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Long-term Heavy Metal Loading to Near-Shore Lake Sediments

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Abstract Long-term sediment pollution by lead (Pb), copper (Cu) and zinc (Zn) is investigated in Karlskärsviken, a typical bay of Lake Mälaren, about 10 km northwest of Stockholm. We find that metal accumulation rates in the sediments of Karlskärsviken increase considerably more in the outer than in the inner bay section, all the time since medieval times and even more so after than before industrialisation. Resulting metal accumulation rates are dominated by increasing sediment accumulation rate and its spatial–temporal patterns. This indicates significant waterborne metal pollution contributions to the bay sediments, which continue until present times and are still increasing for Zn and Cu. The outer Karlskärsviken bay section is then mainly affected by regional waterborne discharges, while the inner bay section is mainly affected by local discharges, in addition to the atmospheric deposition that affects the whole bay and Lake Mälaren similarly. The present results indicate possible important shortcomings in environmental regulation, management and monitoring systems that consider only sediment concentration levels and neglect pollutant accumulation rates and diffuse metal load contributions from slow subsurface transport into surface water environments.

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1 Introduction

In undisturbed environments, heavy metals exist only in low concentrations. In higher concentrations, metals are hazardous for humans, animals and plants, with one rule being that the lower the natural concentration is, the higher are the toxic effects of increased concentrations. When mining and using rocks for metallurgic production, metal concentrations in the environment increase both by atmospheric deposition (Brännvall et al. 2001; Bindler et al. 2002) and because water flowing through mine wastes becomes polluted by acidity and heavy metals and carries the pollution further to downstream to water environments [e.g., Herlihy et al. 1990; Younger et al. 1997; Iribar et al. 2000; Younger 1997, 2001; Milu et al. 2002; Environmental Regulation of Mine Waters in the European Union (ERMITE) Consortium 2004]. In addition to mine wastes, some studies suggest that also abandoned mine voids may yield large water pollution by acidity and heavy metals (Younger et al. 1997; Wood et al. 1999; Younger 2003).

Recent studies of metal transport from mine wastes show further that large transport variability and delay may occur in the subsurface water system (Eriksson and Destouni 1997; Berglund et al. 2003; Malmström et al. 2004, 2007; ERMITE Consortium 2004). Slow and highly variable transport of pollutants through the subsurface water system imply that this system may constitute an important diffuse source of pollution for downstream surface water environments long after abandonment and closure of mining activities. A suite of recent basin-scale studies indicate, for instance, that slow subsurface pollutant transport may yield large and increasing pollutant load contributions to downstream surface and coastal waters (Baresel and Destouni 2005, 2006; Baresel 2007; Lindgren et al. 2007).

Other studies have further shown that the metallurgical development in Europe during the last 4000 years is archived in lake sediments (Brännvall et al. 2001). In Sweden, for instance, Pb pollution history has been established by analyses of sediments in small lakes without any nearby industrial pollution pressures, implying that the found anthropogenic metal loading in the lake sediments has there occurred through atmospheric deposition (Bindler et al. 2002). This small-lake history follows a characteristic pattern for Pb concentrations: first signs of Pb increase already about 3000 B.P., followed by relatively low concentrations until the industrial revolution in the middle of the 19th century, a slow rise from then until World War II, followed by a temporary decrease caused by reduced production and non-petrol based transports in Europe, and then a quick rise until the oil crisis in the 1970s. Thereafter, lead pollution decreases again, as a result of lead-free petrol and exhaust cleaning in the combustion industry.

However, in the sediments of the deep basin S. Björkfjärden of the large Swedish Lake Mälaren, west of Stockholm (Fig. 1), lead contamination has not been found to show the same pattern, but exhibits instead an early rise in the Middle Ages, due to impacts of pre-industrial mining in Bergslagen, a main mining district west of Lake Mälaren (Renberg et al. 1994, 2001; Brännvall et al. 2001). The already high metal concentrations in the sediments of S. Björkfjärden in Lake Mälaren due to these impacts obscure then the later industrial lead pollution impacts since the middle of the nineteenth century, which are characteristic for most smaller Swedish lakes that have not been affected by earlier pollutant sources.

In general, the history of metal mining in Sweden is long and started approximately 3000 B.P. or earlier with numerous small furnaces used for household production of bronze and iron (Jaanusson and Wahlne 1975; Hjärtner-Holdar 1993). The raw material for this production contained also other metals, such as Cu, Zn and Pb, but until the very beginning of 1300 A.D., the mining and its metal pollution effects were limited, because of the relatively low-intensive mining technology and limited scale of production. Around and after 1300 A.D., (Den Svenska Historien 1977) however, mining technology was improved and the Swedish mining industry progressed in full speed, leading to the early increase of lead pollution that has been found in the sediments of the deep S. Björkfjärden basin of Lake Mälaren and interpreted to originate from such intensified pre-industrial mining and metal processing in primarily the Bergslagen mining district (Renberg et al. 2001).

