Distribution of cadmium, copper and zinc emitted from a swedish copperworks, 1750–1900

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Abstract

The present distribution of Cd, Cu and Zn in the soils around and sediments downstream from the Åtvidaberg copperworks which closed in 1900, was investigated to estimate the duration of the environmental stress caused in the surrounding area. Sampling of soils together with literature data on sediments provided the necessary information along with a study of the historical context of the copperworks. Leaching rates from the A- to the B-horizon of the soil were estimated to 0.3% yr^{-1} for Cd and 0.4% yr^{-1} for both Cu and Zn. From the sediment and water data it can be concluded that the soils surrounding the copperworks, a century after the works closed down, still continue to load Lake Håcklasjön with Cd, Cu and Zn. For Cd, it is likely that the approximately 100 years of leaching has substantially reduced the soil reservoir, while for Cu and possibly Zn we are still at the beginning of the loading pulse from the soils of the area to the lake.

1. Introduction

Metals that are deposited within an area accumulate in the soil, which acts as a sink, retaining the metals for some time. Most metals then travel slowly down the soil profile and eventually reach the ground water. The next sink in the transport process is the lake sediment where the metals are incorporated and retained for a much longer time.

When considering the present dispersion of metals to the environment (as described by Nriagu, 1990 and Bergbäck, 1992) to the environment, it is of interest to determine the duration of the problem caused. In the long-term, the concentrations of a metal in a polluted soil will decrease if no more metals are deposited within the area. As the downward transport processes move the metals, the initial soil pollution problem gradually becomes a problem of ground water, surface water and sediment pollution. One approach to assessing the duration of this metal pollution process is to study the metal distribution in metal polluted areas where a century or more has passed since the cessation of the metal deposition. An important advantage with this method is that the leaching occurs in an undisturbed soil under natural conditions.

In an earlier study (Eklund et al., 1997), the Cd, Cu and Zn emissions from the Swedish Åtvidaberg

Copperworks have been estimated by means of archive studies and element analysis of tree rings. The copperworks were in operation between 1750 and 1900 during which about 900 000 Mg of sulphidic ore was processed to extract about 32 000 Mg of copper, thus constituting a significant source of metal pollution.

The method of copper extraction remained about the same during the whole period of operation and included five steps. First, the ore was calcinated in large heaps in order to remove the sulphur as sulphur dioxide. The available copper was thus converted to copper sulphide and zinc to its oxide. Volatile elements such as As, Sb but probably not Cd (GCA Corp., 1981) are likely to have been emitted during this step since the temperature of the heaps was probably between 600 and 800°C (Moore, 1990). During the subsequent steps, the smelting and the converting, a higher temperature was reached by driving enormous amounts of air through the furnaces. The smelt was stirred regularly, producing large particles which were emitted into the air as flue-dust, and which constituted the major part of the metal emissions from historical copper production

(Peters, 1907). The element composition of the flue-dust roughly follows that of the ore and accordingly, abundant elements of the ore, like Cu and Zn, dominate these emissions (Percy, 1861). The chemical characteristics of the element also affect their inclination to be emitted into the air. Volatile elements as As, Sb and Cd are likely to be over-represented in these emissions.

In Atvidaberg, some changes of the production methods took place during the 1840s. New furnaces, where the temperature could easily be regulated, and higher stacks were built (Bredberg, 1849) causing the emissions of flue-dust including Cu and Zn inevitably to decrease. Cadmium emissions were probably not as affected by these changes, since it is more difficult to control the volatile metal emissions than the ones of particulate matter (Coleman et al., 1979). The results from the oak tree-ring analysis in Atvidaberg support this hypothesis (Eklund et al., 1997).

