinc in the Bothnian Sea (Koljonen 1992, Boström et. al. 1978) and in the Gulf of Finland (Emelyanov 1988).

In the Gulf of Bothnia the studied areas could be divided into the four subareas as presented by Leivuori and Niemistö 1995. The used data pool have updated with the data of the Sediment Baseline Study and some metal concentrations are presented in Fig. 4. In the Bothnian Bay in the deposition traps (area 1, Fig. 4) the maximum concentrations of mercury and zinc were found in the surface sediments off Kokkola (Table 1, Fig. 1). In the vicinity of Rönnskär (Fig. 1) higher values of mercury have been found ($1.36 \mu\text{g g}^{-1}$ dry weight, dw.), as well as for arsenic ($1\ 500 \mu\text{g g}^{-1}$ dw.) and lead ($148 \mu\text{g g}^{-1}$ dw., Jonsson and Blomkvist 1992 and Borg and Jonsson 1996), than what found in this study. In the Bothnian Sea in the area 1 high concentration of mercury was found in the study made by Piironen 1992, reflecting probably the effect of the industry along the river Kokemäenjoki (Fig. 1). In the Gulf of Bothnia the area 2 (Fig. 4) is characterized by erosion, transportation and non-deposition (Winterhalter 1972, Tulkki 1977). In this area the found concentrations do not differ from the concentrations in the coastal basins and estuaries.

In the transportation area 3 in the Gulf of Bothnia some high metal (cadmium, Fig. 4) concentrations were found, which indicate that at least temporarily some material may rest on this bottom type, flushing away as soon as suitable properties are established. The largest areas with present deposition are the central deep areas in the open sea in both the Bothnian Bay and the Bothnian Sea (area 4, Perttilä and Niemistö 1993). High concentrations of cadmium, lead and mercury were found in the surficial sediments in the open sea in the Bothnian Bay and arsenic in the open sea area off Raahel (Fig. 1). The sediment surface in these active accumulation basins are populated by a dense benthic fauna and some metals tend to be circulated in the biota (Sandler 1984). The bioturbation brings sediments into suspension which may be even transported outside the Gulf of Bothnia or selective entrapment of elements by microbial redox processes may take place in the abundant concretions (Boström et. al. 1981). In these basing filling areas the concentrations of some elements are smaller than in the sediments of metal loaded coastal and estuarine waters, even though they actually represent the major sinks for these trace metals.

In the Gulf of Finland the bottom structure is characterized by a greatly variable distribution of bottom types, making the bottom mosaic (Foldén and Winterhalter 1981). During the Sediment Baseline Study noticed strong bottom current transportation of fine sediment particles and even rolled off sediment surfaces in the Gulf (Perttilä and Niemistö 1993). Also the bottom sediment surface in the Gulf of Finland have been in the different situations depending on the appearance of benthic fauna, which have fluctuated strongly with the hydrographical conditions (Andersin and Sandler 1991). Sometimes the sediment surface has been uncontaminated and mainly after mid of 1980s there has been possibility of mixing modern contaminants and organic material in an old sediment surface by benthic fauna and their bioturbation.

