Historical pollution trends in a disturbed, estuarine sedimentary environment, SW Sweden

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Abstract The objective of this article is to determine whether pollution trends can be reconstructed from sediments in the outer Göta älv estuary. Detailed interpretation of heavy metal profiles in the recent sediment, in combination with profiles of ^{210}Pb- and ^{137}Cs-activity, organic carbon content and grain size distribution, allows estimation of historical trends. This is based on the assumption that only certain intervals, which can be identified, are disturbed. The sediment data are compared with archive data on historical trends in emissions of heavy metals. Special attention is given to mercury, which shows high enrichment over long periods of time. It is found that the most polluted sediments were deposited between 1930 and 1980, but that episodic discharges have also occurred more recently. Nevertheless, the effects of remediation since the 1970s are apparent.

Keywords Estuary · Heavy metal · Mercury · Pollution trends · Sweden

Introduction

Suspended matter scavenges pollutants while settling through the water column and thus sediments are often enriched in contaminants. This means both that sediments are a potential source of pollution if they are disturbed and that they can act as historical archives. Historical interpretations of pollution in sediment cores can be made, provided that the possible problems related to coring techniques, bioturbation, physical mixing and diagenesis are taken into consideration. If literature data on heavy metal supply are available, documented pollution inputs can validate the interpretation of contaminant trends in sediments. A good agreement between the sediment record and known pollution discharge trends would support the hypothesis that variations in contaminant supply from the main sources are reflected in the sediments. On this basis, discharge records may be further used to constrain the age determination based on sediment data. However, both synsedimentary and post-depositional processes may limit this approach, and thus interpretations will be made in the context of a sedimentological model.

The main sources of heavy metal pollution are usually atmospheric input, industrial discharges and sewage discharges. Studies of heavy metals in mires (e.g. Jensen and Jensen 1991; Jensen 1997) can help to reconstruct the atmospheric input of pollutants to estuarine systems. Jensen and Jensen (1991) report that the deposition rate of Hg in south-western Scandinavia has increased from about 10 to about 30 μg Hg m^{-2} year^{-1} since the beginning of this century. The deposition of Cd, Cu, Pb and Zn in Sweden has also generally increased over the past 150 years (Jensen 1997).

Bricker (1993) found that sediment concentrations of all metals at Rhode Island (United States) began to increase over background levels at around 1900 and that the observed distributions in the cores were generally consistent with estimated long-term trends in inputs from atmospheric and sewage sources. Valette-Silver (1993) made a global review of sediment studies published up to 1990, which show an increase in sediment contamination in the late 1880s, further increases in the 1940s, and a plateau or a maximum in the 1960–1970s. Cooper (1993) found that sedimentation rates, the prevalence of anoxic conditions and eutrophication have increased in the Chesapeake Bay (United States) since the time of European settlement around 1760.

Large areas of estuary systems are commonly affected by human activities, especially by dumped material. Olsen and others (1993) define three types of area characterised by (1) little or no net deposition, (2) natural deposition or

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(3) deposition affected by human activities (dredging, construction). Localised areas were found to serve as contaminant sinks, whereas sediments along natural channels and in wide shallow areas were relatively uncontaminated. Feng and others (1998) point out that variations in deposition of material on both short (seasonal) and long time scales can be a reason for variability in trace metal levels. The effect of changes in sediment accumulation rate was also pointed out by Croudace and Cundy (1995), who therefore suggest that pollutant flux should be used for the reconstruction of pollution trends rather than concentrations.

The objective of this article is to evaluate how far it is possible to reconstruct historical pollution trends in the Göta älv estuary using the sedimentary record in the area, which is influenced by both natural and anthropogenic processes. The age estimates based on sediment data will be compared with archive data on organic matter and heavy metal discharges in the area, and a refinement of age estimates using documented discharge trends will be discussed. Since the Göta älv River has a catchment area of 50,180 km^2, the sediments at its mouth have the potential of recording the pollution history in a large part of SW Sweden, where some of the main industrial areas in Sweden are located. Information about the containment and behaviour of contaminants in estuarine sediments is valuable for understanding the distribution of contaminants in polluted areas and identifying potential sources of reactivated contamination even after discharges have ceased. A stratigraphic framework for the study area spanning the whole Holocene was presented by Brack and others (2000). Recent sediments cover the whole Göta älv estuary area with a thickness of 10–80 cm. They are separated by a hiatus from older clays and distinguished by consolidation, colour and organic matter content.

Environmental setting

The study area includes the mouth of the southern branch of the Göta älv River (Fig. 1) and the outer estuary (Fig. 2). The Göta älv River is the largest river in Scandinavia, with a mean discharge of 180 m^3/s at its mouth and a catchment area that corresponds to one-tenth of Sweden's land surface. The catchment area is largely characterised by crystalline bedrock with a thin cover of till and coarse, glaciofluvial sediment, overlain by clay deposits of up to 100 m thickness. About 10% of the catchment area is used for agriculture (Clason and Granström 1992). After the last glaciation, the area was exposed to changing sea levels because of the balance between isostatic uplift and eustatic sea-level changes. The present rate of land uplift is 0.2 cm/year (Påsse 1987). Water depths in the area mostly vary between 5 and 15 m. Göteborg Harbour is mainly situated in the inner and middle estuary and in the area where the estuary widens, but parts of the harbour extend into the outer estuary on the northern side. The main shipping lanes are situated in 13- to 20-m deep channels that branch into the northern and the southern outer estuary as shown if Fig. 2. Maintenance dredging is only required in the harbour portion of the estuary.

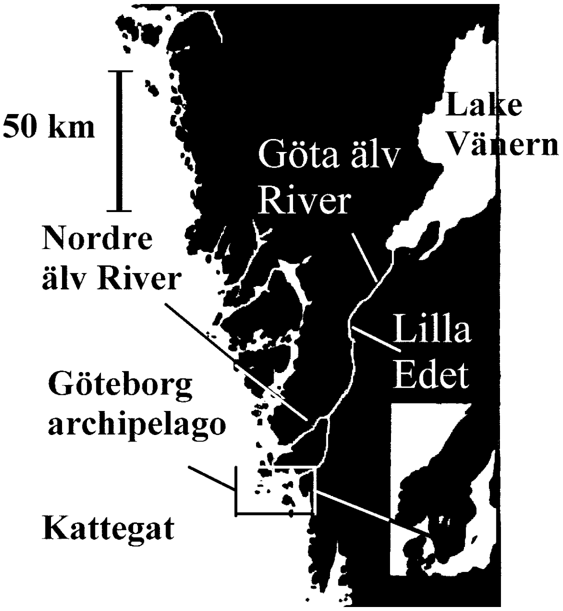


Fig. 1
Location map of the Göta älv River, Lake Vänern and Kattegat

The fresh-water outflow from the Göta älv River and the inflowing deep water from the Kattegat create a strongly stratified estuary (Selmer and Rydberg 1993), situated in a micro-tidal environment. A salt wedge is developed in the inner harbour, with a fluctuating position depending on river discharge and wind directions. The surface freshwater layer is 2–5 m deep and extends into the outer estuary, influenced by local wind and sea-level variations (Selmer and Rydberg 1993). Discharge variations are presently mainly caused by river regulation by the hydroelectric power plant at the outlet of Lake Vänern and a screen at the bifurcation of the river, about 20 km upstream of Göteborg. Where the estuary widens, the surface water is affected by the Coriolis force and is often deflected to the north.