With industrialization came a greater supply of metal products, generally contributing to increased metal contents in water, sediments and soil from the mining and industrial processing, as well as by associated industrial waste impacts. In the middle of the twentieth century, Pb in petrol was the main Pb polluting source, which has now been reduced. In contrast, the use of Cu and Zn in various products is still increasing, for instance in buildings and impregnated wood products. Furthermore, numerous abandoned mine waste dumps and mine voids have been accumulating in and around mining districts from both pre-industrial and industrial mining until modern times, when environmental regulation started to require pollution abatement measures for at least known modern mine waste deposits. Considerable downstream water pollution, however, may still occur from older and yet un-remediated mine and other waste deposits and abandoned mine voids. Abandoned mine voids and numerous unknown old mine waste dumps, for instance, are not explicitly regulated in Sweden or the EU [Swedish Environmental Protection Agency (SEPA) 1998; Salmon and Destouni 2001; European Commission 2003], yet may yield significant long-term metal load contributions (see, e.g., Eriksson and Destouni 1997; Berglund et al. 2003; Malmström et al. 2004, 2007; ERMITE Consortium 2004 for more detailed explanations of the slow transport-transformation processes that may yield such long-term contributions) and have important implications for the success and efficiency of water pollution abatement (Baresel et al. 2006; Baresel and Destouni 2007).

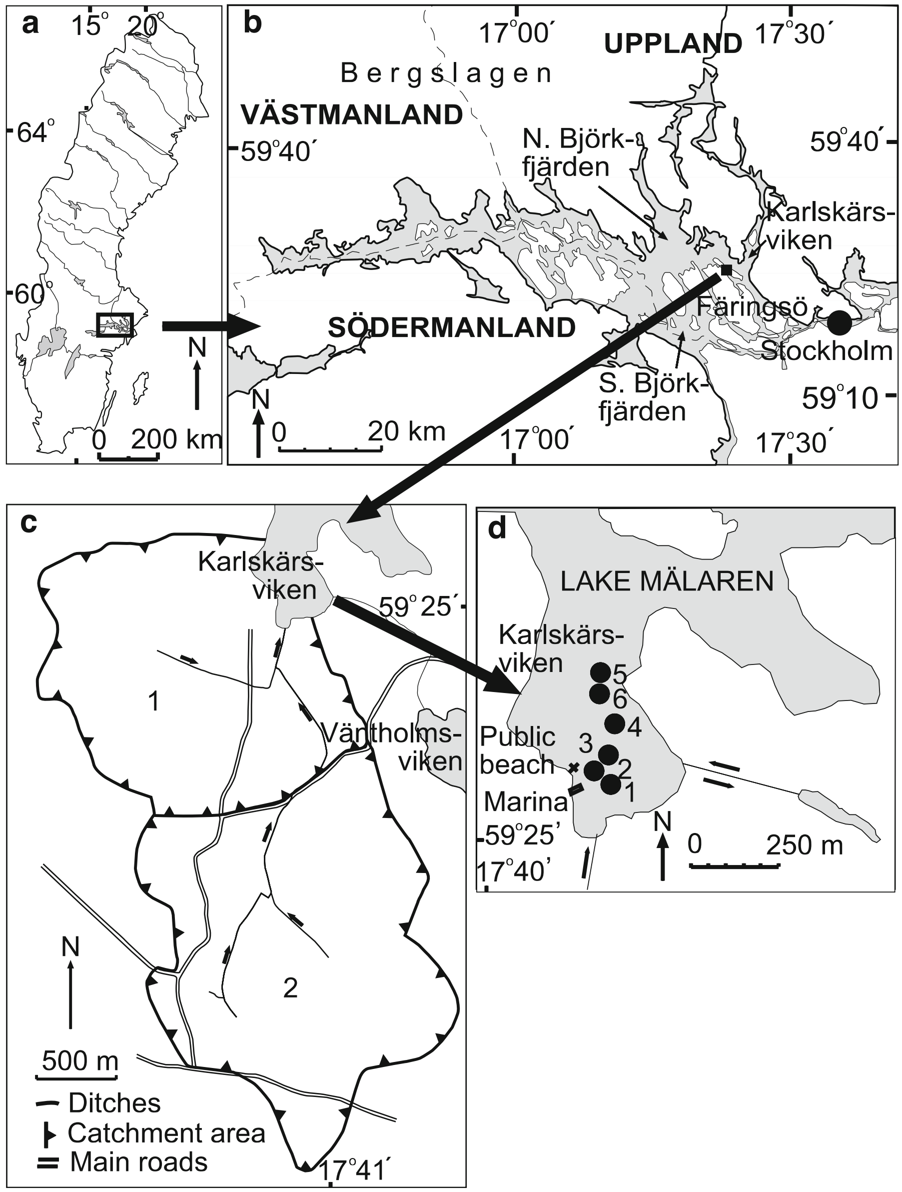

Fig. 1 **a** Sweden. **b** Lake Mälaren with Karlskärsviken to the north of Färingsö. **c** The catchment area to Karlskärsviken and **d** Karlskärsviken with the location of the sampling points. Maps by Sven Karlsson

The present study investigates if Lake Mälaren has been and may still be significantly impacted by such long-term metal load contributions from the subsurface water system into the lake or into streams that discharge into the lake, in addition to the airborne metal pollution from pre-industrial times that has already been found to have greatly affected Lake Mälaren sediments (Renberg et al. 1994, 2001; Brännvall et al. 2001). Long-term waterborne metal load contributions may be expected, for instance from all the mine waste dumps and voids that have been accumulating in the region since the nineteenth century industrial intensification in the Bergslagen mining district until present times. These contributions may not have been found so far because pathways of their waterborne transport discharge into and affect near shore sediments more than the previously investigated deep basin sediments of large lakes such as Lake Mälaren.

