The influence of mining on Lake Tisken and Lake Runn

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An investigation of nine sediment cores from Lake Tisken and Lake Runn provided a basis for establishing the metal distribution in lake sediments downstream of Falun Copper Mine. The study was restricted to the distribution of iron, manganese, copper, zinc, lead, silver, and cadmium and their association with the mining activities in the area. In some of the cores, cobalt, chromium, molybdenum, antimony, selenium, and mercury were also determined together with loss on ignition, water content, wet density, and pH of the sediments.

The concentrations of almost all metals increase in the sediments as a result of the mining operations. As regards copper and zinc the increase is 25 to 100 times the average for the background concentration. By dating one sediment core and comparing with the metal distribution is was possible to calculate that the mining in the area began c. 700 A.D. The recovery of the big cave-in, called the "Great Pit", in 1687 can also be traced in the sediment cores.

Of particular interest is a calculation of the metal content in the sediments which shows that there were probably some unknown metal sources in the area which, in the last 300 years, have been responsible for about 40 to 50 per cent of the metal input to the lake. This figure is in agreement in the main with the calculated amounts of the metals lost through weathering from the mining area during the last 300 years.

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There are mines where silver is dug;

There are places where gold is refined. Men dig iron out of the ground And melt copper out of the stones. Men explore the deepest darkness. They search the depths of the earth And dig for rocks in the darkness. Far from where anyone lives Or human feet ever travel, Men dig the shafts of mines. There they work in loneliness, Clinging to ropes in the pits.

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Introduction

In recent years, discoveries of high concentrations of heavy metals in lake sediments have aroused serious concern. Moreover it has long been known that certain heavy metals such as copper, cadmium, mercury, and lead are highly toxic even in small concentrations. In an attempt to discover both the natural loading and the contamination of heavy metals emanating from some pollutant in a certain environment lake sediment cores are often taken.

There is an intimate relationship between lake type and lake sediment, and the physical and chemical characteristics of bottom deposits closely reflect both natural processes and human activities operating within the catchment area. The use of lake sediment surveys has also been illustrated in studies of lakes suspected of having been polluted by cultural loading by man (cf., e.g., Digerfeldt et al. 1975; Salminen, 1976; Cato, 1977; Håkansson, 1977, 1981, and Qvarfort, 1977). In most of these studies is was found that the uppermost 5-10 cm of the cores usually show the highest concentration of metals. It was therefore thought that the concentration of heavy metals was, in some cases, due to recent cultural loading by man. Furthermore the accumulation of metals in the sediment takes place by either adsorption, organic complexing, sulphide preciptation or, to a lesser extent, enrichment in ferromanganese nodules.

Walters *et al.* (1974) found that mercury diffuses upward through the sediment, resulting in high concentrations in the sediment/water interface. Therefore, an upward accumulation of metals may show only that geochemical and biological reactions within the sediment are efficient in keeping the heavy metals concentrated in the upper few centimeters of sediments, even under continuous sedimentation.

The vertical distribution of some metals in a given sediment core depends also on the compaction, the diagenetic processes, the bioturbation and the diffusion in the sediment and the sediment-water interface. A metal enrichment of about 10 per cent at the sediment-water interface, relative to the background value, can be attributed to sediment diagenesis and is not necessarily an anthropogenic input (cf. Elderfield & Hepworth, 1975).

The primary aim of this study is to report the distribution of various metals in lake-sediment cores, and their correlation with the mining activities in the drainage area of the Falun Copper Mine. The working hypothesis is that the element contents in the lake sediments are closely related to the mining activities. Consequently, provided that proportion of metals in the drainage water to the lake during the mining period was several times that of earlier periods, we ought to find an unusually high metal content in the "mining part" of the sediment. This, however, presupposes a relatively constant sedimentation rate.

Some of the results in this paper have been published earlier, in Påhlsson & Qvarfort (1975; cores nos. 1-6), Qvarfort (1980; core no. 8) and in Gembert (1982; core no. 7).

Heavy metal contamination in a mining area

In recent years there has been a growing interest in the environmental effects of pollutants in the surroundings of sulphide mines, particularly in the Bergslagen region, southern central Sweden. In these areas, thousands of small mines have been worked and economically important amounts of metals recovered by a number of smelting furnaces.

When the smelters were operating, the ground in their neighbourhood must have been contaminated by metals lost as "fume". In addition, piles of slag accumulated around most of these sites, and although the majority were not readily decomposed by weathering, and elements occurring in sulphides were likely to become mobilized. The fine components of slag-heaps near rivers would also be particularly prone to transport by run-off, and thereby reach the drainage system.

Since acid mine-drainage, caused by oxidation of pyritic material, can continue long after the closure of a mine, there is an increasing concern about the environmental effects of disused mines (Qvarfort, 1979).

The degree of contamination (of the environ-

ment) due to the mining and allied activities will vary with the emitted pollutants and also to some degree with the site of the emittor. This transport can be rather complicated as all media, soil, groundwater, streams, and lakes, are involved. This first question is how the atmospheric deposition will be distributed. The second, when and how the metals deposited on land will reach the lakes and be incorporated in the lake sediments.

Previous mining and smelting in the Falun region

The previous mining activities in the Falun region and its industrial history were described by Tunberg (1922), Tegengren (1924), Söderberg (1932), Lindroth (1955), Lundqvist (1963), Boëthius (1965), Hyenstrand (1979), and Qvarfort (1980).