The sediments in the study area are fine-grained, with silty clays dominating. This is both because of sediment sources and transport mechanisms. The Göta älv River valley is filled with glaciomarine and younger marine clays, and the dominant sediment transport mode is suspension. Sediment grain size decreases towards the outer estuary and archipelago (Johannesson and others 2000), probably as a result of decreasing flow energy at the widening of the estuary and an increasing flocculation effect in the middle estuary and outward. Suspended matter concentrations vary between 0.8 mg/l (Sundborg and Norrman 1963, measured April 1958 to April 1959) and 75 mg/l (The Göta River Water Protection Association, unpublished data from 1998 and 1999).

The main industries that have discharged to the Göta älv River are paper mills, an alkali chloride plant at the bi-

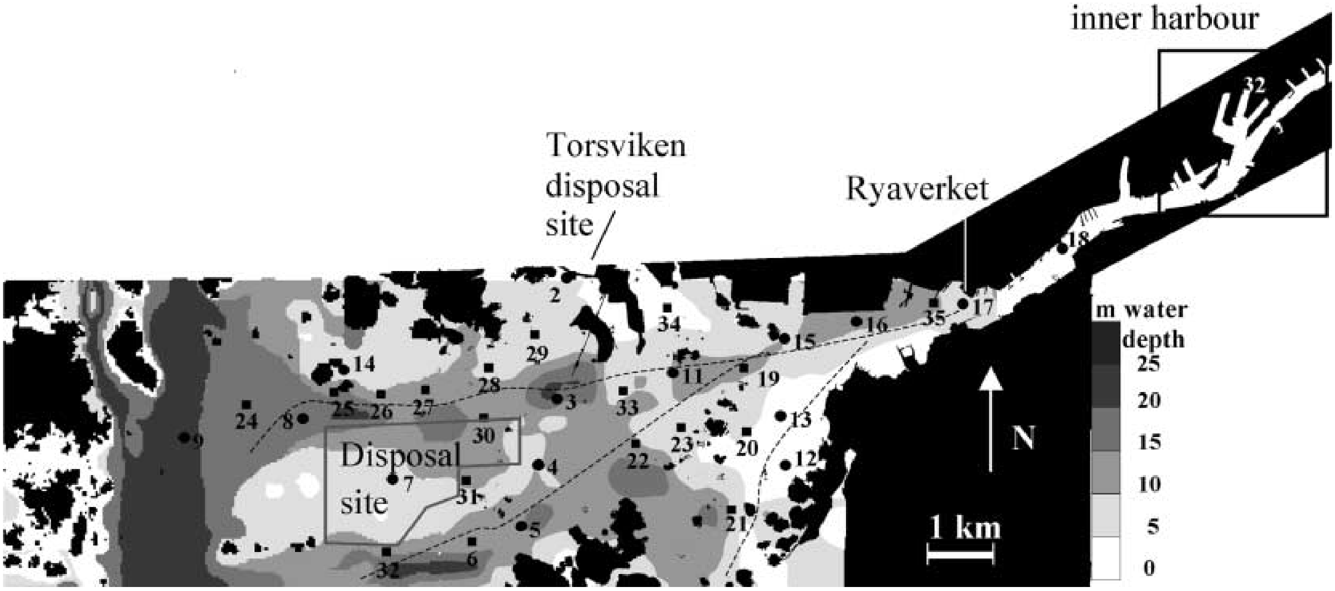


Fig. 2
Water depth and sampling sites (*squares* box cores, *circles* piston cores) in the study area. The shipping lanes are indicated by *dashed lines*

furcation of the river, a battery factory further upstream and wharves in the harbour area. Untreated sewage was discharged from nearly the whole city until 1972, when the Ryaverket sewage treatment plant was built (Fig. 2). The original plant had limited biological treatment capacity and a plant expansion in 1982 added primary settling and greater aeration. Chemical precipitation was introduced in 1984 and purification has continuously been improved.

Methods

Sampling

Sediment sampling was done with a 50×30×30-cm box corer at 35 sites (Fig. 2, see electronic supplementary material, Table 1, for co-ordinates), of which 18 were also sampled with a Kullenberg-type, 3-m gravity corer on a transect from the middle to the outermost estuary (E–W) and on three short cross transects. This scheme was designed to document changes along the river outflow and differences between open and more sheltered settings. The box cores were fairly evenly spaced in different sub-environments. Sediments from the box cores were described and subsampled (~50 ml at 0–2, 10–12, 15–17 and 20–22 cm) with plastic devices within minutes of sampling. For later lithological description, a short core was taken by pressing a plastic tube into the box-core sediments and stored at 4 °C, as were also the gravity cores. Both short and long cores were described for lithology, colour, degree of consolidation and structure of sediments (Ross and others 1978). Three representative cores were chosen for dating of the recent sediment layer (2, 5 and 32). Piston cores were used for all analyses because they are sufficiently long to retrieve the main sediment facies. As a control of the recovery of the piston cores, they were compared with box cores from the same site. It is assumed that box cores are relatively undisturbed in these shallow environments.

Analytical methods

Geochemical analyses were done by XRAL Laboratories, Canada, on dried, ground samples. Concentrations of 33 elements were determined by ICP-AES from multi-acid (HF and $HClO_4$) digested bulk samples. Hg was determined by cold vapour absorption and analysed with a Leeman PS200 Hg autoanalyser. The measurement precision for Cu, Pb and Zn is about 5% and for Hg it is 7%, based on replicate standard measurements and duplicate samples. Detection limits for Cu and Zn are 0.5 ppm, Pb 2 ppm and Hg 5 ppb. Heavy metal concentrations were not normalised, but we have employed the concept of an enrichment factor in the maps in Fig. 3, using pre-industrial sediments as background (Dickinson and others 1996). Total organic carbon (TOC) and nitrogen were measured with a Carlo Erba analyser of type NA 1500 (Hedges and Stern 1984).

Table 1
Correlation coefficients for heavy metals, TOC (total organic carbon) and % clay (<2 μm). All *r* values that indicate moderate or strong correlation are printed *bold*

r	Zn-TOC	Hg-TOC	Pb-TOC	Cu-TOC	clay-TOC	r	Zn-clay	Hg-clay	Pb-clay	Cu-clay	Cu-Zn
Core 2	**0.69**	0.49	**0.93**	0.51	–0.42	core 2	0.09	0.06	–0.23	0.12	**0.91**
Core 5	0.36	0.50	**0.88**	0.18	**0.71**	core 5	**0.86**	**0.75**	**0.66**	**0.65**	**0.94**
Core 32	0.04	0.07	0.17	0.56	–0.13	core 32	0.61	**0.73**	0.33	–0.2	0.09

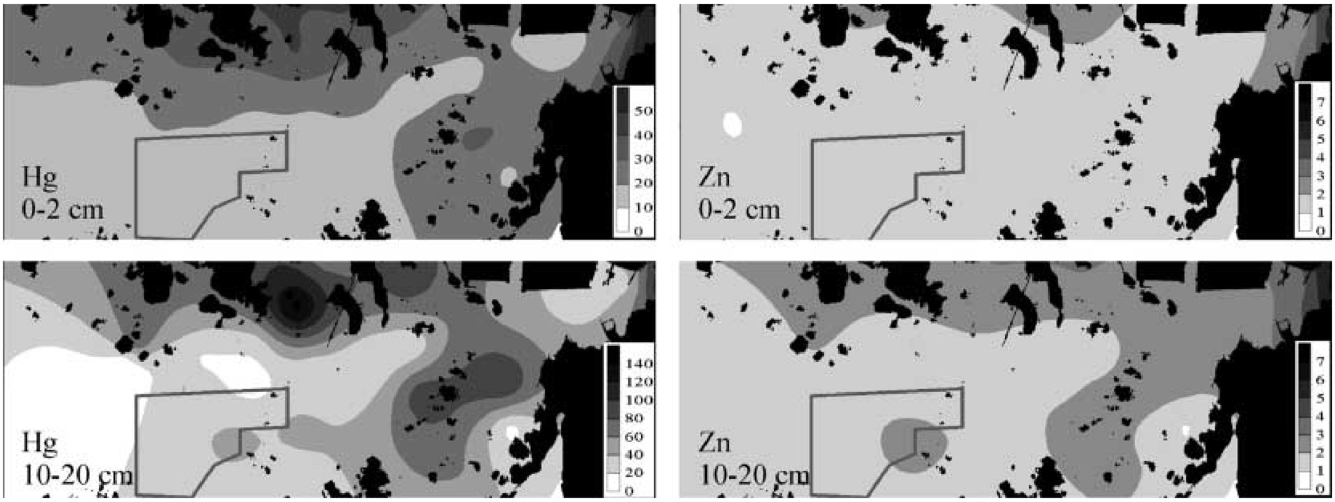


Fig. 3
Geographical distribution of heavy metal enrichment factors in the recent sediments