Furthermore, Swedish environmental monitoring is sparse, with on average only one metal concentration monitoring point per 6,700 km^2 and no particular clustering of monitoring wells in and around main mining areas (Destouni 2005; Karlsson 2005; Ojala

2005; Baresel 2007). Such monitoring essentially neglects subsurface water quality and is more likely than not to miss important pollutant load contributions from subsurface to surface waters. For the present investigation purposes, we therefore study the possible metal load impacts and temporal development directly in the sediments of a typical near shore zone of Lake Mälaren, the bay of Karlskärsviken, located about 20 km west of Stockholm on the island of Färingsö (Fig. 1). This bay has recently been investigated with regard to the sediment and phosphorous inputs and accumulation in its near-shore sediments (Olli 2007, 2008), which provides useful background information also for the present study.

2 Materials and Methods

2.1 General Site Description

The bay Karlskärsviken of Lake Mälaren covers about 20 ha and has an average depth of about 3 m; the depth difference between the outer and the inner section in this investigation is about 0.5 m. The bay's local surface water catchment area is about 560 ha (Fig. 1c). It is dominated by farmland, which is drained by a main ditch with its outlet into Karlskärsviken. There is no population centre or industry, except commercial gardens, in this catchment area. Close to the ditch outlet, which is also the main outlet of the bay catchment, the ditch is presently overgrown by macrophytes, and organic and inorganic particles are deposited in the macrophyte zone (Benoy and Kalff 1999). The present western shore of Karlskärsviken has also a broad transition zone with reeds between the open water and the low lying shore meadows. The eastern side is rocky and steep.

Before the Middle Ages, Lake Mälaren was a bay of the Baltic Sea with brackish water. Thereafter it changed into a major freshwater body, with no seawater entering the bay Karlskärsviken. During the Middle Ages, Karlskärsviken was not a bay of Lake Mälaren but part of a strait between N. Björkfjärden andVäntholmsviken of today (Fig. 2).

2.2 Metal Sources

Metal pollution into water may generally have different sources, including: (1) natural lead pollution from weathered bedrock; (2) airborne pollutant transport from anthropogenic sources; and (3) waterborne pollutant transport through both focused surface and diffuse subsurface water pathways, originating from both nearby local and more faraway regional anthropogenic sources. Pollution sources may at least roughly be estimated by isotope analysis. For instance, the isotope ratio $^{206}Pb/^{207}Pb$ is lower when the origin is anthropogenic activities and higher when

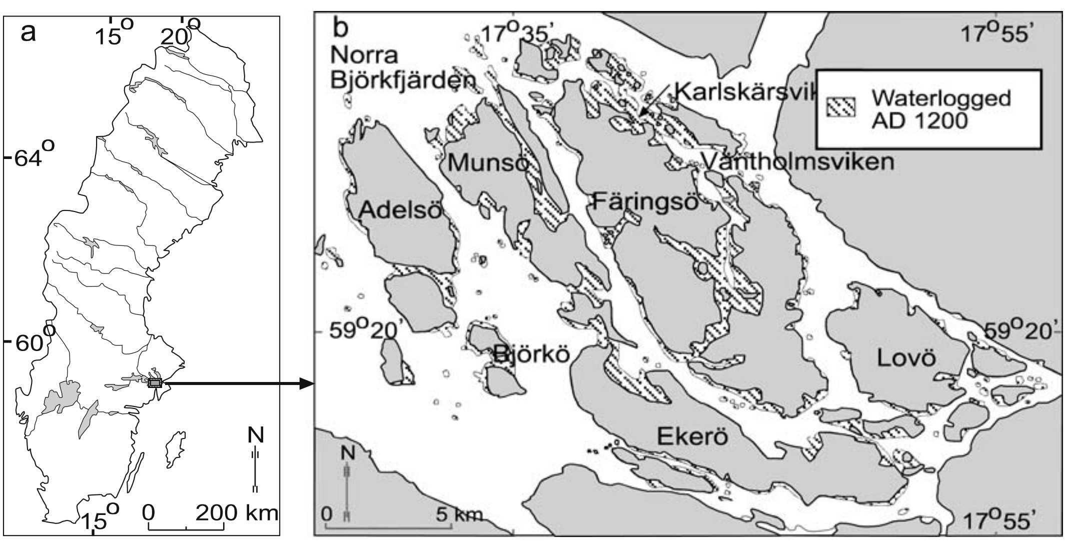

Fig. 2 **a**. Sweden. **b**. Ekerö municipality with the medieval shoreline. Karlskärsviken was a part of a strait at that time. The *dotted line* is the present shore. Map by Sven Karlsson

lead originates from bedrock weathering, and Pb isotope analysis performed for Lake Mälaren (Östlund et al. 1998; Renberg et al. 2001) shows that higher metal concentrations during industrial time coincide with lower $^{206}Pb/^{207}Pb$ ratios, which imply anthropogenic sources.

