

Surface water quality in Bersbo, Sweden – Fifteen years after amelioration of sulphidic waste

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Abstract

In 1987 two tailings deposits in Bersbo, Sweden, with sulphidic waste were covered with cement stabilised fly ash and natural illitic clay, respectively. The deposits contained approximately the same amount of waste and were separated by a water divide in rather hydrochemically homogeneous watersheds. Between 1983 and 1987 the composition of the surface water in the area was monitored weekly at 9 locations. It was found that the concentration changes in the small brooks in the area had two components; one related to season and the other controlled by single storm events. The monitoring programme continued with a similar frequency until 1997. In July and September 2002 additional samplings were performed. In this comparison data collected only during these two months are discussed. Sampling locations are illustrated in figure 1.

There has been a reduction of all trace element concentrations, as well as sulphate, and at all sampling locations. It seems, however, as if the natural clay has been the best barrier to precipitation and oxygen since the metal concentrations are lowered most at the weir where this material was used as cover. The area of amelioration goes, according to these data, to the inlet to Lake Risten. Further downstream other sources to metal pollution are quantitatively more important than the mining waste.

Introduction

Until the end of the 1980s the abatement of acid mine drainage in Sweden was usually conducted by liming of the effluents, sometimes in combination with sedimentation ponds. By maintaining pH at a high level in this system immobilisation of trace metals was enhanced by precipitation of aluminium and ferric hydrous oxides, which act as adsorbents (Karlsson et al., 1988). This approach serves its purpose well when the addition of lime can be optimised in relation to the discharge of the acid mine drainage. It is, however, not so effective when these requirements are not fulfilled, such as in the case of old tailings deposits.

In the Bersbo mining area in Sweden the concept of covering the waste material was evaluated at a full scale for the first time. The site contained some 15 Mtonnes of coarse tailings that were remains from a mining period that goes back to the 14th century. Two different materials were used; cement stabilised fly ash and a natural illitic clay. In this paper we report some results concerning the success of the different approaches as evidenced by the chemical composition of the surface waters in the area.

Field site

The field site is located some 250 km SSW of Stockholm, Sweden, figure 1. Copper mining at the Bersbo site began already in the 14th century, probably earlier (Tegengren, 1924), and it left some 1 million m³ of coarse tailings. These were found in two deposits of equal size, separated by a small ridge that acted as a water divide. Both deposits were put on lenses of clay, consisting of a compacted illite. The tailings consisted of a complex mixture of sulphides (pyrite, magnetic chalcopyrite, sphaelerite, galena) in a matrix of amphibolitic intrusions in leptite (Tegengren, 1924). The metal concentrations in the ore have been estimated to 0.5-3% Cu; 1-3% Zn; 1% Pb and 20% Fe while the sulphur content was approximately 25% (Karlqvist and Qvarfort, 1979). A more thorough description of the area is given in Allard et al. (1987).

The annual precipitation is 650 mm of which some 200 mm forms the runoff. The variability of the water discharge is extremely high with the most intense flow period during snowmelt, usually in early April. Till dominates the soil type, although clay is found in the lower parts of the watershed. Higher vegetation consists of

mixed spruce and deciduous forest. A detailed description of the water balance can be found in Brandt et al. (1987).

The shallow groundwater in the area is of Ca^{2+} - SO_4^{2-} - CO_3^{2-} type without any obvious marine origin (Allard et al., 1987). The alkalinity is 2.5-3 meq/l and pH 7.1-7.5 why neutralisation of the acid effluents was efficient in both surface- and groundwater.

Technical description of the covers

Remediation was performed during 1987-89. Some 200,000 m³ of the tailings were put into the old mine shafts, in order to minimize the area of the new deposits. All areas freed from waste were limed. On the easterly deposit, natural illitic clay from the area was used as a sealing layer, after compaction. A drainage layer of gravel was put on top of the clay and approximately 1.5 m of moraine was finally added. A similar design was used on the westerly deposit with the exception that cement stabilised fly ash was used as sealing material. Scots pine (westerly deposit) or birch (easterly deposit) were planted. A thorough technical description of the procedure is presented in Lundgren (1990) and Lundgren and Lindahl (1991).

