

Environmental Impacts of an Old Mine Tailings Deposit – Hydrochemical and Hydrological Background

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The release and distribution of metals as well as acid from a mine tailings deposit into a stream have been studied, particularly the distribution and neutralization of acids, the distribution and chemical speciation of metals and metal adsorption and precipitation phenomena. The Bersbo mine, some 250 km SSW of Stockholm, was selected for the study. An over-view is given of the local geology, hydrology and hydrochemistry as well as the research program (field measurements, sampling, analytical procedures and modelling). The databases (chemical and hydrological) generated within the project are outlined. The paper serves as an introduction to a series of three papers based on field data from the same area.

Introduction

The spreading of hazardous elements in the ecosystem, both from natural and anthropogenic sources, has become a major environmental problem (Förstner and Wittman 1980; Salomons and Förstner 1984).

Despite the extensive amount of published data related to the releases of metal contaminants it is not generally possible to quantitatively model the transport and redistribution of metals in the aqueous environment. Knowledge of chemical speciation, interactions with geologic phases, transportation and natural background levels is rather limited.

The general aim of the present project is to gain information on the release and redistribution of acid and metals (particularly Al, Fe, Mn, Cu, Zn, Cd and Pb)

from a point source represented by a mine tailings deposit.

The following areas are studied in detail:

- 1) Distribution and neutralization of acid; integrated hydrological and chemical modelling.
- 2) Distribution and chemical speciation of metals
- 3) Metal adsorption and precipitation phenomena.

The Bersbo mine in the municipality of Åtvidaberg, some 250 km SSW of Stockholm (16°3'E, 58°16'N) was selected as a suitable point-source (Törnebohm 1885; Sundius 1921; Sandegren *et al.* 1924; Tegengren 1924; Söderbäck 1974; Hellström *et al.* 1983). The release of metals from the mine tailings as well as biological uptake have previously been studied in the area (Jacks 1976; Qvarfort 1977; Karlqvist and Qvarfort 1979; Noorlind *et al.* 1980; Börkén 1982; Adelsvärd *et al.* 1983; Lundholm and Andersson 1985). Some preliminary studies by the present project group have also been reported (Karlsson and Sandén 1984; Allard *et al.* 1985; Allard *et al.* 1986).

The Bersbo Area

Mining History

Mining of copper in the Åtvidaberg region started already in the 14th century, possibly even earlier, particularly in the Närstad and Bersbo areas (Törnebohm 1885; Tegengren 1924; Sandegren *et al.* 1924) cf. Fig. 1. Rational mining was undertaken from ca. 1760 to 1902 with a peak production during 1855-1870. During this period the Åtvidaberg mining area was the largest copper producer in Sweden.

The mining at Bersbo has resulted in a continuous system of shafts and tunnels down to a depth of 465 m.

The mine is now filled with water. Visible remains of the mining is the large deposit of tailings, covering some 0.2 km² and containing ca. 300,000 m³ of waste rock.

Geology

The oldest rocks in the Åtvidaberg area are of Precambrian age, and supra crustal origin consisting of rocks as amphibolites, leptites and mica schists (Tegengren 1934). Intrusions of younger granitic rocks (oligoclase rich gray gneissic granites as well as intermediate and alkaline red gneissic granites) are frequent, just as formations of metamorphic gabbro and diabases. The Bersbo mines are situated in a zone of leptitic rocks, however with a large intrusive amphibolite body containing hornblende, augite and calcium rich plagioclase as well as some younger pegmatite rich granitic intrusions. The ore veins, which contain pyrite, magnetic chalcopyrite and occasionally sphaalerite and galena are found in the leptite at some distance