No one can tell exactly when and how the Falun Copper Mine began to be worked. The mine is, however, one of the oldest in the world, where work has been in progress at various times from about 700 A.D. (late Iron Age) until the present day.

A brief description follows of a few important events in the history of the mine.

Year A.D. Remarks

- 700- Investigations carried out by various methods
- 1080 such as pollen analysis, sediment analysis and radioactive carbon dating, indicate that mining operations started sometime towards the end of the Viking Age.
- 1288 A document which shows that mining was in progress, organized as a kind of company in which shares could be bought and sold.
- 1347 King Magnus Eriksson issued a charter for the mine-owners. The charter provides detailed specifications concerning the rights to work the mine. This document, long a standard for conditions at the mine, was actually used until the 16th century.
- 1492 The customs records of the Hanseatic town of Lübeck, which have been preserved since 1492, show that the annual production at the mine reached more than 300 tons of copper in this period.
- 1504 Repeated roof-falls began to disturb operations. Up to the 1550's the production was only about 70 tons a year.

- 1556 During King Gustav Vasa's reign, important technical innovations were introduced. Water was required for working the pump system which drained the mine and was obtained by means of extensive regulation of lakes adjacent to the mine. Canals were also dug and wooden pipes were laid. Owing to technical improvements and the fact that richer ore was reached, the production could gradually be increased.
- 1629 Olof Swart's mine-map of the Falun Copper Mine was finished in this year. The map is the oldest mine-map in Sweden.
- 1650 More than 3000 tons of crude copper were produced. The copper from Falun met two thirds of European requirements at this time. The ore was treated in smelters in the immediate vicinity of the mine. During this time, the mine was sunk deeper, and the work was concentrated to more intensive mining of medium-grade ore.
- 1687 "Stora Stöten" or the "Great Pit" resulted from the major collapse at Midsummer this year. The rock wall between the two major shafts of the mine, and the galleries beneath, caved in. Current mining technique was the cause of the collapses as fire-setting was used to open large galleries beneath each other.
- 1700 Christopher Polhem was engaged in the technical administration of the mine. He designed special devices (Swedish "stånggångar") for transmitting power to hoisting devices and pumps.
- 1714 The mine began its renaissance period under the mining inspector Anders Swab. More business like organizational and working conditions were introduced.
- 1750 In 1750, records mention that red paint was burnt from vitriol. This by-product derived from the manufacture of sulphur shows an annual production of 50 to 100 tons from the 1780' to the 1830's. Nowadays the production is about 200 tons a year.
- 1881 The copper production ran at a considerable loss. Instead, gold and silver were worked. In 1888, 81 kg of gold were extracted. An average of 400 kg of silver a year was also obtained. Figure 1 presents the crude copper production during the years 1620 to 1898.
- 1983 At present, the total production of the mine is about 200 000 tons of ore per year. The average composition of the ore mined is: 21% iron, 3% zinc, 1% lead och 0.5% copper. Earlier, however, the percentage of copper was higher. During the 18th century for example it was about 7%.



Fig. 1. Annual crude copper production from the 1620's to the 1890's (Redrawn from Tegengren 1924).

Area of investigation

Lake Runn and Lake Tisken

These two lakes are situated to the south of the town of Falun (see Fig. 2). Lake Tisken was originally a creek of Lake Runn. During the 16th century, however, it was dammed about one metre above Lake Runn. More data about the lakes are given in Table 1.

The evaluation of the lakes has a direct relation to the mining in the area. The main part of the old town of Falun is built on dump-heaps. Leached material from these, together with the pump water from the mine, have long been added to the water and sediments in the lakes. Furthermore, air-borne metals from the melting works in the surroundings of the lakes may also have contributed. The population of the town of Falun has also contributed to the pollution of the lakes. As a result of all these factors the water and sediments in Lake Runn and Lake Tisken are severely polluted. Today the proportions of some metals discharged into the Lake Runn are Fe 112, Zn 70, Cu 2.8 kg h^{-1} and Cd 65 g h^{-1} (Dottne-Lindgren, 1977).

Table 1. Morphometric data for Lake Runn and Lake Tisken

Lake Runn	about 3005 km ²
Catchment area:	about 57 km ²
Lake Area:	maximum 26 m, mean 8 m
Water depth:	5-6 months
Resetting time:	from Lake Tisken and from the Sund-
Inflow:	bornsån River
Outflow:	to the Lillälven and Dalälven Rivers in
Lake Tisken Lake area: Water depth: Inflow: Outflow:	0.5 km^2 maximum 2 m, mean 1–1.5 m from the Faluån River in the north to Lake Runn in the south.



DEPTH BELOW WATER LEVEL

Fig. 2. Map showing the investigated area and the sampling sites in the lakes.



Fig. 3. Simplified geological map of the investigated area.

The bedrock of the area is dominated by granites and leptites of Precambrian age. The chief ore mineral nowadays is pyrite, together with some chalcopyrite, sphalerite, and galena. The pyrite ore forms compact, lens-shaped masses and has to a large extent replaced limestone or dolomite which are now found as scattered remnants in the ore body. Some sulphide minerals also occur as impregnations in quartzite, occasionally with some gold and selenium. The rich copper ore which previously made Falun renowned is now of little importance.

The ore bodies are usually bordered by *sköl*zones which consist of biotite, chlorite, and talc with large crystals of chalcopyrite and other sulphide minerals.