In its Middle Age position, as a strait between N. Björkfjärden and Väntholmsviken of today (Fig. 2), at least the outer section of Karlskärsviken must have been directly exposed to drainage water inputs from parts of the Bergslagen mining district. After that, the strait was silted up, and since 1951 there are no major surface water flows into the former strait extent between Karlskärsviken and Väntholmsviken. Similarly, the strait to the northwest of Karlskärsviken has also decreased. However, delayed groundwater transport of metals from old abandoned mine wastes and voids may still make significant metal load contributions directly and indirectly into the lake.

Furthermore, before 1953, local waste products were deposited directly on the west part of the shore of Karlskärsviken, which is now used as a public beach. The public beach was constructed during 1951–1953, along with construction of a marina and deepening of the main drainage ditch discharging into the bay. All these activities increased the sediment accumulation rate in the inner section of the bay significantly in the period 1951–1953, as shown in Fig. 3 based on the reported detailed sediment accumulation analysis by Olli (2008). Exhaust emissions of lead from the marina and later repairs and extensions of the marina, along with the known construction of a new private bridge in the 1990s and frequent use of impregnated wood in local shore and near-shore constructions may have continued to yield significant local metal loading to the bay even after the main construction period 1951–1953 and subsequent stabilisation or decrease of sediment accumulation rate (Fig. 3).

2.3 Field Work

To assess the metal contamination of the lake sediments, sediment cores were taken from the bay Karlskärsviken (Fig. 1d) during winter time over the period 2001–2003, when the bay was covered by ice. All the core locations are outside the macrophyte zone. In the macrophyte zone, the sediments have been dredged and no historical information is archived there. One meter overlapping sections were retrieved with a Russian peat corer. A turbidity-meter was used to measure the water depth and to know where to start the coring. Cores 1 and 2 measured 1 m in length and core 5 measured 2 m in length. Core 4 was 10 m, in order to establish the sediment accumulation since the retreat of the ice cover. Core

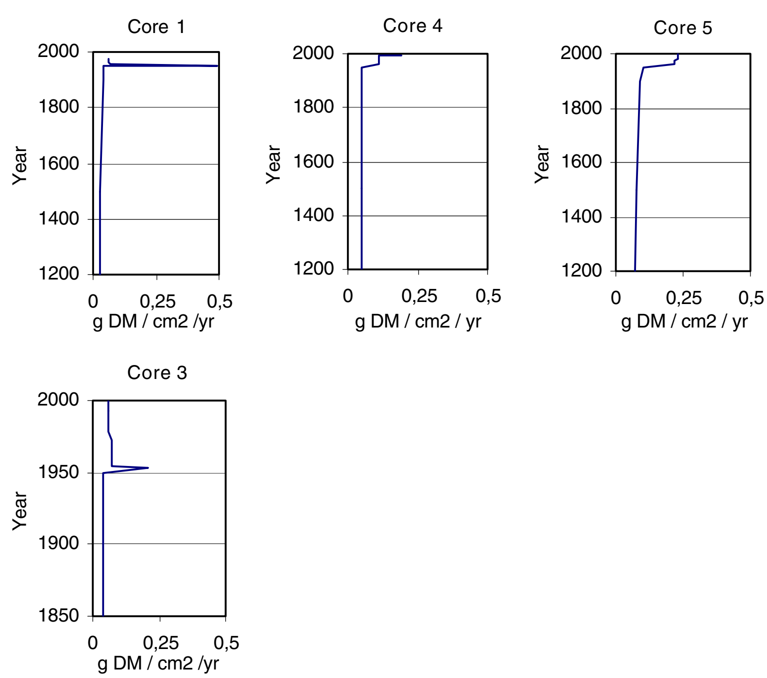

Fig. 3 Sediment mass accumulation rates in the cores 1, 3, 4 and 5. The dates were estimated by SCP analysis as reported in detail by Olli (2008)

2, close to core 1 in the inner section of the bay, was originally taken to study possible pollution from a former near-coastal waste deposit, but appeared to be taken too close to a dredged area and did not archive the chronological history between 3 and 40 cm depth. Cores 3 and 6 were taken with a freeze corer, which restricted the length of these cores as the corer is just dropped down. The length of core 3 was 48 cm and core 6 was 65 cm.

2.4 Laboratory Work

The cores were sectioned into 1 cm slices down to 70 cm and then 2 cm slices. The samples were analysed with respect to Pb, Cu and Zn concentration according to the Swedish standard methods (Svensk Standard 1993) and determined with a flame atomic absorption spectrophotometer. To independently check the lead concentration results, samples from two cores, 1 and 5, were also analysed at the Archaeological Research Laboratory, Stockholm University. Accumulated metal rates (μg cm^{-2} $year^{-1}$) were calculated from metal concentrations and the sediment accumulation rates (g DM) cm^{-2} $year^{-1}$ [dry mass (DM)] that were found and reported by Olli (2008; Fig. 3). The cores consisted of clay and gyttja clay in the sediments from the last decades (in core 1 above 9 cm, core 3 above 18 cm, core 4 above 18 cm and core 5 above 22 cm). In all the cores, the carbon concentration was about 4% at 0 cm. The uppermost 5 cm of core 1 was empty. The recent (2000) metal concentrations in core 2 and also core 3 were therefore used for the year 2000 inner section assessment, due to their proximity to the most inner core 1.