Mine Tailings – Hydrological Background

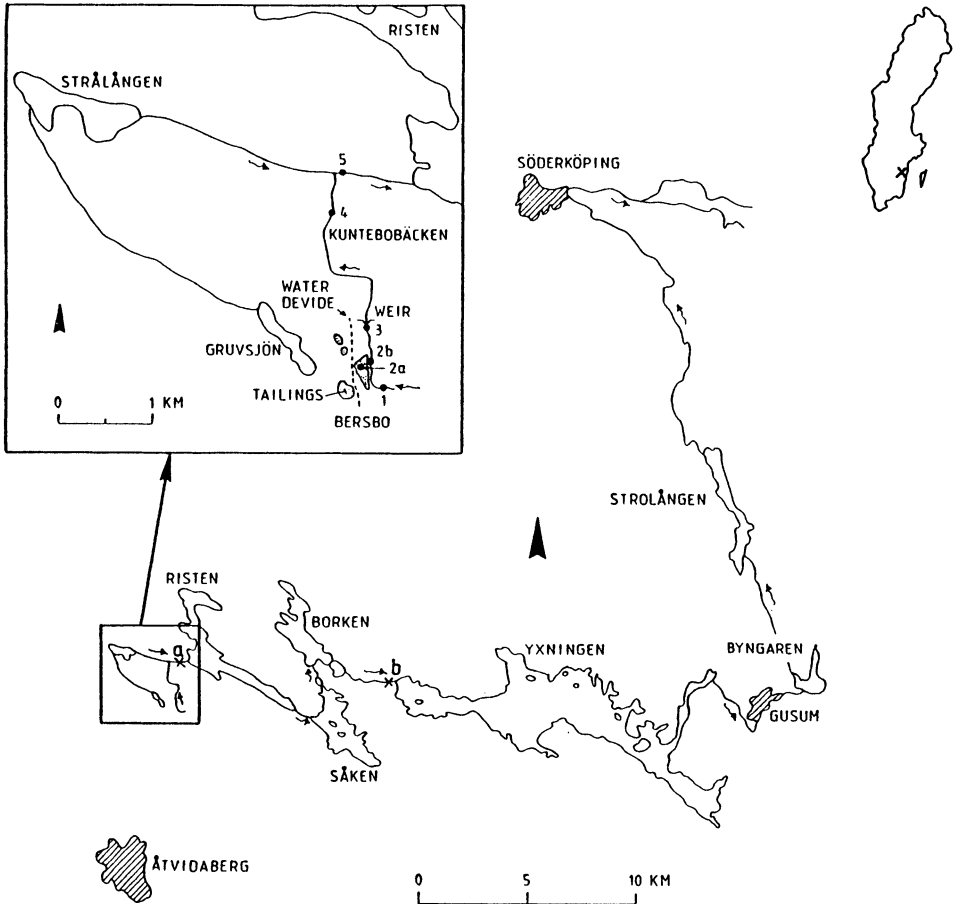


Fig. 1. The Bersbo area with sampling locations in the eastern stream.

from the amphibolite/leptite interphase, or associated with the amphibolite. Thus, they form zones in the leptite that are parallel to the large amphibolite intrusion. All these rock types are found among the tailings.

The concentrations of metals in the ore have been estimated to be 0.5-3% copper, 1-3% zinc, 1% lead and 20% iron (as well as 25% sulphur), based on the available data on the total production (Karlqvist and Qvarfort 1979).

The bedrock in the vicinities of Bersbo is covered with till and, in lower parts, clay. The clay zone has a thickness of 2-4 m underneath the tailings deposit. It is probable that this clay contains substantial amounts of calcareous shells.

Hydrology

A ridge through the deposit area will act as an effective water divide. Surface water from the precipitation and percolating water from the deposit will be drained



Fig. 2. Monthly precipitation, in mm, at the Bersbo area and water flow in the eastern stream.

essentially in westerly direction (to the Lake Gruvsjön, altitude 80.5 m above sea level) and in easterly direction (to the Kuntebobäcken, originating in a bog area at 97.1 m above sea level) Fig. 1. The westerly drainage will pass through Lake Strålängen and meet the easterly drainage at Missmyra, Fig. 1, and further into Lake Risten (at 62.4 m above sea level) and into a series of lakes (Lake Såken, Lake Borken, Lake Yxningen).

