The distribution of heavy metals and arsenic in recent sediments in the Gulf of Finland

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A large number of sediment samples representing most of the active mud accumulation areas of the Gulf of Finland were studied for total cadmium, chromium, lead, mercury, arsenic, copper and zinc. The highest concentrations of metals were found in sediment surfaces in the easternmost part of the Gulf, which is influenced by the inflow of the Neva river and the loads of the city of St. Petersburg. Vyborg Bay and the Kymi River also clearly affect the off-shore sea bottom. The vertical distributions determined in four sediment cores show for all metals overall decreasing concentrations towards the surface of the sediment cores, which in part indicates decreased load of metals. Evidently levels of copper, lead and mercury are still decreasing, while cadmium and arsenic levels are slowly increasing in the eastern Gulf of Finland. From the mean sedimentation rate of wet suspended particulate matter (SPM) and dry weight of the surficial sediment, an annual accumulation of 8.7×10^6 t a⁻¹ of SPM was estimated for the Gulf. With that estimate the annual total accumulation of elements into the sediments was calculated. The annual accumulations of cadmium and lead are clearly higher in the Gulf of Finland than in the Gulf of Bothnia. Accumulation of the other metals is somewhat higher in the Gulf of Finland, while the accumulation of arsenic is clearly lower, only one fifth of that in the Gulf of Bothnia.

Introduction

The Gulf of Finland is a narrow basin of the main Baltic Sea. Depth decreases from west to east, and surface salinity from 6 psu to nearly freshwater conditions in the same direction (Fig. 1). Because it is a direct continuation of the Baltic Proper, the saline water of the Baltic deep flows freely in assuring that the basic hydrographical changes in the Baltic Proper are reflected in the Gulf. High freshwater inflow is an additional influence on the hydrographical conditions. By far the most significant inflow is from the east, from the river Neva (ca. 80 km³ a^{-1}), which flows through the city of St. Petersburg. The river has a clear polluting effect over a large part of the eastern Gulf. Other main loads entering the Gulf are of atmospheric origin or from point sources along the Finn-



Fig. 1. Bathymetry of the Gulf of Finland. Modified from Seifert and Kayser (1995).

ish, Russian and Estonian coasts (HELCOM 1993, 1997).

Sediments act as reservoirs of emitted elements recording the concentrations of metals and changes in emissions over the years. The earlier studies of heavy metals in the sediments of the Gulf of Finland have covered small areas or just a few stations (Jankovski and Pòder 1980, Ott and Jankovski 1980, Tervo and Niemistö 1989, Vallius and Lehto 1998). Since the Gulf of Finland is heterogeneous with active sedimentation basins of different shape and size scattered throughout the Gulf area, a reliable picture of element distributions in the sediments could only be obtained by increasing the number of studied basins. Thus the aim of our research was to study the total concentrations of heavy metals in most of the active mud accumulation basins in the open sea of the whole Gulf of Finland. By adding the metal data by Leivuori (1998) and mercury data by Vallius (1999) to the earlier unpublished data of this study, the present paper incorporates all the latest research, thus providing the first extensive picture of the horizontal distribution of the selected elements in the sediments of the Gulf. The total concentrations of the environmentally interesting elements cadmium, chromium, lead, zinc, arsenic, mercury and copper are reported here.

Additionally, sediment cores from four sites

along the Gulf were examined vertically to asess the change in metal loads through the last decades. This development was of interest since it was expected that the recent slow-down in industrial production in Russia would show up as a decrease in metal concentrations in the cores. The data on the mean metal concentrations in the topmost centimetres of the sediment cores were used in accumulation calculations, and from an average of the mean annual accumulation of suspended particulate matter annual total accumulations of elements in the sediments were estimated for the whole Gulf.

Material and methods

Sedimentary basins

The Finnish coast is part of the Fennoscandian Shield and the bedrock consists of hard crystalline rocks that during repeated glaciations have been abraded into a system of scattered hillocks and valleys. The valleys have later been filled by glacial and postglacial sediments and now appear as small isolated heterogeneous basins of recent sedimentation. Along the southern coast of the Gulf of Finland, as well as at its eastern end, the bedrock consists of younger Paleozoic sedimentary rocks (< 1



Fig. 2. Locations of the sampling stations in the Gulf of Finland.