The Quaternary deposits covering the bedrock are till, glaciofluvial deposits and fine-grained sediments. More geological information is given in Fig. 3.

Methods

Sampling

Sediment samples were collected over the period 1975–1981, with a Livingstone bore giving a core length of 1.5 m and a diameter of 54 mm. Sampling was performed to a depth greater than 1.5 m from the bottom of the lake sediment surface, with 20 cm overlap. To locate suitable sampling sites in the lakes, manual soundings were made. The use of the Livingstone borer has the disadvantage that some systematic sampling errors, due to loss of part of the sediment-water interface, can occur. This fact must be borne in mind when interpreting the analytical data.

Before analysis, the core was cut lengthwise; samples with a thickness of 5 cm were used. In most cases, the core was analysed a few days after sampling. The samples were, however, always stored at $+6^{\circ}$ C before analysis.

In connection with investigations in the area, Lundqvist (1963) collected some samples from Lake Tisken for pollen analysis in the year 1934, using a Hiller auger. The samples were taken every 2.5 cm and stored at the Geological Survey of Sweden.

Parts of Lundqvist's samples have been used for the chemical analysis. It was thereby possible to use the stratigraphical section constructed by Lundqvist (1963). However, in view of the lapse of time since the sampling, determinations of density, water content, loss on ignition, and pH were not made. Lundqvist's core is here named no. 8.

For one of the cores, no. 7, the results come from an investigation made by Gembert (1982). Some samples from cores nos. 1-6 have been reanalysed.

Chemical analysis

The laboratory methods, some of them routine procedures, were chosen as being most suitable for giving information about the lake sediments.

Wet density. – The wet density of the lake-sediment samples was determined with a nutting pycnometer (Krumbein & Pettijohn, 1983).

Water content. – Between 5 and 10 g of the fresh sample were weighed, dried at 110° C for 24 h and re-weighed. The water content was expressed as the weight loss from 100 g of wet sample.

Organic content. – About 10 g of the dry material (100°C) were ignited at 450°C for 12 h in a furnace. The organic content, approximately equal to the loss on ignition, was expressed as the weight loss from 100 g of dried sediment.

pH. – The measurement was made by inserting the electrode directly into the freshly opened sample.

Element analysis. – For the determinations of the trace elements, except those of the more stable, rock-forming silicates, the samples (1 g of each) were digested with a mixture of hydrochloric and nitric acid (1:3, 50 ml) by heat attraction (90°C) for one hour. After dilution of the samples to 100 ml the elements were determined by atomic absorption using a Varian 1200 instrument. The mercury analysis was made by flameless atomic absorption (Per-kin-Elmer, Coleman Mercury Analyser).

Presentation of analytical data

The results of the chemical analysis, calculated on the basis of dry weight, were plotted against depth in a so-called silhouette diagram. The stratigraphy, depth below surface, and some dating notes are arranged in separate columns. Finally, the data obtained were transcribed onto tapes for ultimate storage. Analytical data lists can be obtained from the author.

Dating the sediment profiles

The sediment profile from Lake Tisken was dated earlier by Lundqvist (1963). He used a combination of pollen analysis, ¹⁴C dating, and traces of the increasing influence of the cultivation of the landscape. The results are entered in Fig. 11. This standard diagram from Lake Tisken was also used together with the chemical analyses for interpretation of the historical evolution and the supplementary dating of, and correlation between the different cores (see p. 126).

Results and discussion

The sampling sites are shown in Fig. 2 and the results of the sediment analysis in Fig. 4 to 12. To facilitate the scrutiny of the analyses and the comparison of the values from the lakes the investigated area was divided into four smaller ones. The grouping can be seen in Fig. 13.

With regard to the stratigraphical description, the organic and the water content, the results are summarized in a table, containing the main data for the different sediment layers. The stratigraphical differences between the cores can be seen in detail in the diagrams, Figs. 4 to 12.





Fig. 4. Graphical presentation of the analytical results from core 1.

Core r Water	io. 2 depi	th 2.5m																
tratigraphy	amp depth cm	rg. content %	ater content %	ensity g∕cm³	τ	e mg∕g	b∕br u	n µg∕g	б/бп и	¢∕6r q	6/6rt 6	r µg∕g	i µg⁄g	6/6rt u	6∕6rt o	б/бп о	б/бп [,] р	6∕6u 6
s	S	25	50	1	4	100		2500	2500	600	25	750	< 40	250	125	25	2.5	- E 1250
<u>Ochre</u> Black Gyttja	50 - 50 -																	

Fig. 5. Graphical presentation of the analytical results from core 2.

Core no. 3 Water depth 3,5m



Fig. 6. Graphical presentation of the analytical results from core 3.



Fig. 7. Graphical presentation of the analytical results from core 4.

Core no.5 Water depth 5.0 m



Fig. 8. Graphical presentation of the analytical results from core 5.



Fig. 9. Graphical presentation of the analytical results from core 6.





Fig. 10. Graphical presentation of the analytical results from core 7.

Stratigraphy

In Lake Runn and Lake Tisken the stratigraphy changes, as shown in Table 2, from clay beneath, to

Table 2. Stratigraphical description and organic and water content of the five distinct sediment layers.