For grain size determination, sediments were treated with 15% H_2O_2 to remove the organic matter, followed by sieving with 500- and 63-μm sieves. Five grams of <63 μm material were weighed into a 100-ml centrifuge tube and dispersed in distilled water with an ultrasonic probe. To prevent flocculation, this suspension was washed by repeated centrifugation and resuspension with fresh water. To aid dispersion by the ultrasonic probe, 1.5 g of sodium polyphosphate was added to the sample immediately before analysis by a Micromeritics Sedigraph 5100 down to 0.2 μm. Grain size classes were defined following the Atterberg scale (Krumbein and Pettijohn 1988).

Only 3 of the 35 sampling sites were considered suitable for dating of the recent sediment layer. The restricting factors include (1) coarse grain sizes of the recent sediments, (2) non-recovery of the lower boundary of the recent sediment layer, (3) disturbed lower boundary of the recent sediment layer because of dumping of dredged sediments or dredging, and (4) non-recovery of recent sediments. Dynamic bottom conditions prevail in the whole study area, which will be taken into consideration when interpreting the results.

Age determination

^{210}Pb and ^{137}Cs measurements were done at the Risø National Laboratory in Roskilde, Denmark, using 16 samples per core. ^{210}Pb is the product of the gas ^{222}Rn (half-life τ=3.8 days), whose parent ^{226}Ra (τ=1,600 years) is present almost universally in crustal material. The ^{210}Pb is incorporated into sediments by scavenging. Once in the sediment, the ^{210}Pb starts to decay (τ=22.3 years), but it is also formed in the sediments by the decay of ^{226}Ra at a constant rate (supported ^{210}Pb). The time that has elapsed since particles were last at the sediment surface can be calculated from the decrease in the unsupported (total–supported) ^{210}Pb with depth (Geyh and Schleicher 1990; Libes 1992). ^{137}Cs is an anthropogenic radionuclide (τ=30.2 years) formed by fallout from nuclear weapon tests, releases from nuclear power plants and accidents, such as the one in Chernobyl 1986. It can be used as a time marker when peaks corresponding to known fallout maxima (1954 onset of fallout, peaks at 1958/1959, 1961, 1963 fallout maximum, 1986 Chernobyl; French and others 1994) can be identified. The combination of ^{210}Pb and ^{137}Cs dating is common because the methods are based on two different approaches: ^{210}Pb data are used to calculate sedimentation rates by regression, whereas ^{137}Cs dating is based on marker levels. Furthermore, ^{137}Cs is subject to different post-depositional biogeochemical processes than ^{210}Pb (Farmer 1991).

The ^{210}Pb age was calculated using linear interpolation (Goldberg and others 1979; Alexander and others 1993), assuming a constant flux of ^{210}Pb at the sediment–water interface and a constant sedimentation rate at certain intervals. Mean sediment accumulation rates over undisturbed intervals are derived from the slopes of the excess ^{210}Pb profiles (Sharma and others 1994). Using the regression line, the age of the top and bottom of the interval is calculated by determining A_0 (regression line x-intercept at the top of the core interval) and A_i (regression line x-intercept at the bottom of the interval) and calculating the age t according to the equation $t = \lambda^{-1} \ln(A_o/A_i)$ using λ=0.0311. The full geochemical and radioactive element analysis data set is available in electronic Table 2 (electronic supplementary material).

Radiocarbon dating on three shells in recent sediment from cores 2 and 4 was done at the Leibniz-Laboratory for Radiometric Dating and Isotope Research in Kiel, Germany (Brack and others 2000).

Results and interpretations

The main facies in the area is a soft, recent, silty clay overlying two, different, consolidated clays. One is of distal glaciomarine origin and a younger one deposited in an estuarine setting before 1000 B.P. In some areas, dumped, dredged sediments are found with a thickness of over 1 m.

Table 2
Vertical and geographical variation in heavy metals and sediment characteristics. Vertical variation (σ=standard deviation and σ_r=relative standard deviation) is calculated from different samples at different sediment depth from the same site (five cores), and geographical variation from the same sediment depth (four levels) at 35 different sites. σ_{rv}/σ_{rs} indicates the difference between vertical and geographical variation. n^* indicates the average number of samples for the five cores and four levels respectively

Vertical	Cu (ppm)	Zn (ppm)	Pb (ppm)	Hg (ppb)	Sand (%)	Silt (%)	Clay (%)	Water (%)	TOC (%)
n^*	20	20	20	20	18	18	18	27	17
Mean$_v$	59.06	235.75	68.80	1505.92	7.07	30.45	62.48	52.93	2.70
σ_v	16.16	68.24	21.08	1235.56	2.77	3.17	3.83	2.84	0.48
σ_{rv} (%)	27	29	31	82	39	10	6	5	18
Geographical									
n^*	26	26	26	26	26	26	26	26	26
Mean$_s$	45.42	177.99	50.80	965.60	11.75	26.02	62.03	56.83	2.61
σ_s	26.03	97.34	20.69	857.44	12.98	6.21	11.23	7.40	0.89
σ_{rs}(%)	57	55	41	89	111	24	17	13	33
Difference									
σ_{rv}/σ_{rs}	0.48	0.53	0.75	0.92	0.35	0.44	0.36	0.42	0.54

The non-parametric Kruskall–Wallis test (Ebdon 1977) combined with Dunn's multiple comparison test (Dunn 1964) shows that the recent clay facies differs significantly from the consolidated clay in all tested parameters except clay content (Brack and Stevens 1999). The most predominant recent sediment in the Göteborg outer estuary is a dark brown, organic-matter rich, silty clay. This clay is relatively homogeneous and occasionally shows a diffuse layering. The bottom fauna that contributes to bioturbation is restricted to a few species, which are probably tolerant to salinity variations and post-oxic conditions. The upper millimetres of the sediment are oxic, as indicated by their light brown colour and oxygen measurements. Below this the sediments are dark brown to black, which can be explained by the high organic matter content and the presence of sulphides in the reduced sediment. The presence of ~0.5 ppm dissolved Mn in the pore water at 0.5 cm and the smell of hydrogen sulphide imply that the sulphidic zone (Berner 1981) persists up to the thin oxic surface layer.

Core 2 was taken in 5 m water depth about 100 m south of the barrier dam enclosing the disposal site for contaminated dredged sediments at Torsviken in the northern outer estuary (Fig. 2). This site was chosen to document possible leakage of heavy metals from the disposal site, which seems likely because there is a pipe that connects the water column in the disposal site with the water outside. In core 2, the distal glaciomarine clay is overlain by the older estuarine clay (Brack and others 2000), representing the only sampled co-occurrence of these two pre-industrial (>1,000 years) facies. The upper 42 cm consist of recent, organic-matter rich, silty clay. The pre-industrial sediments appear to be undisturbed, whereas the recent sediments are partly disturbed. A clay layer at 14–17 cm in the recent sediments might originate from a slump in connection with building activities, such as construction of the disposal site dam in 1975, or from dumping before the disposal site was enclosed. The uppermost 4.5 cm was lost during sampling, as indicated by the box core from the same sampling site and the ^{137}Cs profile (Fig. 4).