Ma). These rocks are softer and much more homogeneous than the Precambrian rocks (>1.6 Ma) and have been evenly worn down by the glaciations. Thus the sea basins of glacial and postglacial sediments in adjoining parts of the Gulf are larger and much more homogeneous than to the north and west. The topmost part of the basins is usually covered by a layer of recent sediments, which are poorly compacted (Winterhalter 1992), laminated or bioturbated muddy clays or silts. The thickness of the recent sediments in the Gulf of Finland varies widely even within basins (e.g. at station GF2, Figs. 2 and 3) (Vallius et al. 1996, Vallius 1999b). Very often the thickness is less than one metre, but occasionally it exceeds five metres. In some basins the sedimentation rate is more or less even throughout e.g. at station F41 (Fig. 2), while in others it varies significantly (e.g. at station GF2, between 2.5 and 8.0 mm a⁻¹) (Vallius 1999b).

Sampling and sample handling

Sampling was mainly carried out in the accumulation basins of the open sea where the basins can be assumed large enough $(1 \times 1 \text{ NM or greater})$. Basins with a radius of only some tens or hundreds of metres were considered too small to receive fresh sediment in all conditions. In order to find the most suitable sampling sites the bottoms of all stations were checked by echosounding before sampling (12 kHz, Atlas Deso-25).

Sediments were sampled with different kinds of gravity corers with a core diameter of 6 cm (Niemistö 1974) or greater. The samples were collected at the stations shown in Fig. 2 between 1992 and 1995 during cruises arranged by the VSEGEI (All Russia Geological Research Institute) for the Finnish-Russian MEP (Marine Ecological Patrol) programme and during cruises of FIMR's (Finnish Institute of Marine Research) R/V Aranda and FEI's (Finnish Environmental Institute) R/V Muikku. The total amount of stations is 38 and because of several parallel samples the total number of cores analysed is 50. Immediately after sampling the cores were sliced into 1 cm subsamples, usually down to 10-25 cm depth, and stored frozen (-20 °C) in plastic bags or containers. The samples consisted of recently deposited muddy silt, muddy clay or a mixture of these. The surface sediment (between 0 and 4 cm depending on site) was brown in basins with oxidized near bottom waters and downcore usually grey, greenish grey or black, indicating reducing conditions. Only at one site, V1, the whole sediment column was anoxic (1994 and 1995).

Analyses

On board R/V Aranda and the Russian vessels the cores were analysed by the method of Kyzyurov



Fig. 3. Bathymetry of station GF2 area with thickness of the total sediment cover and postglacial sediments indicated (compiled by Dr. Boris Winterhalter, GSF).

et al. (1994), which uses the gammaspectrometrically detectable ¹³⁷Cs activity to mark off the depth of April 1986 (Chernobyl nuclear power plant accident). These datings have been of significant importance in determining sedimentation rates in the basins of the Gulf. Results of 98 datings, including the data from this study, are presented by Kankaanpää *et al.* (1997). The oxygen concentrations in the near bottom waters were also measured during the cruises, on R/V Aranda with the method described by Grasshoff (1983).

After the cruises the frozen sediment samples were immediately transported to the laboratories for chemical analysis. The sediment cores were analysed from the surface (0–1 cm layer) down to a depth of 10–25 cm. At the Geological Survey of Finland (GSF) the sediment samples (30 stations) were freeze-dried and sieved to grainsize < 2 mm in order to remove large objects, such as plant parts and benthic animals. At FIMR the samples (20 stations) were freeze-dried and handled as bulk samples, from which large objects were removed by tweezers. In both laboratories, samples were mechanically homogenized and analysed for total concentrations of heavy metals and arsenic.