I	Description	
Layer no.	Description	
5	Iron ochre.	Organic content 20–25%
		Water " 80-85%
4	Black gyttja,	Changing downward to
	0.5	stratified clay-gyttja.
		Organic content 15-25%
		Water " 80-85%
3	Clay gyttja.	Organic content 5-15%
		Water " 50-60%
2	Silty layer.	Organic content c 5%
	, ,	Water " c 5%
1	Clay.	Organic content $2-3\%$
	,	Water " 30-50%

a more or less black gyttja-clay or clay-gyttja in the uppermost part. In some of the cores, e.g. nos. 1 and 8, iron ochre forms the uppermost layer. It has been assumed that this layer indicates an increasing pollution of the lakes. According to the cores from Lake Tisken, however, the ochre layer may have some connection with the copper-extraction plant constructed beside the Faluån river in 1862 (Lundqvist 1963). Stapf (1865) – only three years later – described an ochre layer in Lake Tisken which suggests that the deposition of ochre started as early as the close of the 17th century, before the extraction plant (cf. Lundqvist, 1963).

The layers of the cores show variations in thickness. This information is important for giving an idea of the relative sedimentation rate in a given lake area. The quantity of the bottom sediment may vary within the same lake depending on several factors such as allochthonous influence, water circulation, autochthonous production, resuspension etc.

A continuous sequence of sediment can only





Fig. 11. Graphical presentation of the analytical results from core 8.

appear within zones of accumulation, although it may be influenced by e.g. bioturbation and compaction. In some bottom areas, as near a river-mouth, the rate of deposition can, however, be high. Håkansson (1981) demonstrated that the areal distribution of erosion- and transportation bottoms on the one hand and accumulation bottoms on the other can be determined from only three parameters, viz. lake area, mean depth and maximum depth. By means of this model is has been calculated that about 27 per cent of the bottom area of Lake Runn is dominated by erosional and transportational processes and about 73 per cent by accumulational processes. The corresponding figures for Lake Tisken are 26 and 74 per cent, respectively.

In view of the above-mentioned facts it is believed that e.g. the cores nos. 3, 7, 8 and 9 show continuous sequences, and can be used as a direct record to diagnose the pollution and unravel the historical evolution of the Falun Copper Mine.

Organic and water content

From Table 2 and Figs. 4 to 12 it can be seen that the organic and the water contents show a similar vertical distribution in the cores, with increasing values from the lower to the upper parts.

A comparison between the different cores also shows that the northern section of Lake Runn has higher organic and water contents in the upper part of the sediment sequences. This relationship can be attributed in part to the increase of human influence on the landscape, or to the fact that the waste water from the town of Falun was previously discharged directly into lakes Tisken and Runn.

pH

The hydrogen-ion concentration in the cores presents only minor variations, with values around 7. The vertical distribution of the pH values almost



Fig. 12. Graphical presentation of the analytical results from core 9.

invariably follows a general trend with pH values rising as the redox potential decreases with increasing sediment depth.

Distribution of Fe, Mn, Cu, Zn, Pb, Ag, and Cd with depth

Nine cores, nos. 1 to 9, each about 1 to 3 m long, were analysed for some elements. The results, as functions of depth, are given in Fig. 4 to 12. They are presented on a dry-weight basis.

In some of the cores, nos. 1, 2, 3, and 6, diverse other elements, such as Co, Cr, Ni, Sn, Mo, Se, Sb, and Hg were also determined. Their distributions will be discussed below (see p. 127).

The general distribution of the metals with depth is almost the same in all the cores. The contents in the lower part of the profiles are low and fairly even. Higher up in the sediment, a sudden increase in the contents of metals is observed. This increase occurs at different levels depending on the location of the cores, or on the said fact that not all cores encompass continuous stratigraphical sequences.

With the exception of minor variations, the metal increase continues to the top of the cores. The increase can be attributed in some measure to the upwards increasing contents of organic matter. In the present case the upward increase of the metal contents must, however, mainly be an effect of the increasing mining activities in the area. It is, however, still an open question to what extent the changes of the metal contents with depth depend on the secondary cycling of the metals in the sediment.

The maps in Fig. 13 showing samplings sites, core length and thickness of the core-sections affected by mining were constructed to sum up the stratigraphy and the metal distribution in the cores.

As the cores show both the natural metal contents and those of the mining period it is also possible to distinguish the following parts in most sequences. In general, the distribution of the elements in the lower part of the cores is characteristic of glacial deposits in the catchment area. This part of the profiles displays the natural background level of the element in question in pre-mining deposits. Higher up in the sediment, a small increase suddenly occurs for almost all the elements investigated. It may be attributed to the first mining activities in the area.

The upper part of the cores contains the big increase of the metal contents. According to the facts mentioned above, these parts may reflect the more intensive mining period in the area. Moreover during this period the ore was processed in the immediate vicinity of the lakes.

When calculating the pre-mining metal contents of the sediments of the two lakes the clay part was excluded. The following mean values were obtained.



Fig. 13. Map showing the sampling sites, the stratigraphy and the thickness of the parts of the cores affected by mining.

Lake Tisken, area A: Fe 31 mg/g, Mn 0.7 mg/g, Cu 31 μ g/g, Zn 197 μ g/g, Pb 45 μ /g, Ag 2.2 μ g/g and Cd 1.5 μ g/g.

Lake Runn, mean of areas B–D: Fe 25 mg/g, Mn 1.1 mg/g, Cu 19 μ g/g, Zn 105 μ g/g, Pb 38 μ g/g, Ag 1.1 μ g/g and Cd 0.9 μ g/g.