The Bersbo area is largely covered with coniferous forests, with some minor bogs upstream of the deposit, and a few cultivated fields along the stream.

The hydrology of the unaffected part of the Bersbo area is typical for the south-east of Sweden. The average precipitation is estimated to 650 mm/year, the evapotranspiration is 450 mm/year, and 200 mm/year forms the runoff. The variability of the water balance components is high, in particular during winter conditions. Some winters are stable with a pronounced snowmelt flood in spring, while others are mild with frequent episodes of rain, snowfall and melt. The water balance of the study period is summarized in Fig. 2.

The till soil in the area can be assumed to be pervious enough to prevent any surface runoff, unless it reaches full saturation. This is probably also true when the soil is frozen, even if some extreme situations with surface runoff caused by icing on the ground may occur locally. The hydrological pathways in a dry and a wet situation are schematically illustrated in Fig. 3. The combination of superficial groundwater in the more acid upper horizon of the soil during a flood situation is responsible for the temporary drop in alkalinity and pH, while it recovers during dry periods when contributions from deeper groundwater dominate the flow.

The area is rich in lakes. Further down the river system these are dominating the hydrological and hydrochemical response. This is illustrated by Fig. 4, which shows a model simulation of the hydrograph at two points, one upstream lake Risten and one downstream lake Borken, by the PULSE-model (Bergström *et al.* 1985) with a subroutine for routing of the flow through lakes attached to it.

Mine Tailings – Hydrological Background

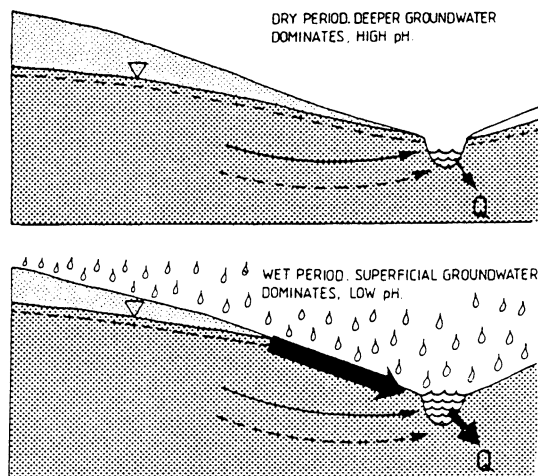


Fig. 3. Schematic picture of the hydrological pathways in the unaffected forested parts of the Bersbo area.

The water balance of the deposit differs from that of the surrounding area, mainly because of lack of vegetation but also due to the coarse size fraction of the waste material. Evaporation is low and transpiration is almost negligible. The hydrological response will therefore be relatively fast even after long dry periods. The deposit under study contains ca. 300,000 m³ of waste rock and covers ca. 0.2 km². Precipitation readily percolates through it since coarse size fractions dominate. Hydraulic conductivity has been estimated to 10⁻³ m/s but the leachate is to some extent retained at the bottom of the deposit before it enters the stream. The drainage areas to the respective sampling sites are given in Fig. 5. Details of the water balance of the area are further discussed by Brandt *et al.* (1987).

Sampling and Chemical Analysis

Sampling in the eastern stream began in Sept. 1983 at the sites given in Fig. 1. Samples have been collected weekly and combined with automatic samplers (1-4 samples per day, sites 3 and 4) to allow sampling under rapid changes in flow

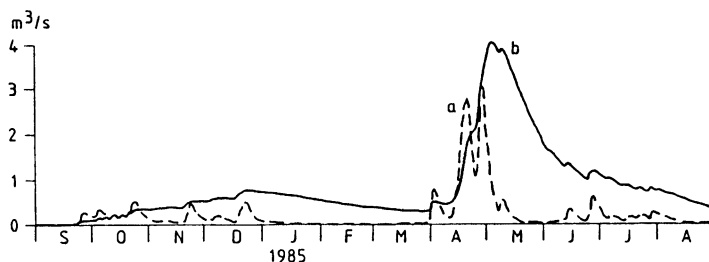


Fig. 4. Model simulated hydrographs at the inlet to the first large lake (a) in the water system downstream the Bersbo area and the outlet of a large lake further down in the system (b), cf Fig. 1.