In this article, the further fate of the metals emitted from Atvidaberg copperworks is examined by means of metal analysis of soils and sediments of the surroundings. The aim is to provide information
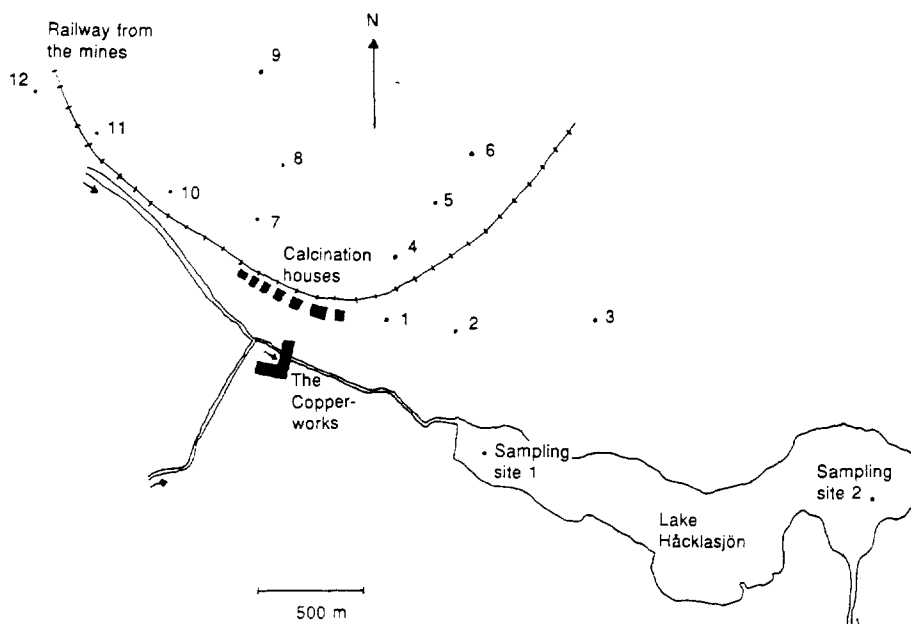

Fig. 1. The location of Åtvidaberg Copperworks, Sweden and the sites for sampling soil and sediments.

about the time span required for the metal transport from the source, over the soil pool to the sediment sink.

2. The study area

Åtvidaberg is situated in southeastern Sweden (58°10''N 16°E) within the county of Östergötland. The prevailing winds blow from the southwest. The main reason to why the copperworks was located here was the occurrence of a stream, where water power could be extracted for the running of the bellows. The ore was extracted, mainly at two different sites, 15–20 kilometres away. The soils close to the site of the copperworks are generally cambisols (brown soil) but with increasing distance podzols become more common. Soil samples for this study were collected in sites with undisturbed soil in the vicinity of the copperworks. Since the copperworks were located more or less within a town, Åtvidaberg, the soil sampling was restricted to four transects directed E, NE, N and NW (Fig. 1). Sediment data are from an earlier study (Qvarfordt, 1977), where two sites where sampled in the lake situated downstream, Lake Håcklasjön. This lake is shallow, eutrophic and receives the effluents from a sewage plant.

3. Methods

3.1. Estimate of the emissions into the air

Based on information from literature and archives about the quantities of copper produced (Fig. 2), emissions into the air have been estimated. In the calculation, the total metal emissions were considered to be of two different types. First, a volatile fraction of small particles and then the emissions of particulate matter of larger size. Since the volatile emissions are difficult to control it is assumed that emission factors developed for modern copper smelters are applicable and provide a conservative estimate of the emissions from the Åtvidaberg copperworks. The elemental composition of the particulate emissions is not known and it is assumed that the concentrations roughly follow that of the ore.
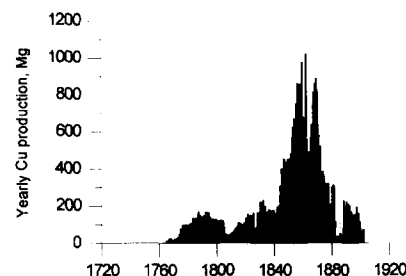

Fig. 2. Copper production at Åtvidaberg copperworks from 1750 to 1900, from Tegengren (1924).