There are some differences between the background values for the two lakes. The highest absolute metal contents are found in Lake Tisken, probably because this lake is located close to the mineralizations.

A similar calculation for the remainder of the sediment sequences, called the first mining part and the main mining part, gives the following values:

Lake Tisken (area A), the first mining part: Fe 98.89 mg/g, Mn 0.59 mg/g, Cu 1457 μ g/g, Zn 1892 μ g/g, Pb 248 μ g/g, Ag 8.8 μ g/g and Cd 1.5 μ g/g.

Lake Tisken (area A), the mining part: Fe 165 mg/g, Mn 0.34 mg/g, Cu 3689 μ g/g, Zn 4925 μ g/g, Pb 518 μ g/g, Ag 29 μ g/g and Cd 7 μ g/g.

Lake Runn, mean of areas B–D, the first mining part: Fe 29 mg/g, Mn 0.8 mg/g, Cu 75.6 μ g/g, Cd 163 μ g/g, Pb 57 μ g/g, Ag 4 μ g/g and Dd μ g/g.

Lake Runn, mean of areas B–D, the main mining part: Fe 77 mg/g, Mn 1.7 mg/g, Cu 1482 μ g/g, Zn 2079 μ g/g, Pb 380 μ g/g, Ag 11 μ g/g and Cd 2.3 μ g/g.

Is should be noted that the calculation also includes the iron-ochre layer, which can be found in the uppermost part of some of the cores (cf. p. 121). Within this layer, almost all elements show unusually high values, approximately twice the average for the main mining part.

From the above results it is possible to calculate the factor of enrichment for the respective metals. As mostly mean values are used, the method eliminates local variation. The factors calculated for the four subareas (cf. Fig. 14) are presented in Table 3.

Table 3. Enrichment factors of some selected metals in Lake Tisken and Lake Runn. The cores have been divided into two parts. No. 1 is the main mining part and no. 2 the first mining part. The different lake areas are shown in Fig. 13.

	Area A	2	Area l 1	B 2	1 1	Area C 2	1	Area D
	5 32	3 10	6.2	_	1.84	1 15	1 18	1 17
Mn	0.49	0.84	1.09	_	1.34	0.94	2.29	0.51
Cu	119	47	95	_	97	2.8	42	5.16
Zn	25	9.64	21	_	30	1.52	7.93	1.62
Pb	12	5.57	13	_	5.38	0.98	12	2.91
Ag	13	4.03	11	_	1.00	1.00	1.00	1.00
Cď	4.71	0.98	2.43	-	2.65	1.05	1.00	1.00
	a farri r	aluar	Cor colo	ulat	ion			

too few values for calculation.

The tables show that the contents of all metals, except manganese, increase in the "mining part" of the sediment. Within the "first mining part", the increase of the metal contents is relatively small. In the "main mining part", however, copper and zinc show unusually high values, approximately 25 to 100 times the average for the natural background. This phenomenon may have several causes, e.g. discharge to the lakes of waste material from the mining activities in the area. In addition, when the smelters and blast-furnace houses were operating, the supply of metals from the air must have been quite large.

Some enrichment factors are notable, viz. those of copper and zinc, which show the highest enrichment, those of lead and silver which are mediumenriched and that of cadmium, which has a relatively low factor. In comparison with values taken from the literature, the factors of the area are rather high (cf. Banat et al., 1972 and Förstner, 1976).

It must, however, be borne in mind that the enrichment factors of metals in the sediments depend on the total metal quantities discharged into the lakes. As shown above the contents of metals in the mine-water follow the order Fe > Zn > Cu > Cd, while in the sediments, the order of the enrichment factors is Cu > Zn > Fe > Cd. The total amount of metals in the sediment, however, follows the same order as that of the mine water.

The enrichment factors in the main mining part of the sediments are higher than those of the first mining part. This indicates that the metal effluents from the mine must have increased in the course of time.

The enrichment factors for those layers of the sediments which include the main mining part (No. 1) are illustrated in Fig. 14. They show that most of the metals accumulated in areas A and B, although the accumulation is high also in area C. This may indicate a supply of copper and probably some other metals such as zinc from the western part of Lake Runn. Despite these irregularities, however, copper and lead clearly tend to be more easily accumulated than e.g. zinc.

The change of the proportions of the metals in the sediment with the distance from the mining site indicates that some of the metals, such as copper and lead, will soon be accumulated and/or co-precipitated by various inorganic and organic precipitates and gels, which exist in the free state or coat the sediment particles. Nevertheless, it should also be noted that the increase of the contents of certain metal in the uppermost part of the sediment in Lake Tisken is partly due to the supply of waste water from the town of Falun.



Fig. 14. Enrichment factors of some selected metals in the lakes. MM represents the main mining part and Mb the background part (see text).

The correlation coefficients were calculated from the analyses of the sediments from Lake Runn. In order to facilitate a comparison between the coefficients the data from cores 3, 7, and 9 were used, split up into the three parts, the main mining, the first mining and the background. These cores show continuous stratigraphical sequences and were analysed for organic and water content, density, pH, and metal contents.

The correlation matrices are presented in Table 4. The results indicate that, in general, the elements Mn, Cu, Zn, Pb, Ag, and Cd show similar patterns in sediments, and high correlation coefficients. These elements may emanate from the same source. The relationship between the absolute amount dissolved and the absolute amount absorbed varies, however, from element to element.