The highest concentrations of heavy metals in the surface sediments (0-1 cm) concentrate mainly in the easternmost part of the Gulf of Finland, which is partly due to the discharges flow by the River Neva (Fig. 1) into the Gulf. In Fig. 5 are presented some mean concentrations of elements for some subgroups in the Gulf of Finland. The groups are defined according the areal and the bottom depth sharing: area I is the opean sea of the Gulf of Finland, II the open sea off Kotka, III the sea area off Vyborg Bay and Neva estuary and IV the sea area off Narva Bay (Fig. 5). The mean concentration of mercury is almost 3 times and cadmium 2 times higher in the areas II and III than in the areas I and II. The concentrations of lead, zinc and copper are also higher in the same areas. The highest value of cadmium (Table 1) founded in the area II near Gogland island (F 44, Fig. 5). In the easternmost station of the Gulf (F 40, area III) founded the highest concentrations of mercury and lead (Table 1). In the area III (GF 6) founded the highest concentrations of copper and for zinc founded the highest value in the area II (XV 1). In the Gulf of Finland the data pool is quite difficult to handle due to the phenomena discuss above. More detail data of heavy metals in the sediments in the eatuaries of Finland, Neva and Estonia are needed to clarify the spreadness of metals in the Gulf of Finland. The results of heavy metals from the sediment samples inside and outside St. Petersburg's flood protection dam (in the Kotlin Island, Fig. 5) from 1990 - 92 (unpublish data, Rybalko et. al. 1993) shows that some part of loads of elements (e.g. Pb,Cd, Zn) by-pass the dam and find their way into the Gulf.

In the Gulf of Bothnia the slight enrichment of elements in the deeps seem to follow the basin filling sedimentation prevailing in the whole Gulf. In the Gulf of Finland similar behaviour was not recognized although sedimentation seem to follow also basin filling (Winterhalter 1992). The higher concentrations of organic material and the sulphidic environment in the Baltic Proper sediments might cause for some elements e.g. cadmium and lead higher values in the Baltic proper than in the Gulf of Bothnia. Also metal budget calculation based on discharge and sedimentation data indicates that only lead probably accumulate totally in the Gulf of Bothnia and mercury, cadmium, copper and zinc might transport into the Baltic Proper (Leivuori and Niemistö 1995).

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TABLE 1. The results of some heavy metal concentrations in the surface sediments (0-1 cm) in the Bothnian Bay, the Bothnian Sea and in the Gulf of Finland 1991-1993 with the statistical specifications (dry weight basis).

The Bothnian Bay (0-1 cm)	DM(%)	N(%)	C(%)	Al(%)	Cu($\mu\text{g/g}$)	Fe(%)	Li($\mu\text{g/g}$)	Mn(%)	Ni($\mu\text{g/g}$)	Ti(mg/g)	V($\mu\text{g/g}$)	Zn($\mu\text{g/g}$)	Pb($\mu\text{g/g}$)	Cd($\mu\text{g/g}$)	As($\mu\text{g/g}$)	Hg($\mu\text{g/g}$)
Mean	17.7	0.37	3.66	4.8	32	5.5	19	0.42	35	3.13	62	202	33	0.59	100	0.21
Standard Deviation	11.6	0.17	1.67	0.8	16	1.8	12	0.47	11	0.5	22	114	30	0.36	96	0.24
Median	12.8	0.35	3.35	4.7	26	5.7	14	0.19	33	3.07	57	160	17	0.50	48	0.12
Minimum	3.0	0.12	1.06	3.2	13	2.6	10	0.05	15	2.08	24	50	11	0.19	8	0.02
Maximum	45.4	0.85	8.31	6.6	80	10.1	60	1.80	58	4.27	120	550	121	1.98	320	0.97
Range	42.4	0.73	7.25	3.4	67	7.5	50	1.75	43	2.19	96	500	110	1.79	312	0.95
Count	32	31	31	35	35	35	32	35	35	35	35	35	35	35	31	35
The Bothnian Sea (0-1 cm)																
Mean	42.8	0.22	1.68	5.4	30	4.3	37	0.72	42	3.14	70	143	31	0.31	33	0.06
Standard Deviation	20.6	0.17	1.18	1.2	11	1.9	25	1.41	22	1.28	35	61	14	0.26	28	0.04
Median	36.5	0.12	0.82	5.1	26	4.5	30	0.20	41	3.06	77	145	28	0.26	2	0.06
Minimum	13.1	0.03	0.34	4.1	10	1.2	12	0.01	14	1.03	16	30	14	0.04	4	0.01
Maximum	72.0	0.62	4.29	8.3	44	7.8	95	6.12	110	5.27	138	240	60	1.28	85	0.15
Range	58.9	0.59	3.95	4.2	34	6.6	83	6.11	96	4.24	122	210	46	1.24	81	0.14
Count	24	24	24	28	24	28	28	28	28	28	28	28	28	28	14	28
The Gulf of Finland (0-1 cm)																
Mean	12.3	6.98	7.82	7.6	43	4.5	63	0.51	42	3.97	77	175	49	1.00	15	0.14
Standard Deviation	4.4	2.20	1.88	1.1	9	0.6	8	0.46	9	0.57	10	39	13	0.50	4	0.08
Median	11.3	1.84	2.27	7.2	44	4.7	62	0.36	40	3.84	77	179	48	0.94	15	0.11
Minimum	7.5	3.68	4.53	6.0	27	3.0	52	0.06	25	2.97	57	107	26	0.34	8	0.05
Maximum	24.1	11.26	11.25	9.9	57	5.4	77	2.00	60	5.19	96	236	80	2.19	28	0.32
Range	16.6	7.58	6.72	3.9	30	2.5	25	1.94	35	2.22	38	129	54	1.85	20	0.27
Count	20	10	10	20	20	20	20	20	20	20	20	20	20	20	20	20