There is some support for the idea that the particulate matter emissions decreased after 1844–1848, when the old furnaces were replaced and higher stacks were built, presented in the contemporary book written by the person who carried out the changes in the smelting processes (Bredberg, 1849). Probably, these estimates are conservative, at least, compared to data from Peters (1907) who described losses of up to 19% of the ore as flue-dust. However, the ore processed in Åtvidaberg was in much larger pieces while roasted than in the example mentioned by Peters.

An alternative approach for estimating the emissions into the air is to make use of contemporary studies of the losses in the process. It is also possible to quantify the share lost into the air and the share lost to the slag. The study on losses (Bredberg, 1825–1828) identifies steps where losses occurred and accordingly, we have estimated the extent to which losses to the slag and into the air would have occurred. Furthermore, there are some data on the Cu concentrations of the slag (Bredberg, 1869) which were used in the calculation of the losses.

3.2. Sampling and analysis

Top soil samples (0–10 cm), approximately corresponding to the A-horizon, and soil from the B-horizon (10–20 cm) were collected with a steel soil auger at twelve different sites in the vicinity of the copperworks. At each site, eight subsamples were pooled to form a general sample to represent the local variation of the sampling site.

The soil fraction < 2 mm of the samples was dried, extracted with nitric acid (conc.) in digestion tubes and analyzed for Cd by graphite furnace AAS,

Cu and Zn by flame AAS. This extraction procedure produce only approximately the total metal content of the samples. Acid strengths of standards were matched with the samples. Soil pH was determined on fresh soil samples after extraction in 0.2 M potassium chloride. Loss on ignition was calculated after heating the dried soil samples at 550°C for two hours.

The sediment data used in this article was produced by Qvarfordt (1977). The sampling was carried out in Lake Håcklasjön which is downstream of Åtvidaberg, with a hand-operated gravitational corer producing a one metre core, one of them near the inlet of the lake and one near its outlet. Metal contents were determined by AAS after acid digestion. The cores were not dated but approximate datings can be made (see Section 4).

In order to get an idea of the present addition to Lake Håcklasjön from the surrounding ground, five water samples from the inflow to Lake Håcklasjön were collected during 1989, both during high and low flow and analyzed by AAS.

4. Results and discussion

4.1. Emissions

The volatile emissions, including cadmium, are difficult to control and therefore, they probably roughly followed the production of copper, for the whole period of activity. Particulate emissions probably decreased after the changes in management and technology during the 1840s. There is support for these ideas both in literature and from the tree-ring

study carried out in Åtvidaberg (Eklund et al., 1997). For the emissions of Cd into the air the volatile fraction is the main contributer while for Cu and Zn the particulate matter fraction completely dominates the emissions (Table 1). These estimates are uncertain but probably conservative, considering the higher estimate where contemporary studies of copper losses formed the basis for the calculations.

From the contemporary studies of losses by Bredberg it can be concluded that during the period from 1750 to 1844 the total losses of Cu were probably about 25% of the total amount of Cu in the ore (Bredberg, 1869). Copper was lost either to the slag or as emissions into the air and these fractions had about the same size (Bredberg, 1825–1828). A quantitative estimate thus results in Cu emissions into the air of about 1400 Mg during this period. For the period from 1844 to 1900 the emissions into the air, relative to the production as well as in absolute values, were much lower. Tree-ring data (from Eklund et al., 1997) indicate that the atmospheric concentrations of Cu, in absolute values, were lower during the production peak of the 1860s than during the early days of production. It is therefore concluded that the emission factor for Cu emissions into the air during the later period was only 10% of the earlier value. Assuming this, another 300 Mg Cu would have been emitted into the air, making the total Cu air emissions to 1700 Mg.