Sampling and analysis

Sampling and analysis of surface water quality began in 1983 at the locations given in figure 1. Weekly sampling constituted the base of the programme although the Gruvsjön outlet and the weir location K (figure 1) were equipped with automatic samplers that collected one sample per day. The latter locations were also equipped with weirs where that allowed for continuous recording of the water flow. This programme was in operation until 1989 where after monthly samples were collected, although with a higher frequency during snowmelt and autumn rains.

Groundwaters were sampled monthly from 1983 to 1986 at a transect across the eastern stream, from some 1 m to 50 m from the deposit. Between 1987 and 1989 groundwaters from the shafts were also included.

Analysis was made with standardised methods, summarised in table 1.

Results and discussion

It has previously been demonstrated that the concentrations of pollutants are highly related to water flow as well as water pathways (Sandén et al., 1995). This is exemplified with the time series of electrical conductivity at the weir location (figure 2). It is evident from the figure that before the remediation an annual amplitude of 5-6 times was present. The magnitude of the amplitude depended on precipitation and its intensity, as reflected by changes of water discharge in the stream. A similar seasonality appears to remain after the remediation but with lower amplitude. This seasonal behaviour of a summary parameter representing dissolved constituents indicates that concentration data must be flow corrected if comparisons are to be made between years. The same applies to any attempts to perform time series analyses to distinguish any trends in the data material.

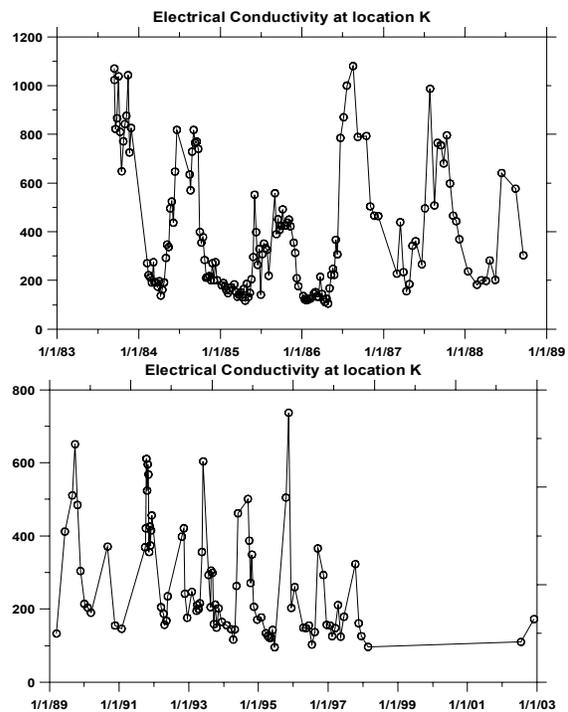


Figure 2. Time series of electrical conductivity ($\mu\text{S}/\text{cm}$) at the weir K. Note the different scales.

Unfortunately it is not possible to perform any flow corrections since water discharge was not recorded during the entire period of water quality measurements. Further, the differences in sampling frequencies between years and sampling sites make it extremely uncertain to perform reliable time series analysis. Instead,

we have used the approach of selecting the period from July to September for comparison between different years. During this time of the years measurements are frequent at all locations. This period also represents the highest concentrations in the surface waters due to minimum dilution with unpolluted groundwater why uncertainties related to measurements are at a minimum. It is also advantageous to base comparisons on these months since autumn rains usually do not alter the chemical composition of the surface waters until late October or November.

According to table 2, the trace metal concentrations in the leachate are lower today, in comparison with the conditions before remediation. The concentration of sulphate at present is about half that of the period until 1986, which infers that the cover has lowered the weathering rate in a similar manner. There appears to have been a concomitant lowering of the other indicators of pollution, i.e. sulphate and trace metals in the leachate. Since liming has been performed on former deposit sites, the pH is evidently not a suitable parameter to evaluate the success of the remediation.