TABLE 2. The mean concentrations of some measured trace metals in the 24-25 cm and in the preindustrial (depth < 10 cm) sediments in the Bothnian Bay, in the Bothnian Sea and in the Gulf of Finland (dry weight basis, n means number of samples).

SEA AREA		Cu $\mu\text{g g}^{-1}$	Cr $\mu\text{g g}^{-1}$	Ni $\mu\text{g g}^{-1}$	Fe %	Mn $\mu\text{g g}^{-1}$	Zn $\mu\text{g g}^{-1}$	Pb $\mu\text{g g}^{-1}$	Cd $\mu\text{g g}^{-1}$	As $\mu\text{g g}^{-1}$	Hg $\mu\text{g g}^{-1}$	Number of samples /Study
Bothnian Bay	24-25	35 \pm 4	41 \pm 35	48 \pm 9	4.9 \pm 0.2	1.72 \pm 0.38	99 \pm 7	30 \pm 8	0.32 \pm 0.14	12 \pm 3	0.01 \pm 0.01	n=2
	Preind.	27 \pm 7	37 \pm 7	32 \pm 18	5.1 \pm 4.0	1.0 \pm 1.0	71 \pm 21	4.2 \pm 2.9	0.37 \pm 0.27	6 \pm 5	0.02 \pm 0.01	n=10 Borg and Jonsson 1996
Bothnian Sea	24-25	44 \pm 4	97 \pm 5	58 \pm 6	6.0 \pm 0.7	1.18 \pm 0.36	157 \pm 25	37 \pm 6	0.48 \pm 0.16	17 \pm 2	0.03 \pm 0.01	n=2
	Preind.	36 \pm 5	40 \pm 7	36 \pm 8	4.7 \pm 1.1	3.2 \pm 1.8	132 \pm 17	24 \pm 6	0.10 \pm 0.07	9 \pm 2	0.03 \pm 0.01	n=5 Borg and Jonsson 1996
The Gulf of Finland	24-25	34 \pm 5	80 \pm 7	39 \pm 7	4.7 \pm 0.6	1.07 \pm 0.29	146 \pm 33	39 \pm 13	0.41 \pm 0.18	8 \pm 4	0.11 \pm 0.1	n=4



Fig. 1. The sampling stations in the Baltic Sea 1991 - 1993.