Sampling site 5 is located in a natural channel in the southern outer estuary. This is one of the few locations with a thick recent sediment layer (70 cm). Core 32 was taken at the end of a harbour basin in the central harbour (Frihamnen). This site was chosen because the seismic record showed a thick layer of soft sediment. The sediments in this core are sandier than in the outer cores.

Correlations of heavy metals with grain size and TOC

To evaluate whether vertical heavy metal trends are caused by changing sediment composition, the correlation of the heavy metals with organic carbon concentrations and clay percentages was tested and the results are given in Table 1. In the case of a strong correlation, normalisation would be needed to remove a dependency, which could mask the contaminant fluxes. The significance of correlations was tested, yielding a threshold level of 0.65 at a probability level of 99% and 0.45 at 95%. We define therefore weak, moderate and strong correlations with r>0.45, 0.65 and 0.8 respectively. In general, the correlations differ between the cores. In core 2, heavy metal concentrations correlate with TOC, but not with the clay content, in core 5 they correlate partly with TOC and clay content, and in core 32 the pairs Cu–TOC and Zn–clay show a weak correlation, and the one for Hg–clay is stronger. Increasing TOC concentrations may be caused by natural changes in climate or sedimentation rates, or anthropogenic impact on land use, nutrient and industrial discharges. Because we consider most of the TOC as originating from discharges coupled to or at least contemporaneous with high heavy metal discharges, we decided to interpret high TOC contents (above ~3%) as indication of pollution rather than naturally changing sediment composition. Furthermore, correlations with sediment parameters vary spatially. Therefore, normalisation of heavy metals with TOC or clay content was not done.

Variability of the sediment heavy metal content

To compare the differences between vertical and horizontal variation and between the different heavy metals,

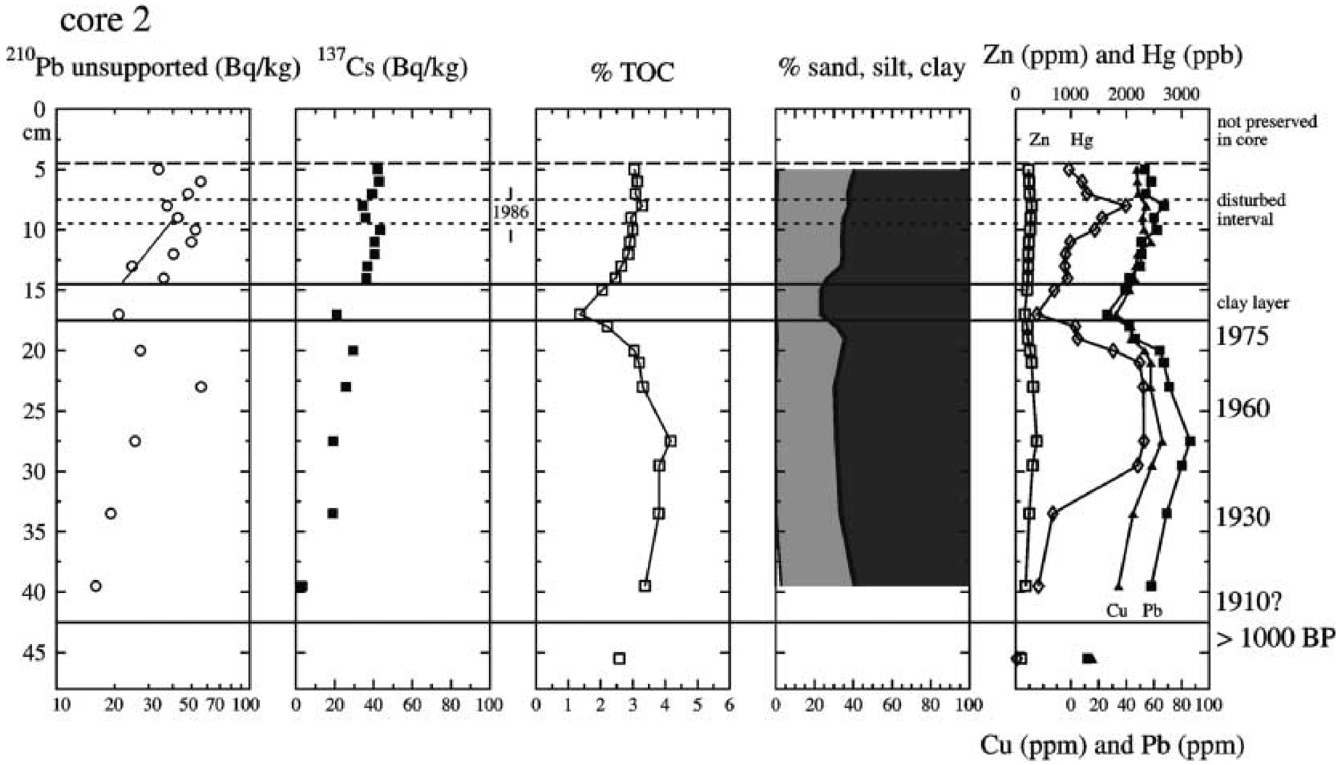


Fig. 4
^{210}Pb, ^{137}Cs, TOC, grain size and heavy metal profiles in the recent sediment layer of core 2 from the northern outer estuary. Lithological boundaries are indicated by *solid lines*. The lowermost sample was taken in the underlying pre-industrial clay. Observe the *different scales* for heavy metals

the standard deviation for four horizontal (0–2, 10–12, 15–17 and 20–22 cm) and five vertical data sets (cores 2, 4, 5, 17 and 32) was calculated. The mean values of the standard deviations, describing vertical and the horizontal variation, are shown in Table 2, and the full data set is available in electronic Table 3 (see electronic supplementary material). Confidence limits for the individual data sets at a 95% level were always less than standard deviation. The heavy metal concentrations are not strictly normally distributed, as assumed for simplicity. However, the comparisons of medians and percentiles lead to the same conclusions. The relative standard deviation values for all heavy metals are higher between samples from different parts of the study area than between samples from one profile in the recent sediment layer. This is mostly because of the increasing distance from pollution sources towards the outer estuary, but partly also the geographical variation of organic carbon and grain size. The contents of silt, clay, water and TOC vary geographically more than vertically within the recent sediment layer of one profile (Table 2). The difference between geographical variation and variation with depth is most pronounced for Cu, and decreases for other tested metals in the order Zn–Pb–Hg. This could be because of a difference in the importance of local sources for these metals. The relative standard deviations of Cu, Zn and Pb are of the same order of magnitude, whereas Hg has about twice the value, suggesting different sources or transport or sedimentation processes.

Heavy metal distribution in recent sediments

The pre-industrial estuarine clay in the study area was used as background (Cu=15, Pb=17, Zn=83, Hg=0.02 ppm), and it has similar concentrations as the background reported by Cato (1977). The mean heavy metal concentrations of all samples from the enriched recent sediment layer in piston cores and box cores are: Cu 45 ppm, Zn 200 ppm, Pb 60 ppm and Hg 1.22 ppm. Heavy metal distribution is shown as enrichment factors relative to background in Fig. 3. Enrichment factors in the study area are highest for Hg, up to 160 in the clays at 10–20 cm, on average 26 in the surface sediments and 47 in the lower sediments. The mean enrichment factors (ef) for Cu, Zn and Pb are lower: Cu (2.7)>Pb (2.5)>Zn (1.9). The highest enrichment for all heavy metals is found close to the Ryaverket sewage treatment plant in the middle estuary (Fig. 2), followed by the northern outer estuary, where especially Hg is enriched (Fig. 3).