Apparently the covering of the easterly deposit has been a success. The trace metal concentrations are significantly lower after remediation (table 2, figure 3). It appears as if the clay cover serves its purpose well and there is no ocular evidence for any rupture of the layer or break through of leachate. Even annual averages of non-flow corrected quality data support the success.

For iron and sulphate, the annual averages do not positively support that the remediation led to improved water quality. A continuous input of iron from the groundwater would explain why the concentrations of this element have not been lowered. For sulphate the levels are far too high above background in the groundwater why other mechanisms are operating. It cannot be ruled out that the deposit still releases sulphate but it is also possible that it emanates from formerly polluted soil and oxidation of metal sulphides in the stream sediment. It seems therefore important to elucidate which secondary sources of pollution that are operating so long time after remediation.

At the westerly deposit there is visible evidence of pollution at the foot of the deposit, even today. Dark, reddish to yellow precipitates of ferric hydrous oxides are still forming and indicate that there is a transport of weathering products from the tailings. This is also supported by the lowering of pH in the leachate after covering of the waste by almost one unit in spite of liming while the concentrations of sulphate were lowered by 50%. Consequently, there has been an addition of acid, other than sulphuric acid, where the oxidation and hydrolysis of Fe(II) is a plausible process. This is also supported by measurements of the oxygen partial pressure inside the deposit, which clearly indicate concentrations below 0.1%. Any abiotic weathering process would have to proceed by Fe(III) as oxidising agent, which corresponds qualitatively with an increased mobility of Fe(II). The reaction involves steps 1 through 4 (Stumm and Morgan, 1981):

Other sulphides would be oxidised in a similar fashion.

In fact, the concentrations of iron in the leachate increased dramatically after completion of the remediation (figure 4). An increased weathering of the waste is also confirmed by increased concentrations of sulphate. If the system's variability is included, it is difficult to conclude any significant improvements of trace metal concentrations at the inlet to Lake Gruvsjön. There appears to be lowered concentrations of Cd, Cu and Zn, whereas Pb is independent of the remediation. If this is really the case, can only be answered if the dynamics of the system are considered. However, at the inlet to Lake Gruvsjön the covering of the waste is thought to have only minor impact on the hydrology why the effect of the remediation would be minor.

The scavenging of metals in Lake Gruvsjön has obviously improved since the westerly deposit was covered. To some extent this can be explained by an average pH approximately one unit higher than before the remediation. Since the trace metal concentrations do not exceed saturation with their stoichiometric solid phases, respectively, adsorption would most likely control their mobilities. Although the annual average concentrations for Cd, Cu

and Zn are not statistically different when comparing the periods before and after remediation the tendency is obvious. All of them show decreasing concentrations after remediation. Lead differs from this general tendency and its annual averages are rather scattered. Probably this phenomenon is caused by a rather efficient adsorption in the lake, or even immobilisation through formation of its sulphide in the sediment.

This explanation is not entirely satisfying since the increase in pH appears to be in a range where adsorption is rather unaffected, considering the properties of possible precipitates (ferrihydrite, goethite, hematite, hydrargillite, gibbsite). Since the loading of dissolved iron to Lake Gruvsjön has increased approximately ten times (figure 4) while concentrations at the outlet of the lake remains essentially the same (figure 5) it seems reasonable to conclude that coprecipitation in combination with sedimentation reduces the concentrations of dissolved trace metals in the lake water. On the other hand, it means also that the lake serves as an accumulation zone, and thereby constitutes a potential environmental hazard in the future since the stability of the principal adsorbent is highly dependent on pH.

These results pose another important question. Which mechanisms cause the improvement of water quality when a deposit of mine waste is covered? These results do not provide the final answer but in this system it seems more important to reduce infiltration of rainwater than lowering the rate of weathering of sulphides.

Conclusions

This study has clearly demonstrated that the surface water quality improves considerably after a successful covering of mine waste. The principal function of the cover is apparently to prevent that any water leaves the waste. The

oxidative weathering of the sulphides does not cease even if the oxygen is depleted. There is just a change of oxidising agent and the weathering will continue as long as there is sufficient amount of Fe(III) in the waste. Preventing oxygen intrusion might under such circumstances lower the oxidation rate of Fe(II) and thereby leading to a more rapid depletion of the Fe(III).