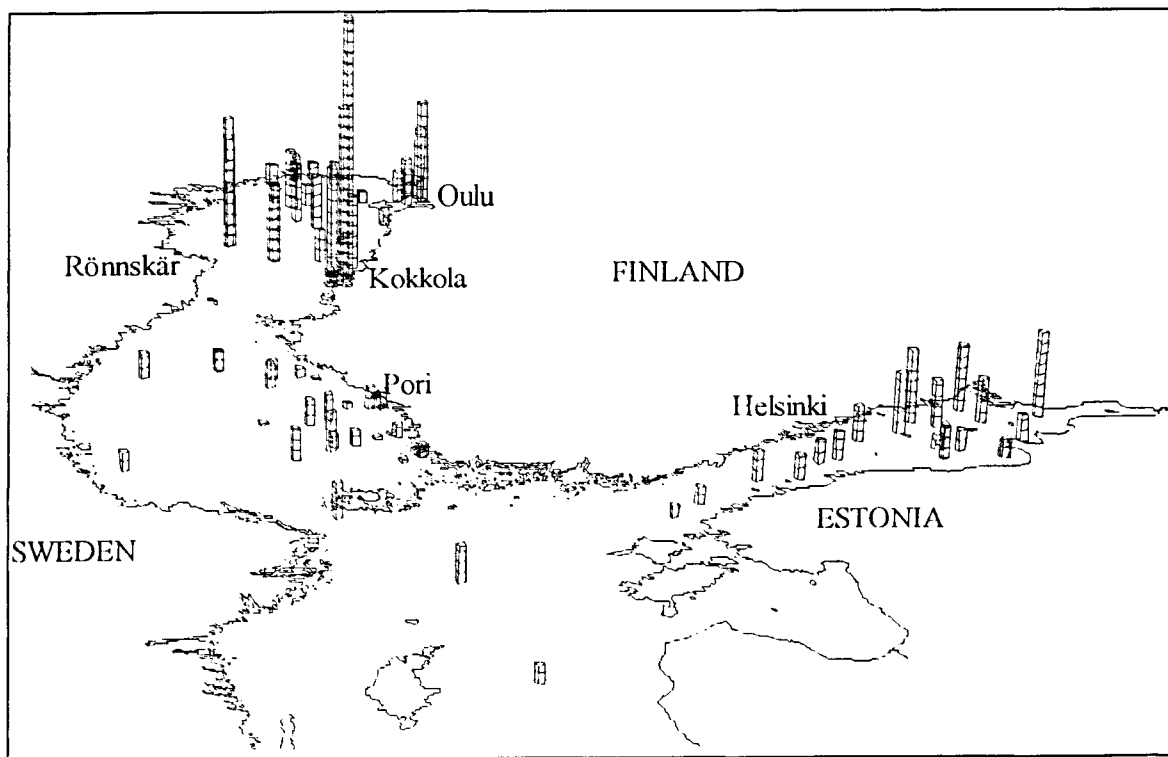


Fig. 2 a. Mercury in the uppermost 1 cm of the sediments of the Baltic Sea. 1 unit = $0.05 \mu\text{g g}^{-1}$ dry weight basis.