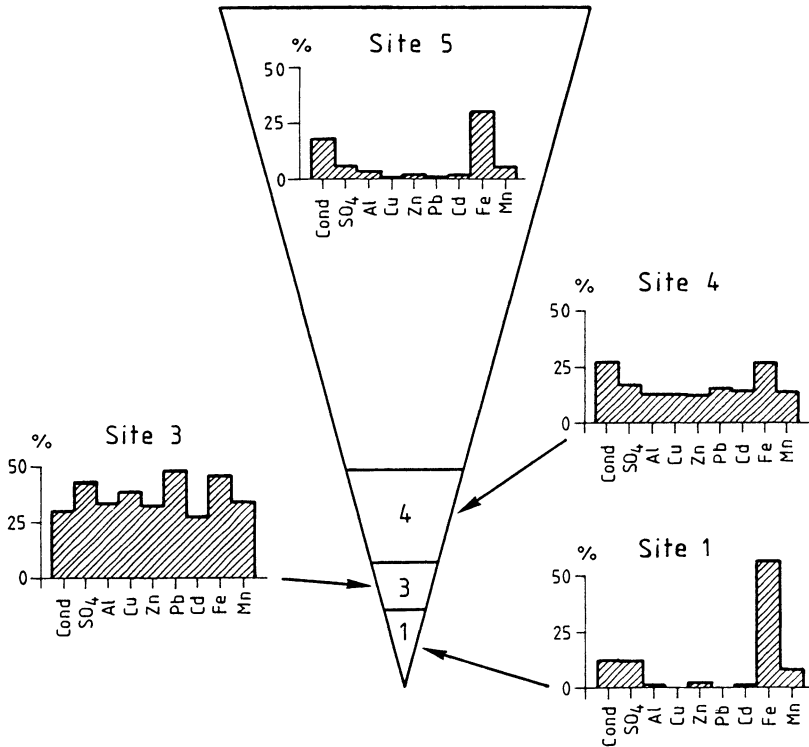


Fig. 5. Schematic presentation of the drainage areas to the respective sampling sites in the eastern drainage. Average values of chemical constituents are given as percentage of those corresponding to the leachate. The triangular areas are proportional to the drainage areas to the five sites, respectively.

conditions. Water flow has been continuously recorded at the weir at site 3 and the daily precipitation at the gauge located close to the water divide (Fig. 1). The sampling site 1 represented »background« conditions of surface water in the area. The leachate was characterized by sampling in the open water table at the clay interface (2a) and just as it entered into the stream (2b). Sampling sites 3 and 4, at 400 m and 1400 m from the outlet, were chosen to represent various input of leachates and subsequent mixing with groundwater (cf. Table 1). The most remote sampling site (5) represented the junction of the two major streams, ca 2,100 m from the deposit with regard to the eastern stream. Samples were also collected from 6 groundwater wells installed perpendicular to the eastern stream adjacent to the deposit. These samples represented water in contact with or just below the illitic clay zone, at 4-6 m below the soil surface.

Sampling and sample preparations were made in acid washed (ca 2 M HNO₃ + 1 M HCl) plastic (polyethylene or polypropylene) vessels. Samples for determinations of dissolved oxygen were collected in a way as to minimize contact with the

Mine Tailings – Hydrological Background

Table 1 – Composition of the Water in the Easterly Stream (Kuntebobäcken), Sept. 1983 – Dec. 1986; weekly samples (Locations according to Fig. 1).