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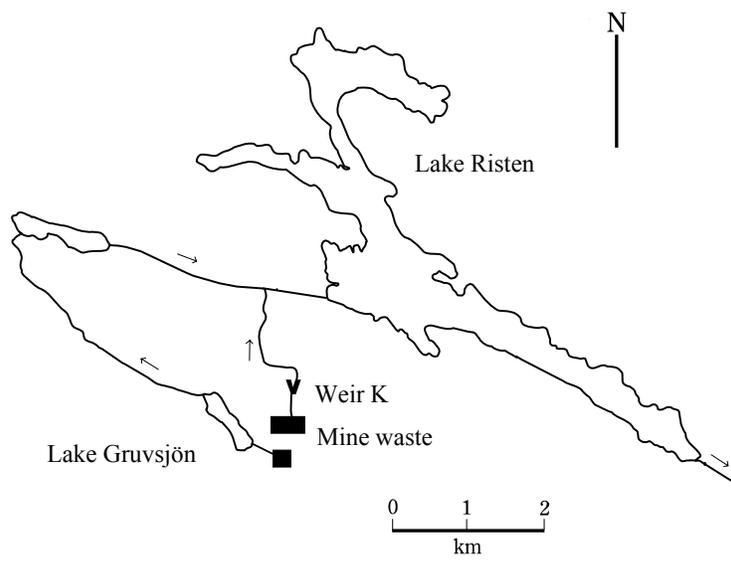


Figure 1. Map over the field site.

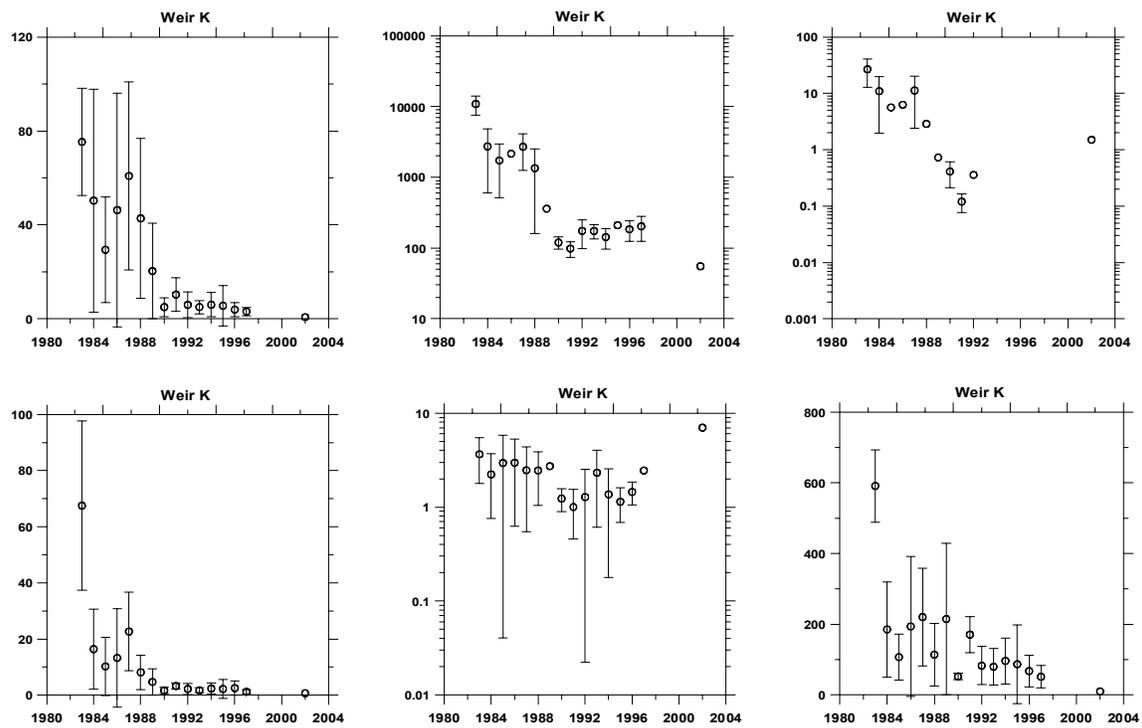


Figure 3. Annual averages for concentrations of trace metals and sulphate at the weir K.