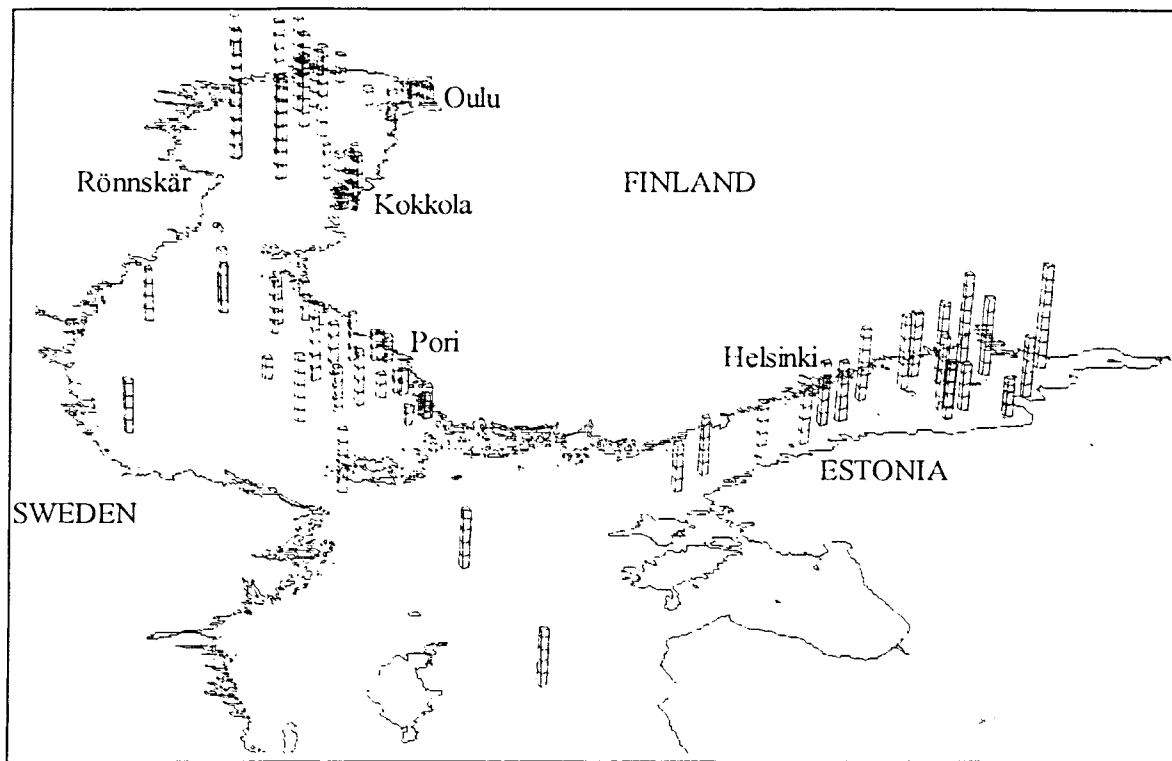


Fig. 2 b. Lead in the uppermost 1 cm of the sediments of the Baltic Sea. 1 unit = $10.0 \mu\text{g g}^{-1}$ dry weight basis.