The recent sediments in core 2 from the northern outer estuary contain elevated mean heavy metal concentrations [Cu 50 ppm (ef=3.3), Zn 253 ppm (ef=3), Pb 59 ppm (ef=3.5), Hg 1.3 ppm (ef=65), see electronic supplementary material, Table 2, for raw data]. The distribution with depth is relatively uniform for Zn, although lower contents occur in the clay layer at 14–17 cm, which consists of older, allochthonous clay (Fig. 4). Hg shows two intervals of enrichment, between 29 and 17 cm and 9 and 7 cm sediment depth. These coincide roughly with intervals of high organic carbon content.

Mean heavy metal concentrations in core 5 from the southern outer estuary are lower than in core 2, but still elevated [Cu 35 ppm (ef=2.4), Zn 168 ppm (ef=2), Pb 50 ppm (ef=2.9), Hg 0.65 ppm (ef=33)]. The heavy metal profiles are somewhat similar to core 2: Hg shows two intervals of enrichment, one between 70 and 40 cm and a sharp peak at 8 cm, which does not appear in the Zn profile (Fig. 5). In this core, however, the highest concentrations are found in the bottom part of the recent sediment layer, and not in the middle, as in core 2. Below 50 cm, the high concentrations coincide with high organic carbon concentrations and relatively high clay contents.
Sediments in core 32 from the central harbour show a coarsening upwards trend (Fig. 6), especially in the uppermost 6 cm. Mean heavy metal concentrations are in the same range as in core 2, but Cu and Pb are more enriched and Hg is less enriched: Cu 88.5 ppm (ef=5.9), Zn 234 ppm (ef=2.8), Pb 83 ppm (ef=4.9), Hg 0.74 ppm (ef=37). Concentrations are highest in the lower part of the recent sediment layer, decreasing strongly from 30–20 cm sediment depth. This is parallel to the grain size trend. A peak in organic carbon at 16 cm corresponds to a slight increase in Hg content.
Because the difference in heavy metal concentrations between recent sediments and the consolidated clay can be attributed to contamination, the whole recent sediment layer is assumed here to have been deposited during industrial times, which means that it has a maximum age of about 100 years.

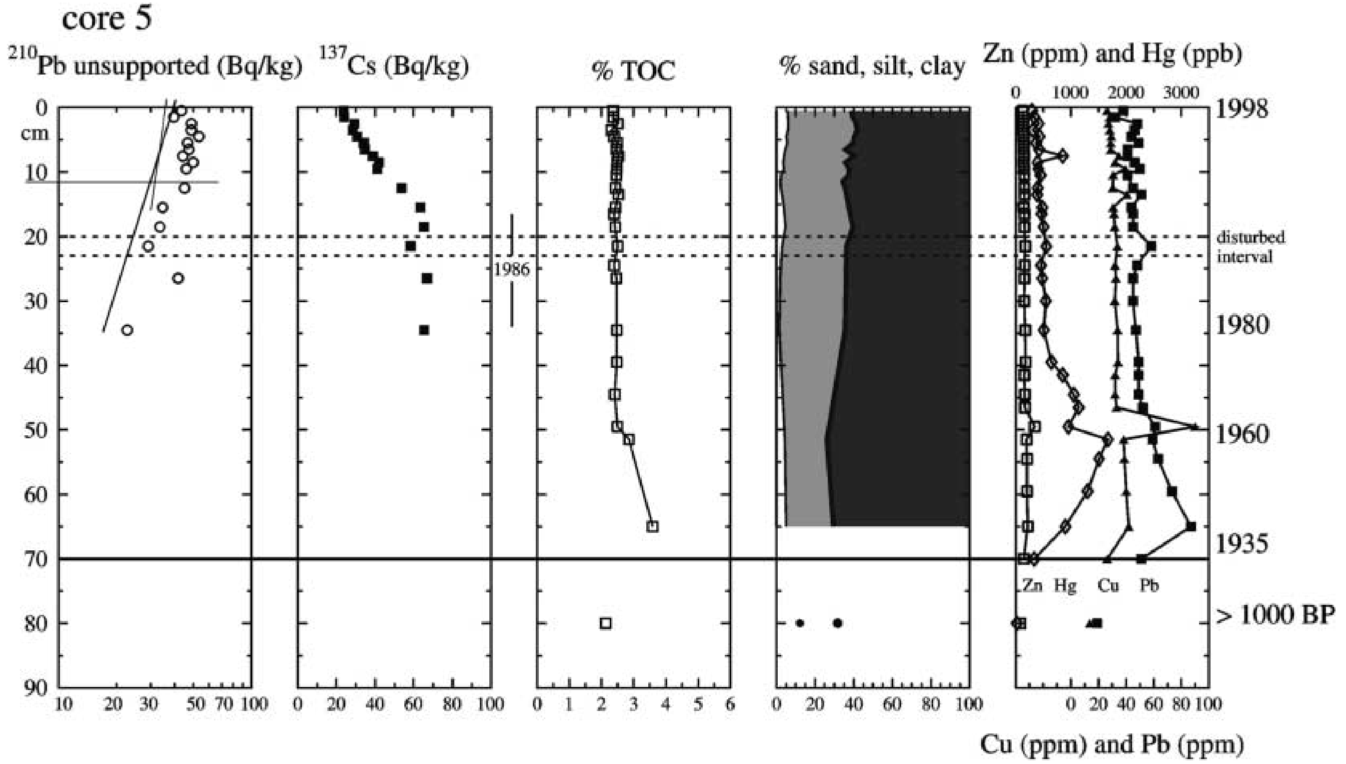
Comparison of Cu, Zn, Pb and Hg profiles

In core 2, the Cu, Zn and Pb profiles are parallel (Fig. 4), whereas the Hg profile shows a wider interval of elevated concentrations. In core 5, Cu and Zn follow the same general trend as Pb, but they show a peak at 50 cm sediment depth that is not present in the Pb profile. Again, the Hg profile shows a wider interval of elevated concentrations, and a peak at 8 cm, which is absent for the other metals. Largest differences between all heavy metals are found in core 32. Zn, Pb and Hg decrease upwards, whereas the Cu concentrations show relatively high variation around an average level. The decrease upwards is most pronounced for Hg, and less for Pb and Zn, in this order. The uppermost sample shows high Pb concentrations. To conclude, Hg is not only more enriched than the other heavy metals, but is also found in wider sediment intervals (Figs. 4, 5 and 6). It is possible that site 2 received mostly regional pollution, possibly from the disposal site, whereas sites 5 and 32 are more influenced by local sources of Cu, e.g. from anti-fouling paints.