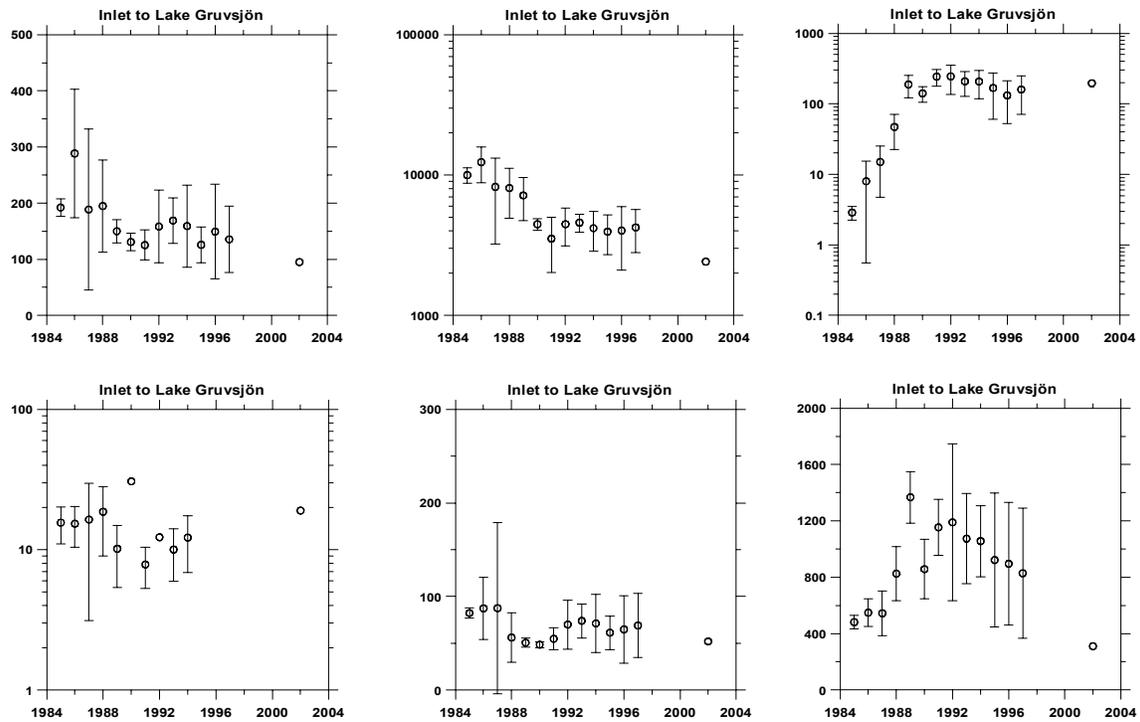


Figure 4. Annual averages for concentrations of trace metals and sulphate at the inlet to Lake Gruvsjön.