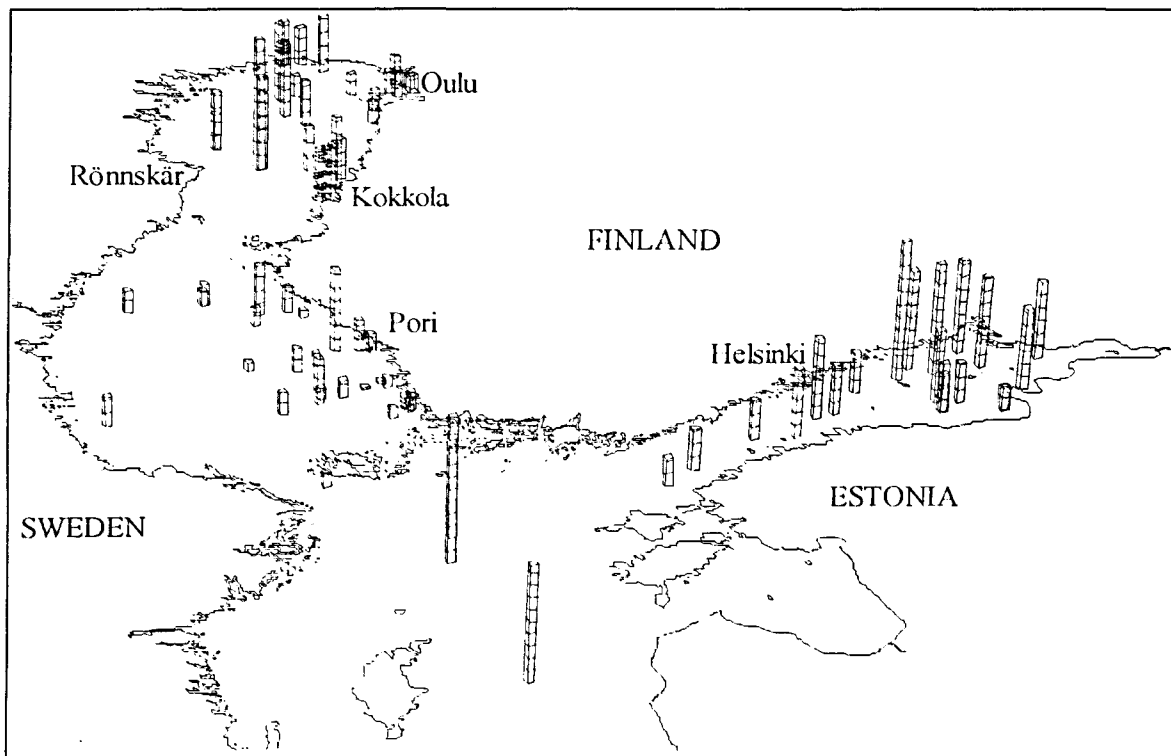


Fig. 2 c. Cadmium in the uppermost 1 cm of the sediments of the Baltic Sea. 1 unit = $0.20 \mu\text{g g}^{-1}$ dry weight basis.