Sediment chronology

The three sites that were chosen for age determination of the recent sediment layer (sites 2, 5 and 32) are located in different, representative environments. The best preserved sediment record was found in the southern outer estuary (site 5; Fig. 2). At most sites, the sediment appears relatively unaffected by bioturbation and seems to have a stable diagenetic zonation. It is assumed that the 1986 Chernobyl ^{137}Cs peak is recorded in these sediments

Fig. 5
^{210}Pb, ^{137}Cs, TOC, grain size and heavy metal profiles in the recent sediment layer of core 5 from the southern outer estuary. The lowermost sample was taken in the underlying pre-industrial clay. Observe the *different scales* for heavy metals



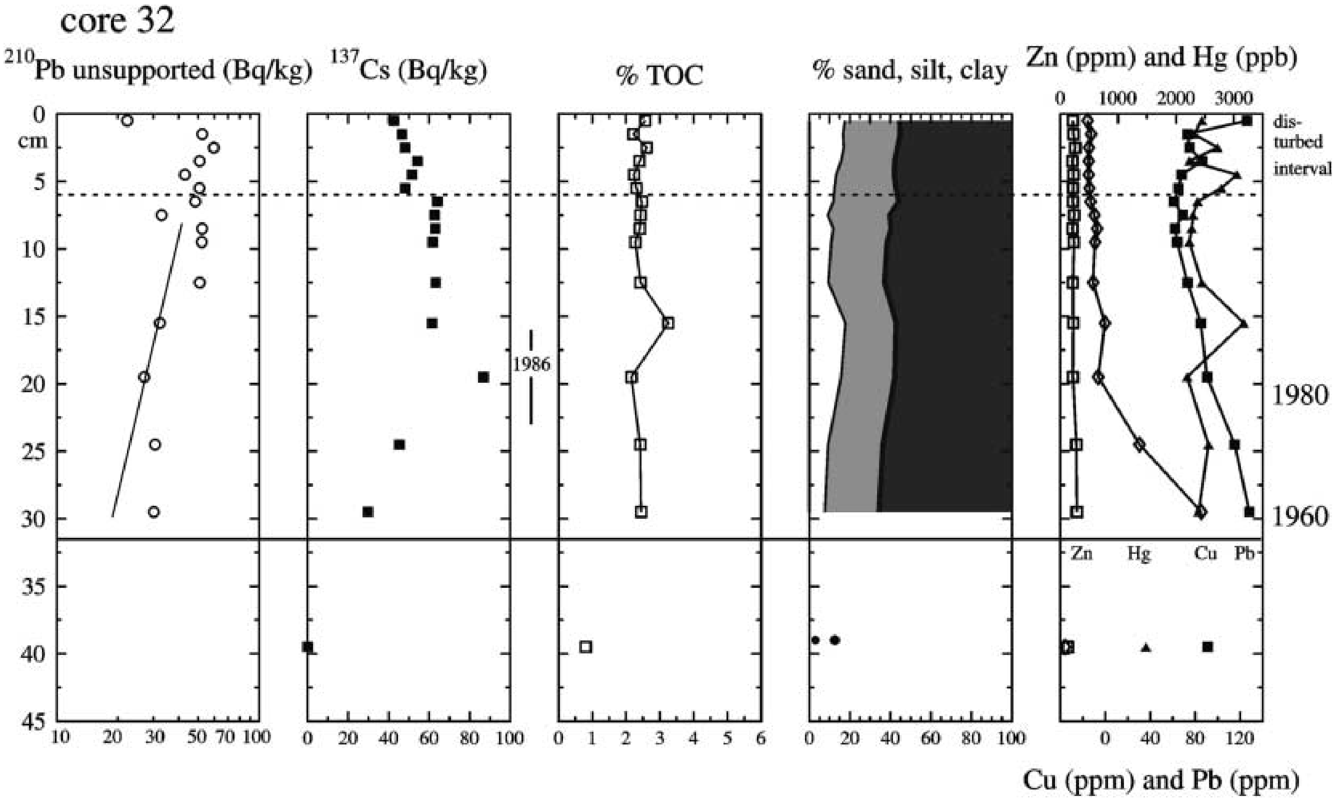


Fig. 6
^{210}Pb, ^{137}Cs, TOC, grain size and heavy metal profiles in the recent sediment layer of core 32 from the inner harbour. The lowermost sample was taken in the underlying pre-industrial clay. Observe the *different scales* for heavy metals

because significant fallout was deposited from the atmosphere in the catchment area of the Göta älv River. Marine sources that would carry the Sellafield-signal (1973–1980, Kunzendorf and others 1996) are of minor importance in the area (F. Klingberg, personal communication).

Radiocarbon dating of shells from site 4 in the middle outer estuary (11.5 cm sediment depth) and site 2 (22 cm) showed high contents of modern carbon from bomb tests, which indicates a maximum age of 1954 A.D. (Brack and others 2000). At 27.5 cm in core 4, the modern carbon concentrations are lower, giving an age of 1954±35 A.D.

Core 2

The ^{137}Cs and ^{210}Pb profiles in core 2 from the northern outer estuary allow several approaches to age determination, yielding a minimal possible sedimentation rate of 0.4 cm/year and a maximal possible sedimentation rate of 0.8 cm/year. A clear ^{137}Cs peak in the recent sediments is not evident other than a higher level of activity above the clay layer (Fig. 4). The uppermost 4–5 cm have been lost during sampling, as indicated by the recovery of these surface sediments in a box core at the same site and by the slight increase in ^{137}Cs activities towards the surface. Undisturbed surface sediments should show a decrease in activity (cf. core 5). An interval of older, contaminated sediment dumped at this site would best explain the low ^{210}Pb and ^{137}Cs activities in connection with high Hg concentrations at 7–9 cm (Fig. 4). Below the clay layer, activities are lower, but still present, so the sediments contain fallout from nuclear bomb testing in the 1950s to 1960s. The lowermost sample has a low activity (3.2 Bq/kg), which might reflect a greater age at this depth. The lowered activity could be a primary signal or an effect of bioturbation mixing with underlying material. It is reasonable to assume that the sediments above 34 cm in core 2 are younger than 1954 when nuclear weapon testing began to introduce significant ^{137}Cs into the natural environment (Farmer 1991). Although the surface is probably not intact, the ^{137}Cs peak above the clay layer is correlated to Chernobyl 1986, suggesting a maximum mean sedimentation rate of about 0.8 cm/year.

If a regression line through the ^{210}Pb profile is used to calculate ages for the top and bottom of the part that seems undisturbed (9–14 cm), lower sedimentation rates of about 0.4 cm/year are possible. In that case, the two lowermost ^{137}Cs samples show activities that are too high, probably because of bioturbation, which does not have to be strong to affect ^{137}Cs activities (H. Kunzendorf, personal communication). Heavy metal concentrations in the sample at 39 cm immediately above the bottom of the recent sediment layer are higher (181 ppm Zn) than in the sample at 45 cm at the top of the pre-industrial clay that is older than 1000 BP. (99 ppm Zn). Therefore, it seems reasonable that the bottom of the recent layer dates from about 1910. However, it might be up to 35 years younger if a higher mean sedimentation rate is assumed, as is suggested by the interpretation of the ^{137}Cs profile described above.

Core 5

In core 5 from the southern outer estuary, the ^{210}Pb and ^{137}Cs datings show that higher sedimentation rates (1–2.5 cm/year) are responsible for a greater recent sediment thickness than at site 2. The ^{137}Cs values (Fig. 5) are nearly constant from 35 to 12 cm and decrease upwards, and there is also a slight coarsening upwards trend from 12 cm. These trends are probably caused by either a real decrease of the ^{137}Cs-supply or by an increasing sedimentation rate. The expected Chernobyl peak is either very broad or not preserved. However, the high ^{137}Cs activities of the sediments above 35 cm indicate that they are younger than 1954, and probably deposited around 1986. In that case, sedimentation rates lie between 1.5 and 2.5 cm/year above 35 cm sediment depth.

^{210}Pb values increase from 35 to 12 cm and are constant above that. Linear interpolation yields a sedimentation rate of about 1.2 cm/year from the whole ^{210}Pb profile. In the upper 12 cm, the estimated mean sedimentation rate is higher (2 cm/year), so that this interval represents about 5 years. The interpretations of the ^{137}Cs and ^{210}Pb profiles agree well with each other. If a mean sedimentation rate of about 1.2 cm/year is valid below 12 cm, the bottom of the recent sediments dates to about 1945. However, without dated samples below 35 cm, the age of the lower boundary remains speculative. The sediment below 35 cm has a different grain size distribution (more clay and sand) and a higher organic matter content (Fig. 5). Together with higher heavy metal concentrations, this may indicate a slightly lower sedimentation rate and an older lower boundary of the recent sediment layer.