2.5 Sediment Dating

Sediment dating for medieval times was based on the presence of brackish- and freshwater diatoms and for modern times on the number of fly-ash particles from fossil fuel, spheroidal carbonaceous particles (SCP) and concentration of the caesium isotope Cs-137. The transition from brackish to freshwater was elongated by various transgression and regression phases from about 800 to 1200 A.D., which have been recognized in other flora stratigraphy studies Åse (1980; Miller and Robertsson 1982; Ambrosiani 1981; Miller and Hedin 1988) and also here in Karlskärsviken. The preparation of diatom slides for analysis followed methods described by Battarbee (1986). The freshwater and brackish water diatom frustules were counted, and at the levels where the freshwater diatoms were 70–80% of the total amount of diatoms the water was considered to be freshwater.

The history of SCP presence in sediments, upon which the modern sediment dating is based, was established by Griffin and Goldberg (1975, 1979), Wik (1992), Odgaard (1993), Rose (1990, 2001), Rose et al. (1995, 1999), Bindler et al. (2002), Renberg et al. (2001). The SCP start to appear in sediments about 1850 A.D. and SCP dating captures the World War years very clearly and also the European industrial progress and crises, and finally also the progress in eliminating the exhausts from fossil fuel combustion. If Pb pollution is primarily through deposition of airborne particles and there is no main local-regional Pb polluter, the increase and decrease of Pb concentrations are expected to coincide with the increase and decrease of SCP in modern times (Wik 1992; Bindler et al. 2002). For SCP counting, the sediment samples were prepared according to Rose (2001).

Concentration of the caesium isotope Cs-137 was determined in core 3 at the Studsvik Nuclear Laboratory. In the Mälaren region, the concentration of Cs-137 increased in the sediments when the Chernobyl accident occurred in 1986. Knowledge of local activities during the second half of the twentieth century, including the known main construction period 1951–1953, when the main drainage ditch was deepened and the public beach and the marina were constructed, as well as the building of a new private bridge (using impregnated wood) in the 1990s, have also provided useful information for chronological control.

3 Results

At medieval times, Pb concentrations in the sediments were on average about 25 μg Pb (g $DM)^{-1}$ where after they increased slowly until industrial time (Fig. 4, Table 1). In the whole bay of Karlskärsviken, Pb concentrations increased further considerably also after the start of industrialisation, which was an effect that has not been seen in the deeper S. Björkfjärden sediments of Lake Mälaren, where earlier Pb loading