At GSF the samples (0.2 g) were digested with hydrofluoric acid (10 ml) and perchloric acid for 24 hours and evaporated to dryness on a hotplate. The residue was dissolved in perchloric acid (2 ml), evaporated to dryness, dissolved in nitric acid and diluted with water. Element determinations were made with an ICP-MS (Perkin-Elmer Sciex Elan 5000). For mercury a microwave-assisted (CEM MDS-2000) nitric acid digestion method was applied (EPA 1990) and the samples were analysed with a Hg-spectrometer FIMS-400 (Perkin Elmer).

At FIMR the samples were dissolved by a modification of the method of Loring and Rantala (1992) as presented in more detail by Leivuori (1998). The samples (0.4 g) were digested with aqua regia (4 ml), hydrofluoric acid (2 ml) and boric acid (12 ml, 3% m/v) in a microwave oven CEM-205, except for mercury analyses where they (1 g) were digested with nitric acid (10 ml) in an autoclave (Nordforsk 1975). A Perkin Elmer 5100 ZL AAS-instrument was used in the determinations of lead, cadmium and arsenic. Mercury was determined by a hybrid technique with a PE 5100 AAS combined with a FIAS-400 and an amalgamation system. An ICP-AES instrument (TJA-25) was used in the measurement of other elements.

The digestion methods that were used give repeatable and intercalibrated total concentrations of elements, which are suitable for distribution studies, but do not easily allow the anthropogenic load to be distinguished from the natural mineralogical background.

Analytical control and laboratory intercomparison

In both laboratories commercial certified reference materials were included in every sample batch as a check on the analytical reliability. At GSF the analytical reliability was checked by measurements on the commercial certified sediment reference material SRM 2704 (NIST, National Institute of Standards and Technology) or rock sample SY-2 (CCRMP, Canadian Certified Reference materials project). At FIMR the analytical reliability was checked with the reference materials SRM 2704 (NIST), MESS-1 and BEST-1 (NRCC, National Research Council of Canada). For the elements studied, recoveries of 61%–108% were obtained for SY-2 and recoveries of 84%-110% for MESS-1 plus BEST-1 (Hg). As the SRM 2704 was used in both laboratories it could be used for intercomparison of the analytical methods. Recoveries were 78%-113% at GSF and 91%–109% at FIMR (Table 1). The data show that the analytical results are comparable. The analytical method for chromium used at GSF appears to be somewhat deficient, perhaps due in part to the matrix of the reference material.

Results and discussion

Table 2 shows the concentrations and some statistical parameters of metals in the surface sediments (0-1 cm) of the Gulf of Finland. The values presented are an average over all stations at which measurements were made and thus give an estimate of heavy metal levels over the whole Gulf. In further handling they can be used for annual accumulation and other estimations. The locations of the stations for which maximum or minimum values are given in Table 2 are shown in Fig. 2.

Horizontal distribution of the elements

The horizontal distributions of the elements exhibit clear trends in the surface sediments of the Gulf of Finland, as shown for selected elements in Fig. 4. Concentrations of all metals investigated are high in the east and decrease towards the west and in the deeper waters of the Gulf. The pattern of decrease towards the west differs for the various elements, however. Arsenic (Fig. 4a) has a more complicated pattern, with an overall low (6–28 mg kg⁻¹ dry weight basis, d.w.) close to background value and slightly anomalous values only in the middle of the Gulf.

	Cert. Value (mg kg ⁻¹ d.w.)	GSF mean (mg kg⁻¹ d.w.)	GSF SD	GSF recovery (%)	GSF count	FIMR mean* (mg kg ⁻¹ d.w.)	FIMR SD	FIMR recovery (%)	FIMR count
		((,-)		((,-)	
Pb	161 ± 17	156	13	97	18	175	11	109	55
Cd	3.45 ± 0.22	3.47	0.29	101	18	3.13	0.34	91	55
As	23.4 ± 0.8	20.3	1.6	87	18	24.2	1.7	103	55
Hg	1.44 ± 0.07	1.63	0.14	113	54	1.39	0.08	94	31
Zn	438 ± 12	431	27	98	18	437	18	100	56
Cr	135 ± 5	106	8	78	18	125	9	93	70
Cu	98.6 ± 5.0	97.6	6.9	99	18	103.8	6.2	105	67

Table 1. Recovery of analytical procedure with standard deviations (SD) for SRM 2704 at GSF and FIMR, related to total metal concentrations of certified reference sediments.