These two different approaches did thus not deliver entirely consistent results, but are nevertheless not too different from each other, 1000–1200 Mg Cu for the modern emission factor approach and 1700 Mg Cu when using the study of losses in the process.

Table 1
Total emissions into the atmosphere of Cd, Cu and Zn from Åtvidberg copperworks, estimated by means of modern emission factors

Element	Emission factors (g/t Cu)	Volatile emissions (Mg)	Conc. in the ore (%)	Particulate emission (Mg)	Volatile + particulate emission (Mg)
Cadmium	350–650 [a]	12–22	0.03	7	19–29
Copper	2500–5000 [b]	85–170	4	900	985–1070
Zinc	9000–11 000 [c]	310–390	10	2300	2610–2690

[a] GCA Corp. (1981).
[b] Ayres and Ayres (1994).
[c] Nriagu and Davidson (1980).

4.2. Soil

Eight of the sampled soils were cambisols, i.e. brown soils, only one typical podzol and three intermediate. Soil pH-values (KCl) ranged from 2.93 (the podzol) to 5.31 and the content of organic matter was below 21%, with one exception, the podzol. The metal contents were higher than average values for Sweden but still within the range found at other localities, especially for cadmium (Table 2). Compared to other soil studies around modern copper smelters and other metal polluters (Hutchinson, 1979; Tyler, 1984), the concentrations are low. This fact could be interpreted either as a result of leaching which has transported the metals further down the soil profile or that the deposition of metals in the surroundings of the works was lower than expected or a combination of both. If soil leaching factors from the literature for Cd, Cu and Zn are applied in this situation, it seems unlikely that a substantial amount of Cu and Zn would have leached through the upper 20 cm:s of soil while, on the other hand, this probably has occurred with Cd.

The metal contents are thus not very high (up to 210 mg/kg for Cu or to 230 mg/kg for Zn) but there is another striking feature of the soil metal data in that the vertical distribution show the B-horizons to contain a large proportion of the metals, despite their lower content of organic matter (Table 2). On average, about 60% of the Cd was found in the B-horizon (10–20 cm). For Cu and Zn the share of the B-horizon was about 40%. The soil metal data

Table 2
Soil characteristics of samples collected around Åtvidaberg copperworks, Sweden

| Direction, number	Soil type	Distance (m)	Horizon	pH (KCl)	Loss on ignition (%)	Cadmium (mg/kg)	Copper (mg/kg)	Zinc (mg/kg)
E, 1	Cambisol	170	A	3.70	7.4	0.68	130	200
			B	3.78	3.5	0.60	210	230
E, 2	C	400	A	3.70	8.9	0.35	150	210
			B	3.62	4.7	0.55	72	82
E, 3	Podzol	880	A	2.93	35	0.66	110	140
			B	3.31	11	0.40	52	33
NE, 4	C	260	A	4.38	7.5	0.60	47	130
			B	3.80	3.6	0.50	28	92
NE, 5	C	480	A	4.20	8.6	0.59	76	210
			B	3.67	5.3	0.80	44	120
NE, 6	P/C	690	A	3.48	21	0.43	90	170
			B	3.81	6.5	0.42	63	130
N. 7	C	240	A	4.12	8.9	0.10	97	170
			B	4.23	4.3	0.70	110	160
N. 8	C	400	A	4.49	12	0.83	140	310
			B	4.22	9.2	1.05	180	230
N. 9	P/C	700	A	3.54	17	0.24	84	170
			B	3.81	8.1	0.55	64	110
NW. 10	C	500	A	5.31	8	0.35	92	270
			B	4.56	5	0.80	100	230
NW, 11	C/P	810	A	3.61	9.8	0.85	160	220
			B	3.86	4.2	1.60	130	180
NW, 12	C	1070	A	4.08	10	0.22	97	140
			B	3.89	6.1	0.73	77	110
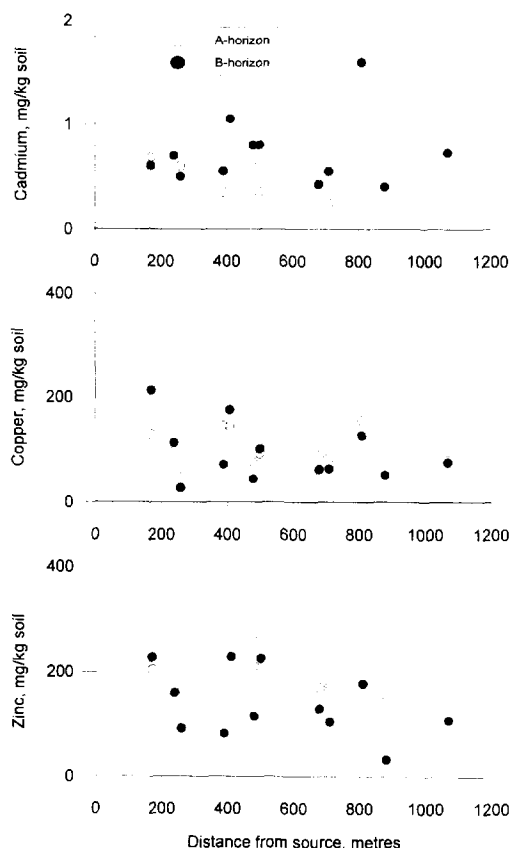
Fig. 3. Soil Cd, Cu and Zn at different distances from the source in A- and B-horizons around Åtvidaberg copperworks, Sweden.