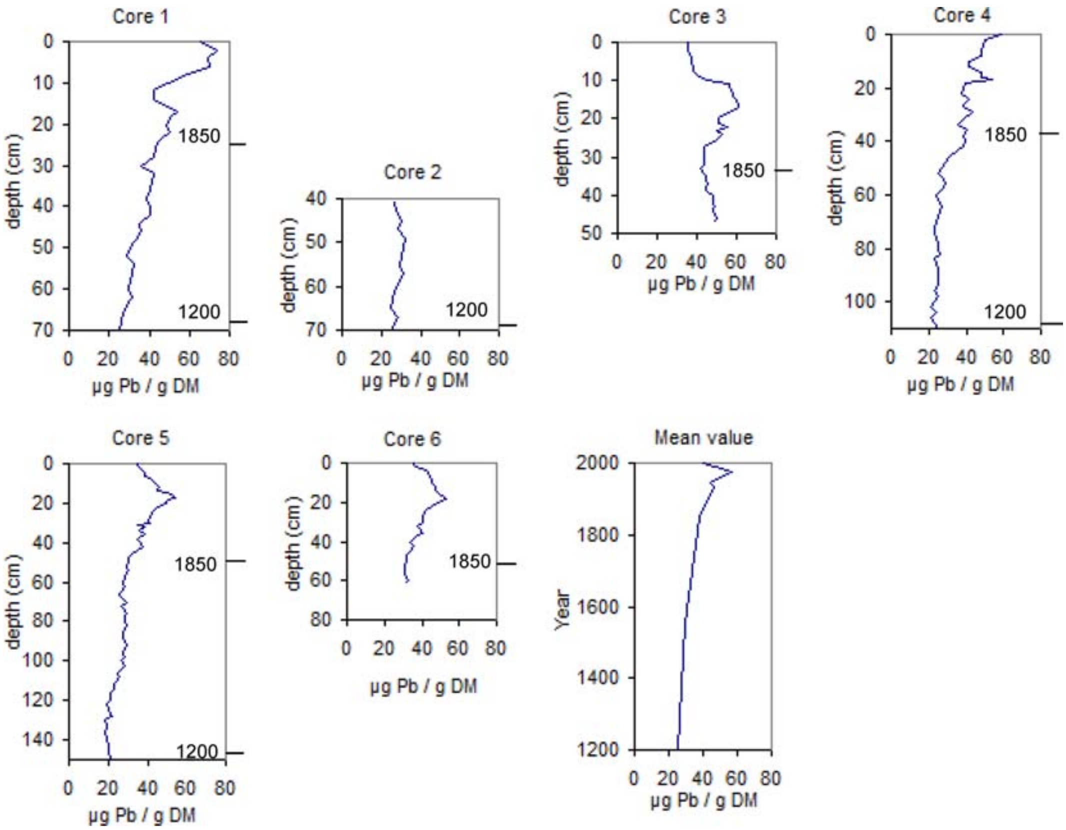

Fig. 4 Pb concentration versus depth and mean concentration versus dated time in the sediments from Karlskärsviken. The different panels are positioned and scaled so as to be comparable with respect to time. The sediments in core 2 were perturbed at 1–40 cm depth

obscured the later industrial time loading (Renberg et al. 2001). The Pb concentrations in Karlskärsviken continued to increase slowly until about 1950 and then quickly accelerating until about 1973, after which they decreased somewhat, conforming to the typical pattern for small Swedish lakes that lack significant local waterborne Pb loading (Bindler et al. 2002). The expected Pb decrease during World War II was detected in all cores and also for Zn (Tables 1 and 2).

The average Pb mass accumulation rate in the whole Karlskärsviken bay was further estimated to have been around 1 μg Pb cm^{-2} $year^{-1}$ in medieval times (Table 3, Fig. 5b–c). In the sediments deposited after medieval times, Pb mass accumulation rates increased, similarly to Pb concentrations. Around 1973, Pb mass accumulation rate was estimated to have increased to about 4.4 μg Pb cm^{-2} $year^{-1}$ in the inner section (core 1, Table 3) and much more so, to about 5–12 μg Pb cm^{-2} $year^{-1}$, in the outer section of the bay (cores 4–6, Table 3). In 2000, Pb accumulation rate was finally estimated to have decreased to about 2.1 μg Pb cm^{-2} $year^{-1}$ in the inner section

Table 1 Pb concentration [μg Pb (g DM)$^{-1}$], with corresponding depth (cm) in parentheses

Year/core	1	2	3	4	5	6	Mean value
2000		36 (0)	35 (0)	58 (0)	35 (0)	35 (0)	40
1973	69 (4)		61 (15)	49 (16)	54 (17)	49 (18)	57
1945	50 (16)		50 (23)	40 (27)	35 (31)	38 (32)	44
1935	54 (17)		53 (24)	43 (29)	38 (33)	40 (34)	46
1850	45 (25)		42 (33)	39 (38)	31 (49)	31 (51)	38
1200	28 (68)	27 (66)		23 (108)	22 (148)		25

Table 2 Cu–Zn concentrations [μg (g DM)$^{-1}$] in Karlskärsviken bay sediments since mediaeval times

Year/core	1	2	4
2000		84–155	59–240
1973	139–208		42–156
1945	38–112		42–132
1935	39–120		42–153
1900	33–104		39–141
1850			40–135
1200		24–60	

(cores 2–3, Table 3), while it remained greater at about 7–8 μg Pb cm^{-2} year^{-1} in the outer section (cores 4–6, Table 3).

The around three times increase in the inner bay section, from 0.8 μg Pb cm^{-2} year^{-1} in medieval times to 2.1 μg Pb cm^{-2} year^{-1} today, is typical for small Swedish lakes (Bindler et al. 2002). In contrast, the outer section increase, from 1–1.5 to 7–8 μg Pb cm^{-2} year^{-1}, exceeded considerably that expected by only atmospheric deposition (Renberg et al. 1994; Brännvall et al. 2001; Bindler et al. 2002). This indicates significant waterborne Pb loading to the outer bay section, in addition to atmospheric Pb deposition.

According to SCP dating and detailed sediment accumulation analysis by Olli (2008), when the main drainage ditch into the bay was deepened and the marina and public beach were constructed in 1951–1953, the sediment accumulation rate increased greatly in the inner bay section (Fig. 3). This resulted in a decrease of metal concentrations in the sediments, especially in core 1. After that, Pb and even more so Cu and Zn concentrations and accumulation rates increased until 1973 (Tables 1, 2, 3, 4 and 5). In the last decades, Cu and Zn accumulation rates decreased in the inner section, while they continued to increase further out in core 4 (Fig. 5b, Table 4). On average for the whole bay, mean Cu and Zn accumulation rates continued to increase after 1973 until present time,

Table 3 Pb mass accumulation rate (μg Pb cm^{-2} year^{-1}) since mediaeval times

Year/Core	1	2	3	4	5	6
2000		2.16	2.1	7.03	8.05	8.17
1973	4.4		4.2	5.32	11.8	11.7
1945	2		2	1.95	3.5	3.77
1935	2.16		2.12	2.15	3.8	4
1850	1.32		1.68	1.9	2.4	2.77
1550	0.9			1.2	2.24	
1200	0.84	0.81		1.15	1.47	