Core 32

Sedimentation rates calculated in core 32 from Frihamnen are in a similar range to the others (0.9–1.3 cm/year). The ^{137}Cs values increase upward to a peak at 19.5 cm, above which they are constant until 6 cm, where slightly lower activities are recorded (Fig. 6). Because the sample at 19.5 cm is the only one between 15 and 25 cm, the peak of activity may be located anywhere within this interval, corresponding to sedimentation rates between about 0.9 and 1.3 cm/year. In the ^{210}Pb profile an increase in activity between 20 and 9 cm separates the relatively high values above from the lower values below. Interpolation between 30 and 9 cm yields a sedimentation rate of 0.9 cm/year. This suggests a ^{137}Cs peak position at 17 cm, excluding the interval above 6 cm from the estimation because it is assumed to be disturbed (Fig. 6). In this core we cannot expect to find the bottom of the recent sediment layer preserved because this part of the harbour basin was deepened in the 1960s and 1970s (Lars Dahlin, personal communication). When the estimate above is applied to the whole recent sediment layer, the age of the lower boundary is calculated to 1963–1973.

It can be concluded that sedimentation rates are lower at site 2 (0.4–0.8 cm/year) than at sites 32 (0.9–1.3 cm/year) and 5 (1.2–2.5 cm/year). The lower boundary of the recent sediment layer in the harbour basin at site 32 relates to dredging in the 1960–1970s, whereas this boundary is natural at sites 2 (about 1910) and 5 (possibly about 1945). Using the estimates of the lower boundary and the ranges of possible sedimentation rates, the intervals with distinct heavy metal enrichment are estimated to have been deposited between 1935 and 1980 in core 2, 1945 (possibly earlier) to 1982 and a Hg peak at about 1994 in core 5, and between 1965 and 1982 in core 32. There is good agreement between these estimates, especially if the possibility of lower sedimentation rates in the lower part of core 5 is taken into consideration.

Heavy metal associations

To facilitate the interpretation of historical heavy metal trends we have so far concentrated on Zn and Hg. Even though Cu and Pb show broadly similar trends, there are some differences in the profiles that might be related to different sources for these elements (Figs. 4, 5 and 6). It has been reported by Bricker (1993) that Cu is often supplied by sewage, whereas Zn and Pb are mainly deposited from the atmosphere or from urban surface runoff. Enrichment factors between 2 and 10 usually correspond to mixed inputs of industrial effluents, sewage and runoff from agricultural sources (Förstner and Wittmann 1979). According to Cato (1977), the order of total heavy metal discharge in Sweden is Zn>Pb>Cu>Ni>Hg. This is also the order of concentrations in the sediments of the Göteborg outer estuary, although Ni has concentrations close to background, whereas all other heavy metals are elevated above background. Cu, Zn and Pb are found in the recent sediments at average concentrations of two to three times the background. There is a good correlation between Cu and Zn in all cores except core 32 (Table 2).

Manheim and others (1998) argued for using inter-element ratios to characterise contamination, and in particular plotting other heavy metals against Zn, which often displays highest concentrations amongst anthropogenic metals. The Cu/Zn ratio in the sediment of core 17 taken at Ryaverket (0.24) and most box-core samples from the outer estuary (0.25) agrees well with the ratio of these two metals in the discharge from Ryaverket (0.28, Göteborg regional sewage work, unpublished data). Sewage might therefore be the main point source for Cu and Zn, whereas surface runoff, erosion and other diffuse sources contribute to an unknown extent. The correlation of Pb with Zn in the sediment cores is less pronounced than for Cu, and the Pb/Zn ratio in sediments is generally two to three times higher in sediments than in the treated sewage discharge. This implies that other sources, such as atmospheric input and surface runoff, might supply significant amounts of Pb (Cato 1977). The high Hg-enrichment is mostly caused by high industrial discharges in the 1950s to 1970s, which dominated the Hg supply in the study area. Even the uppermost deposits may have received considerable Hg from the reworking of the older deposits. The asynchronous deposition of Hg and the other heavy metals is explained by the dominating Hg supply from cellulose works and alkali chloride industry during the 1950s to 1970s. These industries did not discharge significant amounts of heavy metals besides Hg.

Discussion

Historical data regarding heavy metal sources and pollution supply trends

Heavy metals in the study area have different sources, including atmospheric fallout by wet and dry deposition, weathering and erosion of contaminated soil, sewage and industrial discharges to the aquatic system (Lindqvist 1991). Land use and lake acidification may also play an important role, as acidified lakes have a lower capacity for binding heavy metals in sediments and heavy metals may even be released from contaminated sediments (Renberg and others 1990).

From the data presented above, it appears that mercury is the most enriched metal. The atmospheric fallout is partly transported to Sweden from central Europe, for example Germany, Great Britain and Poland (Pacyna and Münch 1991), and comes partly from sources in Sweden (Fig. 7). At the beginning of the 20th century, metal production, coal combustion and pyrite burning were the main sources of atmospheric mercury in Sweden, whereas from the 1930s to the 1970s the alkali chloride industry and waste incineration were mainly responsible for high emissions (Lindqvist 1991). The discharge of Hg was previously dominated by industrial point sources. Inorganic Hg was discharged from the alkali-chloride industry, whereas phenyl mercury was used as preservative for pulp from the 1940s to the middle of the 1970s (Åke Larsson, personal communication) and was discharged directly from cellulose works in Lake Vänern (Lindqvist 1991). Flue gas cleaning systems were installed on all Swedish waste incinerators from 1985 to 1992 (Lindqvist 1991), which has probably reduced the relative importance of waste incineration. Diffuse sources of Hg include landfill deposits of slag and waste, dentistry, hospitals, chemical laboratories, the production of electrical equipment and paint applications.

The lead input to the Swedish environment has been studied by Bergbäck and others (1992). The main sources were found to be metal products and chemicals at the end of the 19th century, cables and batteries since about 1920, the use as petrol additive since 1946 (Cato 1977) with emissions peaking around 1970, and industrial emissions to air and water, which culminated in the 1970s. Metal discharges have been decreasing since then because of improved controls. It appears that, at present, Pb is mostly supplied to the environment by diffuse sources. In the densely populated area of Göteborg the soil has a mean Pb content twice that of background concentrations (Bergbäck and others 1992).

Organic matter discharges from sewage and the paper industry to the river were very high during the 20-year period 1955–1975 (Fig. 8). Because sewage and paper production waste products also contained considerable amounts of pollutants, this trend is presumably connected to a heavy metal discharge trend. Furthermore, organic matter scavenges pollutants, so that high levels of pollution are expected in sediments from the 1950s to 1970s.

Environmental measures

Environmental measures were first taken in the 1950s and 1960s, concentrating on reducing the discharge of organic matter. Before the Ryaverket sewage treatment plant was built in 1972, nearly all sewage was discharged untreated to the Göta älv River. The treatment of the sewage resulted in a rapid decrease of organic matter in the river water (Fig. 8). In addition, the paper industry has reduced discharges since the beginning of the 1980s (Åke Larsson, personal communication). The early sewage treatment probably did not remove heavy metals as efficiently as the organic matter. Therefore, discharges of heavy metals from Ryaverket have decreased mainly between 1980 and 1985, when improvements in sewage treatment and the decoupling of industrial discharges from the municipal system took place (Göteborg regional sewage work, unpublished data).