As regards iron, however, the results reveal differences. In the mining parts of the sediment, iron shows a negative or a low correlation with the elements Mn, Cu, Pb, and Cd. In the background part, however, iron and these elements show a high or medium correlation such as Fe with Ag. As to iron, several mechanisms give rise to the sharp decrease in the correlation coefficients when the sediment changes from the pre-mining to the mining stage. The metals discharged from the catchment area are partly absorbed on suspended organic and inorganic particles or aggregates or form metallo-organic complexes. The sorption also involves adsorption on fresh hydrous oxides of iron and manganese. Concerning the mining part of the sediment, the metal adsorption, excluding silver, falls outside the relationship found in the lower (background) part. This divergence indicates a lesser adsorption between the metals and iron in the mining part of the sediment.

The lack of relationship, mentioned above, may be an effect of changes in the oxidizing-reducing parts of the lake during the mining period. Lee (1970) has shown that iron and manganese hydroxides are dissolved and migrate upward through the interstitial water in the reducing part of a sediment. But it is uncertain whether the other elements will also be mobilized or precipitated as sulphides within the sediment.

The results of this study are, however, confirmed

Table 4. Correlation coefficients for selected elements in the sediment cores, nos. 1-7 and 9. The cores are divided into three parts (see text).

4 Th					
*Part L.	(main	mining	period) n =	22
	(/	

	Org	Water.	Density	рH	Fe	Mn	Cu	Zn	Pb	Ag	Cd
	cont.%	cont.%	g/cm ³	r	mg/g	mg/g	µg/g	µg/g	µg/g	µg/g	µg/g
Org.cont.%	-										
Water.c.%	0.87	_									
Density g/cm ³	-0.78	-0.87	-								
pH	-0.34	-0.54	0.53	_							
Fe mg/g	0.84	0.72	-0.52	-0.42	-						
Mn mg/g	-0.31	-0.06	0.24	0.03	-0.90	-					
Cu µg/g	0.61	0.72	-0.51	-0.44	-0.13	0.92	_				
Zn µg/g	0.36	0.50	-0.64	0.09	-0.10	0.94	0.94	_			
Pb µg/g	0.44	0.37	-0.25	-0.29	-0.14	0.97	0.90	0.96	-		
Ag µg∕g	0.53	0.49	-0.28	-0.33	0.94	0.53	0.83	0.95	0.96	-	
Cd µg/g	0.49	0.35	-0.21	-0.63	-0.40	0.70	0.68	0.77	0.76	0.73	_
* Part 2 (first min	ing period) r	n = 64									
	Org.	Water.	Density	pH	Fe	Mn	Cu	Zn	Pb	Ag	Cd
	cont.%	cont.%	g/cm ³	g/cm ³	mg/g	mg/g	µg/g	µg/g	µg/g	µg/g	μg/g
Org.cont.%	_										
Water.c.%	0.97	_									
Density g/cm ³	-0.90	-0.94	-								
pH	-0.73	-0.76	0.82	-							
Fe mg/g	0.79	0.73	0.35	-0.43	-						
Mn mg/g	0.94	0.88	-0.13	-0.43	-0.41	-					
Cu µg/g	0.75	0.75	0.13	-0.24	0.21	0.40	-				
Zn µg/g	0.38	0.38	-0.28	-0.51	0.32	0.73	0.87	-			
Pbµg/g	0.84	0.82	-0.50	-0.73	0.39	0.89	0.77	0.95			
Ag µg/g	0.72	0.66	-0.65	0.67	0.93	-0.88	0.93	0.89	0.73	-	
Cu µg/g	0.59	0.61	-0.11	-0.32	0.15	0.88	0.49	0.75	0.85	0.65	-
* Part 3 (backgro	und) n = 60										
	Org.	Water. I	Density	pН	Fe	Mn	Cu	Zn	Pb	Ag	Cd
	cont.%	cont.%	g/cm ³	mg/g	mg/g	µg/g	µg/g	µg/g	µg/g	µg/g	µg/g
Org.cont.%	-										
Water.c.%	0.62	0.01									
Density g/cm ³	-0.77	-0.84	-								
pH	0.16	0.17	-0.23	-							
Fe mg/g	0.35	0.49	0.50	-0.44							
Mn mg/g	-0.29	0.07	0.01	-0.55	-0.72	-					
Cu µg/g	0.64	0.31	0.23	-0.60	0.59	0.85	-				
Zn µg/g	0.59	0.27	-0.30	-0.36	0.62	0.92	0.96	-			
Pb µg/g	0.67	0.75	-0.77	-0.53	0.73	0.82	0.94	0.94	-		
Ag µg/g	0.54	0.61	-0.46	-0.31	0.94	0.21	0.96	0.85	0.81	-	
Cd µg/g	0.40	0.25	-0.15	-0.56	0.49	0.84	0.94	0.94	0.92	0.93	_

by studies of Kuijpers (1974) carried out in the western Baltic Sea. He found that the decomposition of organic matter at the sediment surface in accompanied by a relative enrichment of mercury, lead and zinc, but not iron. This element seems to be recycled by the decomposition processes. According to Kuijpers, the decrease of iron relative to the other elements is probably due to differences in the formation of metallo-organic complexes. Distribution of Co, Cr, Ni, Sn, Mo, Se, Sb, and Hg with depth

In some of the cores, e.g. nos. 1, 2, and 3, analyses of the sediments also include Co, Cr, Ni, Sn, Mo, Se, Sb, and Hg. The results are given in Figs. 4, 5 and 6.