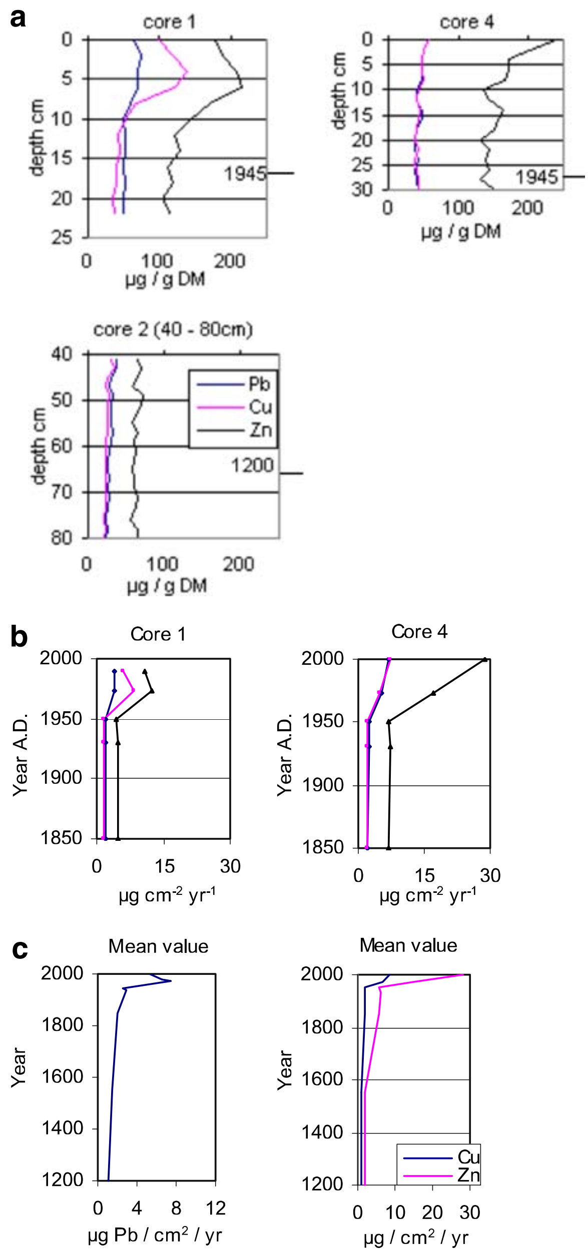

Fig. 5 **a** Pb, Cu and Zn concentrations [μg (g DM)$^{-1}$] versus depth. **b** Pb, Cu and Zn accumulation rates (μg cm^{-2} year^{-1}) versus dated time, and **c** mean values of Pb, Cu and Zn accumulation rates (μg cm^{-2} year^{-1}) versus dated time, in the sediments of Karlskärsviken. Core 2 is disturbed between 1–40 cm, and therefore not taken into account in this increment

Table 4 Cu and Zn mass accumulation rates (μg cm^{-2} $year^{-1}$); corresponding graphs are shown in Fig. 5b, along with related Pb mass accumulation rates

Year/core	1	4
2000	5.01–9.32	7.1–28.8
1973	8.33–12.5	4.67–17.2
1950	1.54–4.48	1.89–6.9

while mean Pb accumulation rate started to decrease around 1973 (Fig. 5c, Table 5).

4 Discussion and Conclusions

In general, we find here that metal accumulation rates in the sediments of Karlskärsviken increased considerably more in the outer than in the inner bay section, all the time since medieval times and even more so after than before industrialisation. Similar temporal patterns were also exhibited by the metal concentrations, but without the differences between the inner and the outer section, and without the large recent increases that were exhibited by the metal accumulation rates. A main reason for these differences between metal concentrations and metal accumulation rates may be that in Karlskärsviken, not only the metal concentrations in the accumulating sediments, but also the sediment accumulation rates have increased since medieval times and even more so during industrial times (Fig. 3; Olli 2008). Furthermore, the sediment accumulation rate increase has been sustained until present times in the outer section of the bay, whereas in the inner section there was a clear sediment accumulation rate peak during the period 1951–1953 of main local constructions (Fig. 3). Parts of the differences in Pb sediment concentrations between the inner and outer section may be explained as a particle size distribution effect, but this effect does not explain the differences in Pb mass accumulation rates. In medieval times, the sediment accumulation rate in the inner section was about 55% of that in the outer section. This relation remained until about 1950 before it decreased to the present value of only 27%. Consequently, in Karlskärsviken, sediment accumulation rate is a better descriptor of metal mass loading than the metal concentrations in the sediments.

The higher sediment accumulation rate in the outer than in the inner bay section, except for the period 1951–1953 when sediment accumulation rate increased also in the inner section due to the known main local construction works (Fig. 3, Olli 2008), explains the generally greater metal accumulation rate in the outer bay section relative to metal concentrations. The short large increase of sediment accumulation rate during the period 1951–1953 is reflected also in simultaneous decreases of SCP, C (Olli 2008) and BSi (Olli 2007) concentrations in the sediments. All these simultaneous anomaly signals are clear indicators of increased deposition of perturbed minerogenic material and transported sediments from the bay's local catchment area into the lake. Furthermore, the short inner-section sediment accumulation rate increase during 1951–1953 explains also why the differences between the inner and outer sections increase after this main local construction period. Especially in the last decades, the macrophyte zone has grown into the main local drainage ditch, retaining transported sediment particles within the ditch and decreasing the sediment transport to and accumulation rate in the inner bay section after the 1951–1953 peak. The high values of Cu and Zn in core 4 in the beginning of the 1990s are also explained by known local construction activities at that time, including dredging and construction of a new bridge made of impregnated wood.