In the 1970s and 1980s industrial heavy metal discharges were also reduced. From river water concentrations, discharges of 200 kg/year Hg in 1973, 50 kg/year in 1974,

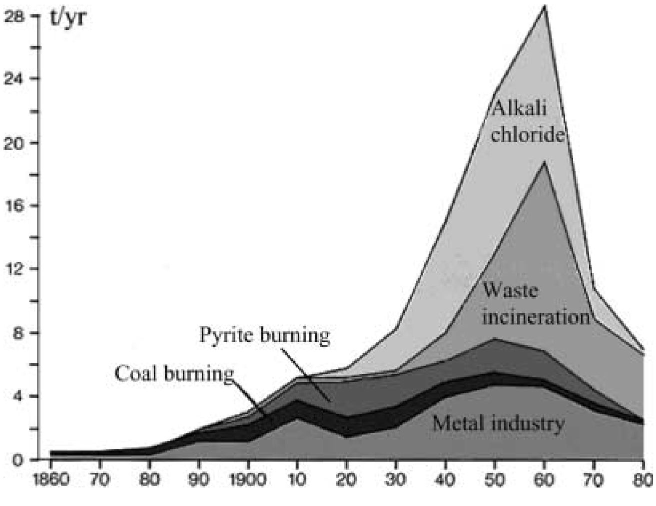


Fig. 7
Discharge of mercury (t/year) to the atmosphere from different sources in Sweden under the period 1860–1980. (Lindqvist 1991)

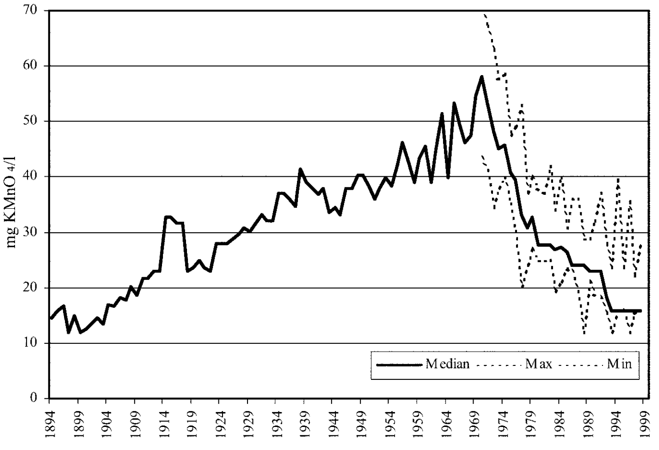


Fig. 8
$KMnO_4$-uptake as a measure of organic matter in the water of the Göta älv River. (The Göta River Water Protection Association, unpublished data)

8.25 kg/year in 1976, and negligible discharge in 1977 has been estimated (The Göta River Water Protection Association, several yearly reports). If all available information on the emission of mercury through time is combined, it appears that a major increase occurred from the 1930s to the 1960s (Fig. 7 and Jensen 1997). Atmospheric emissions have decreased since then, returning to the level of the early 1930s by 1970. The reduction of industrial discharges to the river started later, probably at the beginning of the 1970s.

To conclude, notably high organic matter concentrations in sediments are to be expected from the 1950s to the 1970s, with a peak around 1970, and high heavy metal concentrations from the 1930s to the 1980s. From the data presented in Jensen and Jensen (1991) and Jensen (1997), it also appears that there might have been a slight increase in heavy metal concentrations in the early 1990s.

Comparison of sediment data with historical trends of human intervention

With this background information in mind, it is now possible to refine the dating of the heavy metal profiles. Based on the interpretation of the ^{137}Cs and ^{210}Pb profiles regarding minimal and maximal possible sedimentation rates, we estimated the intervals with distinct heavy metal enrichment to date from ~1935 to 1980. In core 5, the timing of the onset of pollution is unclear, and in core 32 it is not preserved. However, the age estimated for the level at which heavy metal concentrations decrease agrees very well among the three cores. This is presumably connected to the decrease in heavy metal supply because of improved sewage treatment and reductions in industrial discharges, as described above. It is therefore reasonable to assume that the onset of increased discharges is also preserved in the sediment, especially as the estimate of 1935 from core 2 agrees well with the literature data.

It is similarly assumed that the metal enrichment in core 5 started about 1935, which is in accordance with the sedimentation rate of 1.2 cm/year predicted by the interpretation of the ^{210}Pb profile. Also, the possible position of the Chernobyl-influenced content of ^{137}Cs in the sediments at about 17 cm strengthens this interpretation of the heavy metal profile. The Hg peak at 52 cm is consistent with high inputs about 1960, and the small peak at 8 cm would date to the 1990s. The increase in sedimentation rates and the correlation of all heavy metals with the clay content in this core (r=0.65 to 0.86) imply that the peak at 8 cm originates from a heavy metal flux in the same order of magnitude as in the lower part of the core.

Using the above interpretations, the lower, recent-sediment boundaries in cores 2 and 5 seem to be non-synchronous. River regulation changes in 1916, 1934 and 1937 have probably increased sediment transport through increased erosion in and along the river (Brack and others 2000) and thus caused an onset of sedimentation in the outer estuary. The regulations in 1916 and 1937 increased erosion mainly upstream from Lilla Edet (Fig. 1), whereas the regulation in 1934 increased discharge in the Göteborg-branch of the river (Sundborg and Norrman 1963). This might have caused transport of suspended material further out into the southern outer estuary than before 1934, and an onset of sedimentation at site 5 around that time.

From the considerations above it is clear that environmental measures have decreased the supply and accumulation of heavy metals. It has to be considered, however, that all cores show a coarsening upward trend, and possibly also an increase in sedimentation rates, such as is interpreted in core 5. Therefore, heavy metal concentrations in the sediments may be diluted and probably do not linearly reflect the original flux of heavy metals. Also, there have been recent episodes of heightened discharge, as indicated by the Hg peak in core 5. Nevertheless, the decrease since the 1970s is so pronounced that a remediation effect is the most logical explanation.

The historical trends of heavy metal accumulation in sediments in the Göta älv estuary agree well with results of other studies in Sweden (Renberg and others 1990; Bergbäck and others 1992), the Skagerrak (Longva and Thorsnes 1997) the North Sea area (Kersten and others 1988), and also in other industrialised areas outside Europe (Bricker 1993; Valette-Silver 1993; Rood and others 1995). The common general trend is an increase starting in the beginning of the 20th century and peak values during the 1960s and 1970s. This is probably because of a similar development in industry and economy in the highly developed industrial countries, and is enhanced by atmospheric long-distance transport of pollutants (Rood and others 1995).

Conclusions

The reconstruction of historical pollution trends in the Göta älv estuary is possible if disturbed intervals are identified and taken into consideration. The estimates from three approaches, ^{210}Pb dating, ^{137}Cs dating and comparison of historical discharge trends with heavy metal profiles, agree well with each other. Disturbances in this setting are mostly caused by dumping of dredged sediments, land-slides and resuspension or loss of the sediment surface during sampling. Bioturbation is considered to be generally low, but can affect the ^{210}Pb and ^{137}Cs profiles. The historical pollution-supply trends match the trends that are found in the sediment record. The most polluted sediments were deposited between 1935 and 1980, but episodic discharges have also occurred lately. Hg is not only more enriched than the other heavy metals Cu, Pb and Zn, but has also had high levels during longer time spans. Another difference between Hg and the other heavy metals is its higher variability, expressed as a relative standard deviation. This probably reflects different sources and transport and sedimentation mechanisms. High heavy metal concentrations are usually associated with high organic matter concentrations, which is caused by both scavenging and synchronous anthropogenic discharge of organic matter and heavy metals. Concentrations in the sediments do not exactly mirror the original flux of heavy metals. Nevertheless, the decrease since the 1970s is so pronounced that a remediation effect is obvious. In an environment that is becoming cleaner, the contaminated sediments may become increasingly important pollution sources, especially if they are brought into contact with water by dredging or turbulence related to shipping.

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