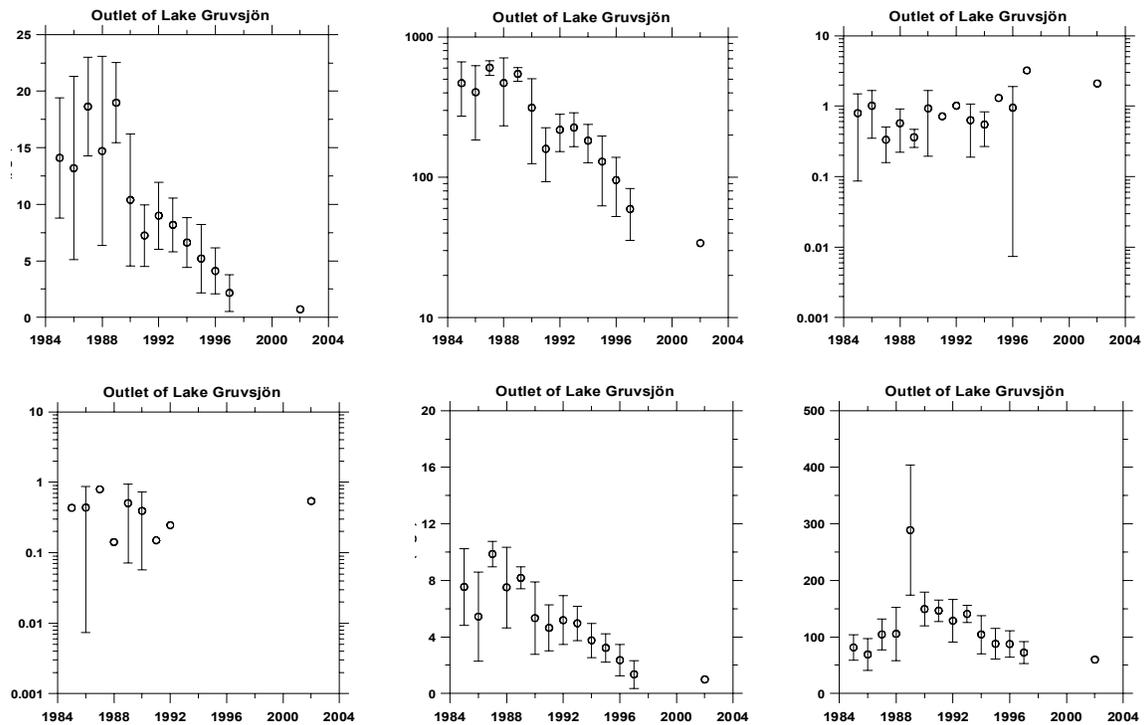


Figure 5. Annual averages for concentrations of trace metals and sulphate at the outlet of Lake Gruvsjön.

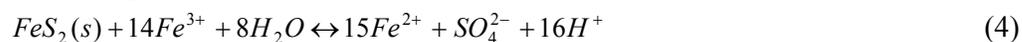


Table 1. Analytical parameters and procedures.

Parameter	Method
pH	Combination electrode
Alkalinity	End point titration to pH 5.2 with 0.02 M HCl
Electrical conductivity	Electrode
Chloride	Precipitation titration with Ag^+ , CE from 1990
Sulphate	Flow injection, CE from 1990
Dissolved oxygen	Winkler method
Metals (Al, Ca, Cd, Cu, Fe, K, Mg, Mn, Na, Pb, Zn)	Atomic absorption until 1994, thereafter ICP-MS
TOC	Shimadzu 5000 TOC Analyzer

Table 2. Average values of some selected indicators on pollution based on average values for July-September (b.d. below detection).

	Period	pH	SO ₄ ppm	Cd ppb	Cu ppb	Pb ppb	Zn ppb
Leachate	1983-86	3.36	603	197	9643	35	66000
	2002	2.87	307	69	5368	22	30470
Inlet Lake Gruvsjön	1983-88	4.4	700	223	11594	16	99000
	1989-97	3.67	994	148	4477	10	66000
	2002	3.21	307	94	2418	19	52410
Outlet Lake Gruvsjön	1983-88	5.1	84	14	479	0.8	7047
	1988-97	5.06	117	7	205	0.1	3817
	2002	6.07	59	0.55	14	0.52	186
Weir	1983-88	3.93	333	89	4282	14.3	28116
	1989-97	5.47	123	26	793	2.3	5550
	2002	6.21	10	0.71	55	1.49	700
Inlet Lake Risten	1983-88	6.86	29	4.46	90	0.48	1107
	1989-97	6.74	22	0.44	16	0.49	201
	2002	6.67	4	0.02	5.6	0.49	20
Outlet Lake Risten	1983-88	7.39	24	0.03	11	b.d	117
	1989-97	7.55	22	0.05	6.4	b.d	50
	2002	6.77	16	0.23	6.1	0.36	25