Apart from Ni och Co all elements investigated show increasing contents in the upper, mining part of the sediment. This in turn suggests that the upward increase of the element must, as stated above, be an effect of the increased mining in the area.

Some of the enhanced Hg concentrations in the northern part of Lake Runn, e.g. in core no. 1, are no doubt caused by discharges from a pulp mill north of Lake Runn, and not by the mining activities in the area.

The relationship between the mining activities in the area and the metal distribution in the sediment

Sediment cores have been used as historical records of intensified metal pollution due to human activities around the world (cf. Cato, 1977). In some of these studies a comparison is often made between the lower, "background part" and the upper, "polluted part". In polluted areas high-grade enrichments of certain metals are often found in the uppermost part of the lake sediments.

As regards the historical record of Lake Tisken and Lake Runn, two levels in the cores are of special interest in the geochemical investigation. One refers to the beginning of the mining activities in the area, and the other to the big cave-in, when the "Great Pit" (see p. 113) was formed. The first level can be traced in, e.g., core no. 8 from Lake Tisken, as a sudden increase in the element contents about 200 cm from the sediment surface. It is, however, hard to find in the other cores. Depending on the small amount of metals liberated by the first mining activities, the contrast between the background and the pollution levels will, however, not be very large. Furthermore, most of the metals will also have been absorbed in the vicinity of the mine, e.g. in Lake Tisken. The beginning of the mining activities has been dated to c. 700 A.D. (see Fig. 11).

The more pronounced level of increased mining activities in the area is shown by the "big" metal increases which appear in almost all cores. In Lake Tisken, for instance, this level is found about 50 cm from the sediment surface. It has been dated to c. 1700 A.D. (see Fig. 11).

This level appears also in the cores from Lake Runn. It may be connected with the formation of the "Great Pit" in 1687.

Obviously, the nature and amount of contamination due to the mining and the distribution of metals to the lake depends on many factors, such as composition of ores, weathering, leaching etc.

When the "Great Pit" was formed in Falun the amount of leaching must have increased enormously. As the whole cave-in is composed of large blocks of rock, water and oxygen can readily penetrate it. Maximum weathering and oxidizing conditions and maximum removal of metals are therefore to be expected. It is also obvious that the cave-in constituted a very extensive underground drainage system for groundwater penetration.

Hallberg & Rickard (1973) calculated the metal amounts lost through weathering and leaching from the "Great Pit" and the Falun Copper Mine since 1687:

Fe	570 000 tons,
Cu	11 400 tons,
Zn	103 000 tons,
Ag	91 tons.

In the past the draining of the mine was extremely primitive and irregular. The groundwater table of the surrounding rocks must have ben very variable. At the present time, however, both the ore deposits and residual material from the cave-in are mined by underground methods. The leaching products from the cave-in area will therefore be mixed with the drainage-water from the mine. Some metal-rich groundwater may, however, enter the lakes directly.

The amounts of metals nowadays discharged to Lake Runn are given below. The measurements were made in the outflow from Lake Tisken to Lake Runn (Dottne-Lindgren, 1977, 1978).

Fe 981 tons/year, Cu 26 tons/year, Zn 650 tons/year, Cn 0.6 tons/year.

The mean contents of metals of the "mining part" of the sediment sequences, reduced by the background values, were used to calculate the amounts of metals which have been bound to the sediments of Lake Tisken and Lake Runn since 1687:

 Fe
 500 000 tons,

 Cu
 14 000 tons,

 Zn
 17 000 tons,

 Pb
 1 900 tons,

 Cd
 33 tons,

 Ag
 72 tons.

Assuming that the outflow of metals to the lakes has been constant since the formation of the "Great Pit", it is possible to arrive at a rough calculation of the outflow during the last 300 years:

Fe	294 000 tons,
Cu	7 800 tons,
Zn	195 000 tons,
Cd	180 tons.

Several mechanisms could give rise to the differences between the two tables. The distribution pattern of Fe, Cu, Zn, and Cd is obviously very similar to the order of adsorption of these elements in the sediments. Fe and Cu prove to be more completely bound to a sediment than Zn and Cd (see e.g. Förstner & Müller, 1974).

Apart from that, all elements will to some extent escape from the lake system by different processes (cf. Olausson, 1972). This leads to lower contents of a metal in the sediment than those calculated from lake-water analyses.

What causes the contents of Fe and Cu in the sediment to be higher than those calculated from the water analyses? One reasonable explanation suggests the outflow of metal-rich groundwater directly to the lakes from the mine, or from some other sources in the catchment area. These unknown sources must have some connection with the mining activity or with the formation of the "Great Pit" as the increase of the metal contents in the sediments occurs during the corresponding period.

The amounts of Fe, Cu and Ag found in the sediment are almost of the same order as those given by Hallberg & Rickard (1973) concerning metals lost through weathering since 1687. Nevertheless, it must be kept in mind that the calculation of both the sediment-metal content and the metal outflow, were simplified.

The amount of air-borne metals supplied to the lakes was not taken into account in the calculation. An attempt to use a peat profile from the area for such a study failed as the analyses showed that the bog area had been affected by water from Lake Runn.