are presented according to their distance from the sampling site to the source in Fig. 3. For all three metals the highest soil contents were found at 300 to 500 metres distance from the works and not in the most closely situated area, which is a common phenomenon. The lowest metal contents were found in the sites furthest from the works (500 to 1100 m), but there is possibly a difference between the vertical distribution of the different metals. On the sites more than 500 metres away from the works there were lower contents of both Cu and Zn in the B-horizon of the soil. Only about 35% of the Cu and Zn is found in the B-horizon while about 60% of the Cd is in the B-horizon. The relatively lower proportion of Cu and Zn in the sites farthest away could be explained by the metals there being deposited later. The dispersal of metals from the source probably changed over time, since higher stacks were built during the later part of the activity period.

An equation assuming exponential distribution with a peak at 400 metres distance was fitted to the soil data in order to assess the deposition pattern of the metals. The calculation resulted in an estimate where 20% of the Cd was expected to have been deposited within 1200 metres. For Cu the share was about 40% and for Zn about 50%. The deposition pattern assessed from the formation of exponential equations can be compared to the share found in the soil of the emissions to air presented in Table 3. For Cd, 15–20% of the estimated total amount emitted into the air was found in the soils up to 1200 m away. For Cu the result was 55–65% and for Zn 30–35%.

On the basis of the metal distribution between the A- and B-horizons and the knowledge of when the emissions occurred, it is possible to calculate the leaching rates of the metals. The method contains the assumptions that the leaching is constant, proportional to the amount of metal in the soil and that no leaching to the C-horizon has occurred. Thus, if substantial amounts of metals have leached to the C-horizon, our calculation will underestimate the leaching rate. The risk for this error is especially great for Cd. When calculating the leaching rates, it was assumed that all deposition of Cd took place during the median year, 1855. For deposition of Cu and Zn the median year was 1840. The following equation describes how the calculation was made:

$$x^{\text{yrs}} \cdot T = A \qquad (1)$$

where x = remaining share of the A-horizon after one yr, T = total elevation above background level, A = present elevation in the A-horizon, and $(1 - x) \cdot 100$ gives the leaching rate in % yr^{-1}.