* Leivuori (1998).

The highest concentrations of most of the metals occur in the Neva Estuary, which can be divided into the Inner and Outer Estuaries (Pitkänen 1994) (Fig. 4b-f: Table 2). Cadmium concentrations (Fig. 4b) are high in the whole Neva Estuary (1.20–2.00 mg kg⁻¹ d.w.) but even higher towards the west $(1.71-2.19 \text{ mg kg}^{-1} \text{ d.w.})$. Values of chromium, in turn, are highest in the Neva Estuary (113–138 mg kg⁻¹ d.w.; Fig. 4c), clearly lower (71–101 mg kg⁻¹ d.w.) in the sea between the Neva Estuary and Lavansaari, which can be considered as the next hydrological region and lower still in the rest of the Gulf (< 102 mg kg⁻¹ d.w.). Copper (Fig. 4d) is present in varying concentrations in the eastern Gulf, with higher concentrations in the Inner Neva Estuary (56-63 mg kg⁻¹ d.w.) and along the northeastern coast (48–63 mg kg⁻¹ d.w.). This indicates large-scale mobility. Mercury concentrations (additional stations from Vallius 1999a) are clearly highest in the Neva Estuary $(0.213-0.392 \text{ mg kg}^{-1} \text{ d.w.}; \text{ Fig.})$ 4e); they decrease more or less evenly to Suursaari, and still more from there westwards (< 0.180 mg kg⁻¹ d.w.). Lead (Fig. 4f) is fairly evenly distributed within the Neva Estuary (70–88 mg kg⁻¹ d.w.) and from there westward the concentrations decrease ($< 71 \text{ mg kg}^{-1} \text{ d.w.}$).

Altogether the horizontal distributions of the metals seem to follow the different hydrological regions in the eastern Gulf of Finland. Also, the natural submarine barriers at Seiskari and Lavansaari cause the currents to flow west along the north coast of the Gulf (Fig. 1). Metals are additionally released into the eastern Gulf of Finland from the Kymi river and Vyborg Bay (Fig. 1). Copper cadmium, lead and mercury show higher concentrations at sites along the northern coast of the Gulf. Metals from Luga Bay migrate to the deeper channel north of Lavansaari on their way to the western, deeper part of the Gulf, since the channel between the mainland and Lavansaari is very narrow (Fig. 1). The concentrations of metals in Luga Bay sediments are relatively low (e.g. at station GF5 in Fig. 2: cadmium 1.27, lead 47 and mercury 0.103 mg kg⁻¹ d.w.). Concentrations in sediments in Narva Bay (Fig. 1) are not very high for any metals even though the notorious Sillamäe oil-shale power plant is located close to the shore of the Bay.

According to measurements made on board R/V Aranda the oxygen conditions of the Gulf were satisfactory (> 2.5 mg l^{-1}) throughout the study period. An almost total loss of oxygen was though noted at some sites north of Suursaari in 1996 (Pitkänen and Välipakka 1997). Oxygen variations may partly explain the distribution of the elements. The concentrations of metals like chromium, which under oxic conditions are easily bound into the sediment (Petersen et al. 1996), decrease rapidly beyond the Neva estuary. Others, like copper, cadmium and zinc, which are partly released by decomposition of organic substances from freshly deposited material under oxic conditions (Petersen et al. 1996), may easily migrate westward into deeper parts of the Gulf waters, and thus explain their more even distribution in the eastern part of the Gulf.

Usually it is thought that high metal concentrations are related to the high organic carbon content in the sediments. The relationship between the total carbon and the distribution of metals in the Gulf is under further investigation, but observations during the present study seem to confirm findings presented by Leivuori (1998) that in many

Table 2. Concentrations and descriptive statistics for studied elements in the surface sediments (0–1 cm). Concentrations on dry weight basis. For location of stations, *see* Fig. 2.