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REFERENCES

- Banat, K., Förstner, U. & Muller, G. 1972: Schwermetalle in den Sedimenten des Rheins. Umschau Wiss. Technol. 726, 192–193.
- Boëthius, B. 1965: Kopparbergslagen fram till 1750-talets genombrott. Uppsala. 218 pp.
- Cato, I. 1977: Recent sedimentological and geochemical conditions and pollution problems in two marine areas in south-western Sweden. *Striae Vol. 6*, Uppsala. 158 pp.
- Digerfeldt, G. 1975: The Post-Glacial development of Ranviken bay in Lake Immeln. III. Palaeolimology. *Geol. Fören. Stockholm Förh.* 97, 13-28. Stockholm.
- Dottne-Lindgren, Å. 1977: Falu gruvas inverkan på sjön Runn (1). Limnol. Inst. Uppsala universitet. Uppsala.
- Dottne-Lindgren, Å. 1978: Falu gruvas inverkan på sjön Runn (2). Limnol. Inst. Uppsala universitet. Uppsala.
- Elderfield, H. & Hepworth, A. 1975: Diagenesis, metals and pollution estuaries. *Mar. Pollut. Bull.* 6, 85-87.
- Förstner, U. & Muller, G. 1974: Schwermetallanreicherungen in datierten Sedimentkernen aus dem Bodensee und aus dem Tegernsee. *Tschermarks Mineral. Petrogr. Mitt.* 21, 145–163.
- Förstner, U & Wittmann, G.T.W. 1976: Metal accumulations in acidic waters from gold mines in South Africa. *Geoforum* 7, 41–49.
- Gembert, B. 1982: En sedimentundersökning i sjön Runn. Forskningsrapp. No. 91. Kvartärgeol. avd. Uppsala universitet, 14 pp. Uppsala.
- Hallberg, R & Rickard, A. 1973: De små gruvarbetarna i Falun. Svensk Naturvetenskap 1973, 102–107. Stockholm.
- Hyenstrand, Å. 1977: Hyttor och järnframställningsplatser. Jernkontorets forskning, ser. H, 14. Stockholm.
- Håkansson, L. 1977: Sediments as indicators of contamination – investigations in the four largest Swedish lakes. Naturvårdsverket, Rapport. SNV PM 839/NLU Rapp. No. 92, 155 pp. Uppsala.
- Håkansson, L. 1981: Lake sediments in aquatic pollution control programs; principles, processes and practical examples. *Naturvårdsverket, Rapport. SNV PM 1398*, 242 pp. Uppsala.
- Krumbein, W.C. & Pettijohn, F. 1938: Manual of Sedimentary Petrography. Wiley Interscience, New York. 635 pp.
- Kuijpers, A. 1974: Trace elements at the depositional interface and in sediments of the outer parts of the Eckernförder Bucht, western Baltic. *Meyniana* 26, 23–28.
- Lee, F.G. 1970: Factors affecting the transfer of materials between water and sediments. Eutrophication Inf. Program Water Resour. Cent. univ. Wisconsin.
- Lindroth, G. 1963: Gruvbrytning och kopparhantering vid Stora Kopparberget intill 1800-talets början. I. Uppsala. 535 pp.
- Lundqvist, G. 1963: Falu gruvas ålder i geologisk och arkeologisk belysning. Stora Kopparbergs Bergslags AB, Falun. Almqvist & Wiksell. Uppsala. 78 pp.
- Olausson, E. 1972: Water-sediment exchange and recycling of pollutants through biogeochemical processes. *In:* Ruivo, M. (ed.): *Marine Pollution and Sea Life*, 158-161. FAO, Fishing News. Ltd., London.
- Påhlsson, I. & Qvarfort, U. 1975: Sedimentkemiska undersökningar i sjön Runn. Kvartärgeol. avd. Uppsala univ. Uppsala.
- Qvarfort, U. 1977: Some problems associated with exploration geochemistry in mining areas. *Striae Vol.* 7, 77 pp. Uppsala.
- Qvarfort, U. 1979: Sulfidmalmsupplag som miljöproblem. Naturvårdsverket, Rapport. SVN PM 1152. Stockholm.

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- Qvarfort, U. 1980: Sedimenten i sjön Tisken och Falu gruvas ålder. Jernkontorets forskning. Ser. H, 19. Stockholm.
- Salminen, R. 1976: Contamination of stream and lake sediment in Kuopio urban area. J. Geochem. Explor. 5, 406-409
- Stapf, F.M. 1865: Om sjömalmers uppkomst. Jernkontorets Annaler. Ny serie. Tjugonde årgången. Stockholm.

Söderberg, T. 1932: Stora Kopparberget under medeltiden

och Gustav Vasa. Stora Kopparberg Bergslag Ab.

- Falun. 278 pp. Tegengren, F. 1924: Sveriges äldre malmer och bergverk. *Sver. Geol. Unders. Ca 17.* Stockholm.
- Tunberg, S. 1922: Stora Kopparbergets historia. 1. Förberedande Undersökning. Uppsala. Walters, Jr. L.J., Volery, T.J. and Myrer, R.D. 1974:
- Occurrence of As, Cd, Co, Cr, Cu, Fe, Hg, Ni, Sb and Zn in Lake Erie sediments. Proc. 17th Conf. Great Lakes Res., 219-234.