The calculated average leaching rate for Cd was 0.8% yr^{-1} and for Cu and Zn 0.4% yr^{-1}. The rate

Table 3
The metal flow induced by the Åtvidaberg copperworks, Sweden. Soil and sediment content exclude estimated natural occurrence of the metals. All data are in Mg

Metal pools	Cd	Cu	Zn
Total amount, ore	275	40000	90000
Emissions to the air	20–30	1000–1200	2600–2700
Total amount in soil < 1200 m	3.2	650	880
Total amount in sediments	5.5	200	1100

for Cd is in the same order as in other studies with different approaches, like 0.8% yr^{-1} from Bergbäck and Carlsson (1995) and 0.52% yr^{-1} from a laboratory study by Tyler (1978). From mining polluted soils relative retention factors have been assessed as 0.01 to 0.17 for Cd, 0.11 to 0.19 for Zn and 0.32 to 0.63 for Cu (Merrington and Alloway, 1994). It would thus be expected that the leaching rate of Zn would be similar to Cd rather than to Cu but in this case Zn is fairly immobile. To find the explanation for this it is necessary to consider the nature of the copperworks' emissions which were dominated by large particles, flue-dust, spread in a limited area. These particles contained sulphides of Cu and Zn (Percy, 1861) and when deposited on the soil their leaching characteristics are not comparable with ionic water-soluted metals. Before any leaching of Cu and Zn can occur, these particles must weather and probably this is the reason to why Cu and Zn display similar leaching characteristics. There is some additional support for this idea: there is a fairly good correlation between the quotients A-horizon Cu/B-horizon Cu and the A-Zn/B-Zn while there is no correlation between Cd and any of Cu and Zn.

It is very difficult to assess whether leaching from the B-horizon of the soil has occurred but, if the calculated leaching factors for A- to B-horizon-leaching are applied to leaching from the B- to the C-horizon it is obvious that no substantial amounts (10% or less) of Cu and Zn have been transported down to the C-horizon. This calculation, however, results in a loss of about 30% of the Cd to the C-horizon. It has been shown (Bergkvist, 1987) that leaching of Cd and Zn increase with increasing soil depth and thus, there is a great uncertainty in these calculations.

When converting the soil contents to an estimation of the total amount of each metal originating from the copper production, assumptions must be made for the background metal content. This assumed background metal content has been subtracted from the analyzed soil metal contents and the amount of metals deposited have been calculated. The background data used here were 0.2 mg/kg soil in the upper soil layer and 0.1 mg/kg soil for the 10–20 cm layer for Cd. For Cu, 15 mg/kg soil was used for both horizons. The background Zn content was assumed to be 60 mg/kg soil (0–10 cm) and 30

mg/kg soil (10–20 cm). These assumptions were based on general data from Ek (1987) and local-specific data from surrounding areas (Eklund et al., 1997).

The amount of Cd, originating from the copper production, in the upper 20 cm soil of the surrounding area is thus estimated as 3.2 Mg. The corresponding calculations for the other metals result in estimations of 650 Mg for Cu and 880 Mg for Zn.

4.3. Sediments

Here, only the data from the inlet to the lake is presented (Fig. 4) while the results from two sites have been used when estimating the total amount of the metals in the lake sediment (Table 3). No dating of the sampled sediment cores was made but a few events can be identified by their chemical chronol-
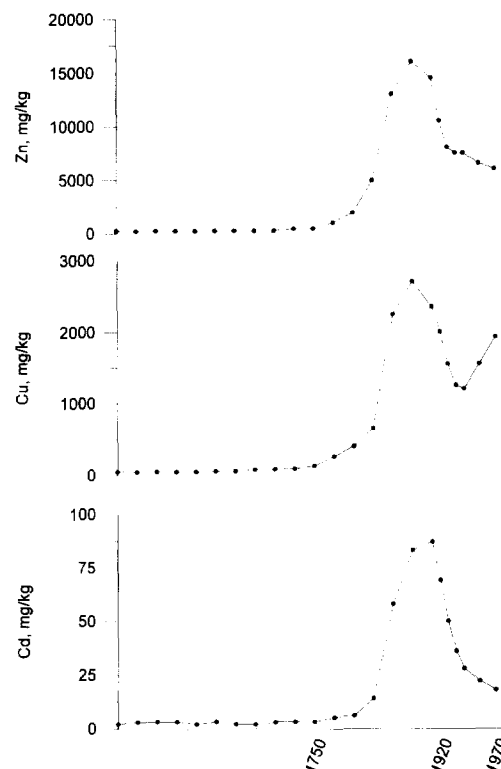