The dominance of sediment accumulation rate in explaining metal accumulation rates indicates con-

Table 5 Mean Pb, Cu and Zn mass accumulation rates (μg cm^{-2} $year^{-1}$)

Year	Mean value Pb	Cu	Zn
2000	5	6.05	19.06
1973	7.3	6.5	14.82
1950	2.6	1.72	5.69
1935	2.85	1.76	5.96
1850	2	1.76	5.72
1550	1.31	0.8	2
1200	1.07	0.81	2.09

Mediaeval times values exist for all metals Pb, Cu and Zn in the cores 1 and 2. Core 1 (and 2 for 2000 values) represents the inner section of the bay and core 4 the outer section in this joint mean value; corresponding graphs are shown in Fig. 4c siderable waterborne metal pollution contributions to the bay, which continue until present times and are still increasing for Zn and Cu. The outer Karlskärsviken bay section is then mainly affected by regional waterborne sediment discharges, while the inner bay section is mainly affected by local sediment discharges, in addition to the atmospheric deposition that affects similarly the whole bay and Lake Mälaren.

With regard to metal concentrations in the sediments, potential risk levels for biological effects of sediment-adsorbed metal contaminants are for the first step in the food chain proposed by Long and Morgan (1990) to be: possible risk for Pb 42 μg (g DM)$^{-1}$, Cu 70 μg (g DM)$^{-1}$ and Zn 120 μg (g DM)$^{-1}$; and probable risk for Pb 110 μg (g DM)$^{-1}$, Cu 390 μg (g DM)$^{-1}$ and Zn 270 μg (g DM)$^{-1}$. Higher up in the food chain, risk levels increase. In all of the present cores, Pb has been at the possible risk level since the beginning of the twentieth century until about 1980, but is now below that level; it has further never exceeded the probable risk level and may now be on a decreasing trend. In contrast, Cu concentration is still increasing and exceeds so far the possible risk level in the inner section. For Zn, no concentration after the start of industrialisation falls below the possible risk level, and in the sediments of core 4, deposited around 2000, Zn concentration is close to the probable risk level.

Considering further the environmental pollution classification of the Swedish Environmental Protection Agency (SEPA; Naturvårdsverket 1991), we regard here the Middle Age concentration value as the background concentration of about 25 μg Pb (g DM)$^{-1}$ (before the Middle Ages the water in Lake Mälaren was brackish and Pb concentrations are not comparable with those in freshwater systems) and note that Pb concentration in recently deposited sediments is on average about 40 μg Pb (g DM)$^{-1}$. Such an increase of ≤1.6 (recent concentration/background concentration) is in the SEPA classification regarded as an insignificant perturbation. However, in terms of Pb accumulation rate, instead of concentration, the corresponding increase is about 5 (from about 1 μg Pb cm^{-2} year^{-1} in the Middle Ages to about 5 μg Pb cm^{-2} year^{-1} presently), which is much greater and would in concentration terms be considered significant. The Cu and Zn accumulation rates have further increased even more, from about 0.8 μg Cu cm^{-2} year^{-1} and 2 μg Zn cm^{-2} year^{-1} during the Middle Ages to about 6.1 μg Cu cm^{-2} year^{-1} and 19.1 μg Zn cm^{-2} year^{-1} presently, implying relative increases of 7.6 for Cu and 9.6 for Zn, which would in concentration terms be regarded as strong perturbations.

The large difference in environmental change classification between concentration and metal mass load terms is due to the large increase of sediment accumulation rate in the last century. Pollutant concentration change in sediments is only indicative of environmental pollution load change for constant sedimentation rate. However, such constancy does not apply in Karlskärsviken and is probably also generally inapplicable. Karlskärsviken is a typical bay of Lake Mälaren and similar metal loading behaviour may be found along most of the shore line of this large lake, as well as of many other lakes.

In general, the present results indicate possible important shortcomings of pollution classification and risk assessment systems that consider only sediment concentration levels and neglect pollutant accumulation rates. Moreover, they indicate that, whatever Cu and Zn pollution abatement measures that have been employed regionally and nationally in Sweden so far, these measures do not appear to have been working very well. Rather than these abatement measures stopping or significantly reducing waterborne leakage and transport of metals to downstream water environments, it is the sediments of the water environments themselves that appear to have so far been retaining the metals. However, this retention may in the future also be reversed, for instance if sediment accumulation becomes too large and/or biogeochemical conditions shift, with potentially serious environmental and health effects. The present study combines with a series of other recent studies (Salmon and Destouni 2001; Destouni 2005; Karlsson 2005; Baresel et al. 2006; Baresel and Destouni 2007; Baresel 2007) in indicating that environmental regulation, management and monitoring may currently be neglecting essential diffuse metal load contributions from slow subsurface water transport into surface water environments.

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