	Cr (ma ka ⁻¹)	Cu (ma ka ⁻¹)	Cd (ma ka ⁻¹)	Zn (ma ka ⁻¹)	Pb (ma ka ⁻¹)	Hg (ma ka ⁻¹)	As (ma ka ⁻¹)
	(((((((
Mean	87	43	1.23	209	52	0.182	14
Standard error	3	2	0.09	13	3	0.012	1
Median	84	44	1.28	205	50	0.176	14
Standard deviation	22	11	0.58	87	17	0.087	5
Maximum (station)	138 (V4)	63 (V4, V14)	2.19 (F44)	513 (SL32)	88 (V4)	0.392 (V4)	28 (LL3A)
Minimum (station)	57 (E1)	25 (E1)	0.28 (E1)	77 (E2)	26 (GF4)	0.048 (F62)	6 (V4, E2)
Count	43	43	42	42	43	50	41



Fig. 4. Distribution of arsenic (a), cadmium (b), chromium (c), copper (d), mercury (e), and lead (f) in recent surface sediments (0-1 cm). The scale is logarithmic and the difference between sampling sites is important, not the actual value at each specific site. Unit mg kg⁻¹ dry weight.

cases there is a poor correlation between metal concentrations and total carbon content in the surface sediments.

Vertical distribution of the elements

Different distribution patterns were found in the vertical profiles of the elements. The changes in emissions during recent decades can easily be seen with the help of the gammaspectrometrically (¹³⁷Cs) determined depth of 1986, as indicated in Figs. 5 and 6. The uppermost parts of the sediment in the Neva Estuary contain overall higher concentrations of heavy metals than cores from the western part of the Gulf. The lead concentra-

tions in the sediment profile at station SL2 (Neva Estuary, Fig. 5), where concentrations were highest in the 1970s and 1980s, have now decreased to three quarters of the 1986 level. Concentrations of copper and mercury decrease also while cadmium is slightly increased in the upper part of that core. The vertical profiles from other parts of the Gulf show decreases in the element concentrations of almost all metals towards the top of the cores, evidently in part due to decreased loads. Copper, lead and mercury are still decreasing, cadmium and arsenic seem to be increasing slowly and levels of zinc remain more or less stable.

In the vertical profiles in Figs. 5 and 6, the maximum peaks of the metals indicate concentrations two to at least five times higher than the



Fig 5. Vertical distributions of arsenic, copper and lead in recent surface sediments (*see* stations in Fig. 2). The deposition in 1986 is marked with an arrow. At the GF stations the sediment was oxydized down to depths between 1 and 3 centimetres and at station SL2 down to 0.5 cm. Unit mg kg⁻¹ dry weight.

concentrations at the bottom of the cores. The lowest concentrations found during this study are comparable to the the concentrations in the lowermost part of the GF1 core (24–25 cm): mercury 0.018, zinc 100, cadmium 0.11, lead 21, arsenic 10 and copper 25 mg kg⁻¹ d.w.

The decrease in the deposition of metals in the eastern parts of the Gulf of Finland can in part be attributed to the present slow-down in industrial production in Russia. Another factor may be the mechanical-biological Central Aeration Plant in St. Petersburg, which began operations in the early 1980s and which in 1990 had a capacity of 1 500 000 m³ d⁻¹ (NoPEF 1990). The Kotlin dam (Fig. 1) in Neva Bay just outside St. Petersburg has probably also played a role since the concentrations of metals at the stations (e.g. SL2 in Figs. 5 and 6) outside Kotlin appear to have peaked immediately before the dam construction was begun (1979). A source of bias might be the dilution caused by the increased amount of organic matter in the top layer of sediment coming from the profuse algal blooms of recent years (Pitkänen 1994); on the other hand, one would expect the



Fig. 6. Vertical distributions of mercury, cadmium and zinc in recent surface sediments (*see* stations in Fig. 2). The deposition in 1986 is marked with an arrow. At the GF stations the sediment was oxydized down to depths between 1 and 3 centimetres and at station SL2 down to 0.5 cm. Unit mg kg⁻¹ dry weight.

deposition of heavy metals to increase in pace with the deposition of organic matter.