Fig. 4. Cadmium, Cu and Zn (mg/kg) in sediment from Lake Håcklasjön, downstream from the Åtvidaberg copperworks, Sweden. The dating is approximate and based on elevated contents of metals (1750) and nitrogen and phosphorous (1920, installation of water closets).

ogy. There is a distinct increase of the Cu and Zn content at the 50 cm level. Since the copper production discharged emissions directly into the water upstream of the lake it is reasonable to assume that this level of the sediment corresponds approximately with 1750 when the production of copper commenced in Åtvidaberg. The elevation of the Cd content of the core occurs later than for Cu and Zn. This could indicate that the Cd from the copper production mainly reached the lake through emissions into the air and deposition onto the lake surface and the surrounding ground. This idea is further supported by the fact that the Cd sediment chronology peaks later than the Cu and Zn ones.

The decrease in metal content of younger sediments is probably caused by the cessation of emissions directly into the water from the copperworks. After that, the metal contents have not dropped to the levels existing before the copperworks' activity which could be interpreted either that a resuspension of the sediment has occured or that further metals from the surrounding soils are continually loading the lake. There is some support for the latter assumption from water analysis data which gave values ranging from 0.7 to 1.0 μg/l for Cd, 13-34 μg/l for Cu and 196-360 μg/l for Zn, all at least 15 times above background levels.

The concentrations of Cd, Cu and Zn in the water and sediments are very high compared to those of the soil. It is thus likely that there are other sources of metals than the investigated soils. Within the area of the copperworks huge amounts of slag have been deposited. The thickness of the slag layer is in some parts more than 10 metres and the slag contains high concentrations of Cu (median 2800 mg/kg, $n = 15$) and Zn (median 3400 mg/kg, $n = 15$), but not Cd (median 1.5 mg/kg, $n = 15$), according to unpublished report by Ragnarsson (1996). This provides a potential for a significant contribution to the metal load of the lake.

The latest development of the sediment chronologies could be interpreted as a result of the different leaching characteristics of the metals where the most mobile element, Cd, displays the largest decrease and the least mobile, Cu, even an increase. Zn is intermediate with a moderate decrease of the content in the younger sediments. An alternative explanation could be that the elements display different upward

mobility. However, if the former interpretation is correct, the conclusion must be that the store of easily mobile Cd in the soil is becoming exhausted and that further leaching to the lake will continue to decrease. In contrast, the Cu store seems by no means exhausted and can be expected to continue as a source to the lake.

5. Conclusion

Cadmium was the most mobile of the metals in the soil study and the data from the sediment support this finding since a large proportion of Cd is found in the sediment sink. From the soil analysis we conclude that both Cu and Zn are much less mobile and display about the same leaching rate in the soil. This finding is somewhat contradicted by the sediment data, where the amount of Cu is much less than Zn. In the copper production process the Zn was enriched in the slag and later deposited around the copperworks. Since there is a high proportion of Zn in the slag, the water leaching from the deposited slag piles is bound to contain large quantities of Zn. This could be the reason why the sediments contain much Zn.

It seems as if the soils surrounding the copperworks, a century after the works closed down, continue to load Lake Håcklasjön with Cd, Cu and Zn. For Cd, it is likely that these approximately 100 years of leaching have substantially reduced the store of the soils while for Cu and Zn we are still at the beginning of the loading pulse to the lake.

Acknowledgements

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