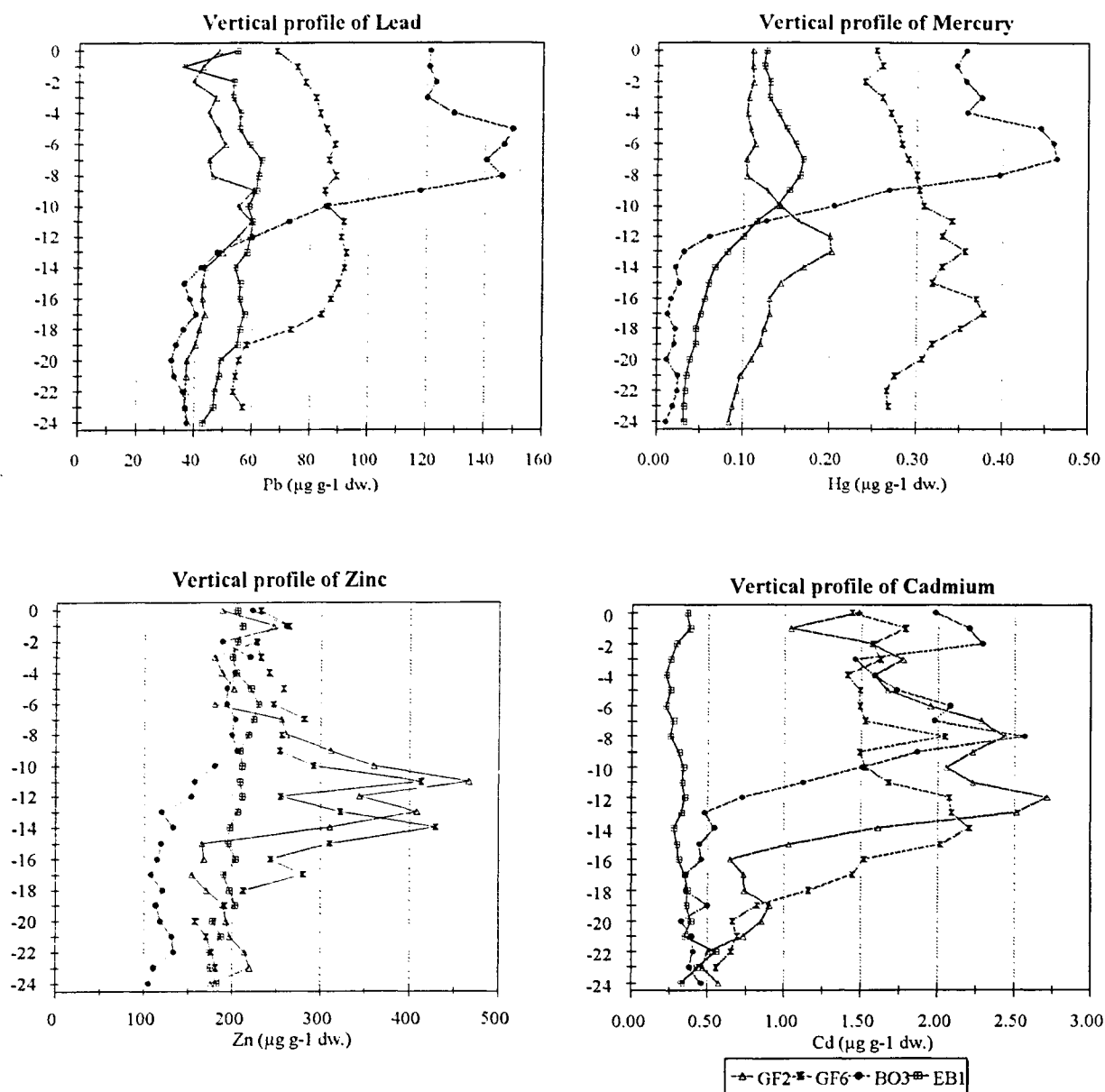


Fig. 3. The vertical distribution of some elements in the Baltic Sea. The stations are GF 2 (59.505, 25.5191) in the middle of the Gulf of Finland, GF 6 (60.2041, 28.0045) in the eastern part of the Gulf of Finland, EB 1 (60.5935, 19.4395) in the Bothnian Sea and BO 3 (64.1876, 22.1920) in the Bothnian Bay.