Comparison with the Gulf of Bothnia

Comparison of the mean concentrations of elements in surface sediments in the Gulf of Finland and the Gulf of Bothnia confirms earlier observations (Leivuori 1996, Leivuori 1998). In the case of cadmium, for example, the mean concentration in the Gulf of Finland is about twice that in the Gulf of Bothnia. Reported maximum values of 0.392 mg kg⁻¹ d.w. for mercury, 513 mg kg⁻¹ d.w. for zinc, and 138 mg kg⁻¹ d.w. for chromium are higher than earlier reported (0.32, 243 and 117 mg kg⁻¹ d.w., respectively, Leivuori 1998). The maximum concentration of mercury in the Gulf of Finland is slightly lower than the maximum value in the accumulation basins of the Bothnian Bay (0.48 mg kg⁻¹ d.w., Leivuori 1998) and 50% of the maximum concentration of 0.97 mg kg⁻¹ d.w. in a near-shore area in the Bothnian Bay (Fig. 1; Leivuori and Niemistö 1993).

4. Annual deposition of heavy metals and a comparison with the Gulf of Bothnia

According to the estimates of Kankaanpää et al. (1997), the average sedimentation rate of wet suspended particulate matter (SPM) in accumulation basins in the Gulf of Finland is 6 mm a⁻¹. The mean dry weight of the surficial sediment in the Gulf of Finland is 12.3% of wet weight (Leivuori 1998) and since the wet sediment density is 1.2 g cm⁻³, we calculate a mean accumulation rate of 886 g m⁻²a⁻¹ particulate matter. This rate is equal to an accumulation of 8.7×10^6 ta⁻¹ of SPM, assuming sedimentation basins to cover 1/3 of the total area of the Gulf of Finland (Voipio 1981). Annual accumulations in tonnes per year in the Gulf of Finland and, for comparison, in the Gulf of Bothnia, are shown in Table 3. Estimates from an earlier study of Borg and Jonsson (1996) for the Gulf of Finland are also included. The estimates show clearly higher annual total accumulation of cadmium (over 2 times) and lead (over 1.5 times) in the Gulf of Finland than in the Gulf of Bothnia. Total accumulation of the other metals is somewhat higher in the Gulf of Finland, while the accumulation of arsenic is clearly lower (only one fifth) in the Gulf of Finland. Metal and arsenic accumulations in the Gulf of Finland calculated in our study are somewhat higher than the estimates of Borg and Jonsson (1996). Comparison between the two studies is not really informative, since Borg and Jonsson used a value of annual dry matter accumulation about half of ours and their data were calcuted from metal concentration estimates for the 1980s.

Conclusions

This study on the concentrations of selected elements in the surface layer of recent soft sediments in the Gulf of Finland shows an improving trend in the most loaded parts in the eastern Gulf. The metals seem to be well trapped in the sediment column. However, this will hold true only as long as physico-chemical conditions in the Gulf remain stable. For example, a total loss of oxygen in the near-bottom water would release metals bound in iron-manganese hydroxides into the water column. If oxygen depletion were accompanied by changes in hydrographic conditions and currents, the final depositon pattern would be hard to estimate. The situation in the Gulf of Finland is indeed unstable, as evidenced by the extensive algal blooms (Rantajärvi 1996) and variable oxygen content in the near bottom waters (Laine et al. 1997). Further studies on the behaviour of the metals in different hydrological and physico-chemical conditions are of great importance. The transportation of metals from source areas to more distant accumulation areas needs to be more fully studied and we need to know the amounts of anthropogenically released metals that are actually available to biota.

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Element	Gulf of Finland (tonnes a ⁻¹)	Gulf of Finland (tonnes a ⁻¹)*	Gulf of Bothnia (tonnes a-1)**
Dry matter	$8.7 imes10^{6}$	$3.9 imes10^6$	10 × 10 ⁶
Hg	1.6	1.2	1.2
Cd	10.7	7.4	4.2
Pb	452	277	257
Cu	374	246	266
Zn	1 818	1 400	1 655
As	122	59	570

*Borg and Jonsson (1996). ** Leivuori and Niemistö (1995)

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