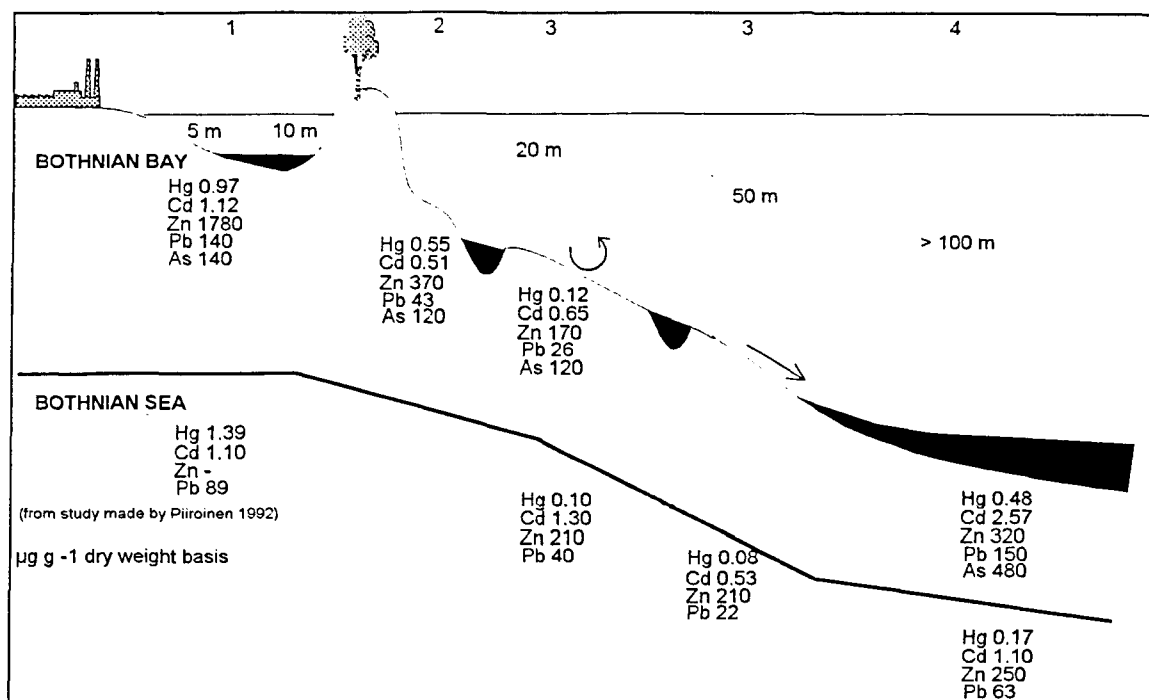


Fig. 4. Some example concentrations of elements ($\mu\text{g g}^{-1}$ dry weight) in the sediment in the Bothnian Bay and Bothnian Sea. 1. Deposition traps in the coastal and estuarine basins, 2. The occasional sedimentation basins in the nondeposition areas with water depth < 60 m, 3. The transport and erosion bottoms and 4. The sedimentation basins in the open sea area with water depth > 60 m (modified Leivuori and Niemistö 1995).

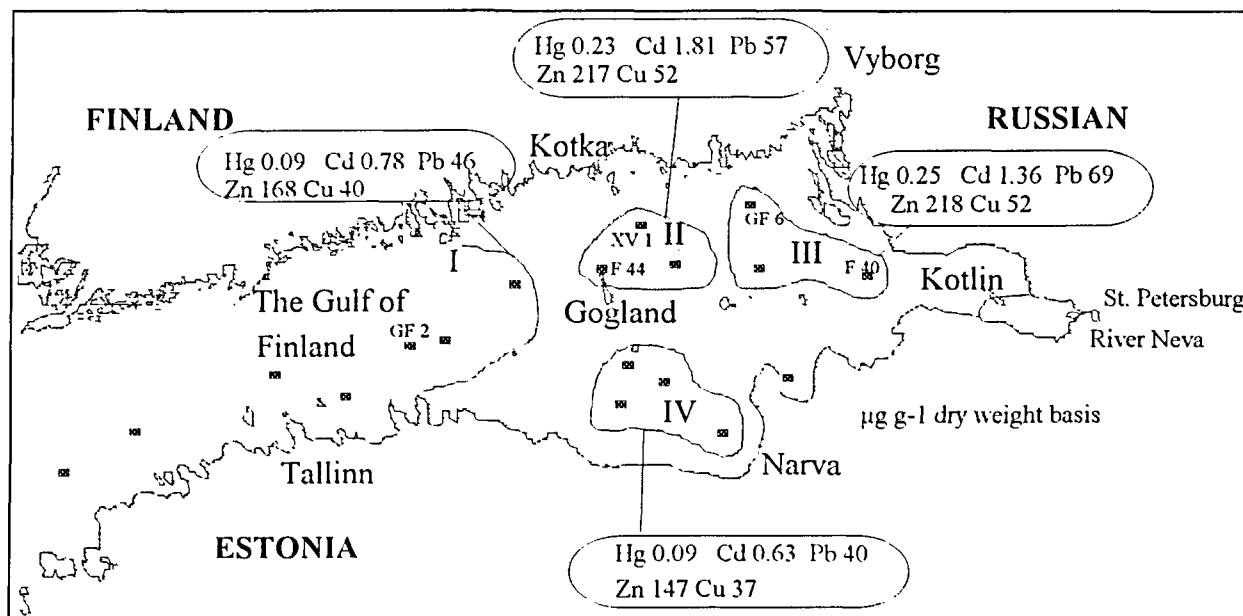


Fig. 5. The mean concentrations of some elements ($\mu\text{g g}^{-1}$) in the surface sediments (0-1 cm) in the Gulf of Finland. Areas: I the open sea the water depth > 60 m, II the open sea area off Kotka with $60 < \text{water depth} < 80$ m, III the sea area off Vyborg Bay and Neva estuary with water depth < 60 m, IV the sea area off Narva Bay with $30 < \text{water depth} < 75$ m.