| Constituent ^a | Site 2b (n=65) | | | | Site 3 (n=21) | | | | Site 4 (n=86) | | | | Site 5 (n=90) | | | |
|-------------------------------|-------------------|-------|-------|-------|------------------|-------|-------|-------|------------------|-------|-------|-------|------------------|-------|-------|-------|
| | Mean | S.D. | Min | Max | Mean | S.D. | Min | Max | Mean | S.D. | Min | Max | Mean | S.D. | Min | Max |
| Na | 249 | 68 | 128 | 374 | 281 | 60 | 96 | 450 | 400 | 126 | 118 | 827 | 337 | 67 | 169 | 501 |
| K | 104 | 24 | 48 | 166 | 84 | 44 | 31 | 227 | 98 | 53 | 35 | 291 | 115 | 29 | 66 | 257 |
| Mg | 1270 | 496 | 115 | 2068 | 857 | 1342 | 32 | 6244 | 680 | 950 | 40 | 4800 | 240 | 56 | 118 | 392 |
| Ca | 437 | 160 | 137 | 885 | 356 | 184 | 44 | 1324 | 520 | 240 | 67 | 1204 | 493 | 142 | 149 | 1089 |
| Al | 1760 | 735 | 7 | 3543 | 481 | 567 | 0.7 | 2539 | 175 | 222 | 2 | 1510 | 52 | 35 | 7 | 220 |
| Fe | 90 | 68 | 17 | 445 | 50 | 42 | 13 | 305 | 23 | 14 | 1 | 83 | 28 | 16 | 11 | 103 |
| Mn | 92 | 33 | 23 | 153 | 29 | 29 | 3 | 125 | 13 | 11 | 3 | 87 | 4 | 4 | 0.7 | 19 |
| Cu | 151 | 49 | 15 | 254 | 49 | 54 | 3 | 250 | 19 | 25 | 0.5 | 173 | 1 | 0.9 | 0.2 | 5 |
| Zn | 992 | 438 | 2 | 2031 | 283 | 361 | 18 | 2431 | 140 | 152 | 3 | 936 | 16 | 12 | 0.5 | 67 |
| Cd | 1.742 | 0.426 | 0.507 | 2767 | 0.394 | 0.364 | 0.018 | 1.432 | 0.210 | 0.165 | 0.009 | 0.889 | 0.021 | 0.022 | 0.001 | 0.106 |
| Pb | 0.170 | 0.057 | 0.063 | 355 | 0.047 | 0.052 | 0.001 | 0.265 | 0.009 | 0.015 | 0.001 | 0.126 | 0.002 | 0.003 | 0.001 | 15 |
| Cl ⁺ | 62 | 16 | 25 | 113 | 231 | 70 | 68 | 415 | 378 | 182 | 99 | 1117 | 327 | 100 | 132 | 646 |
| SO ₄ ²⁺ | 6220 | 2010 | 1700 | 10300 | 2130 | 2000 | 300 | 7940 | 1257 | 980 | 243 | 5752 | 336 | 181 | 160 | 1160 |
| CO ₃ ²⁺ | 0 | – | – | – | 0.11 | 0.06 | 0 | 0.22 | 0.42 | 0.44 | 0.01 | 2.15 | 0.94 | 0.40 | 0.24 | 2.45 |
| O ₂ ^b | 10.2 | 2.2 | 7.1 | 15.8 | 7.6 | 2.3 | 0.6 | 10.8 | 10.4 | 1.9 | 2.0 | 14.3 | 6.6 | 2.6 | 0.7 | 11.1 |
| pH | 3.40 | 0.21 | 2.93 | 3.91 | 4.42 | 0.60 | 3.41 | 5.88 | 5.82 | 0.73 | 4.20 | 7.3 | 6.75 | 0.24 | 6.22 | 7.28 |

^a Concentrations in µ mol/l

^b Concentrations in mg/l

Table 2 – Analytical methods

| Parameter | Analytical method/equipment |
|-----------------------|--|
| pH | Radiometer PHM84, glass electrode (G2040C) and calomel reference electrode (K4040) |
| Alkalinity | Titration with 0.02 M HCl to pH 5.4 using the electrodes above under continuous flux of carbonate free N ₂ |
| Electric conductivity | Radiometer CDM83 instrument with electrode CDC104 automatic temperature compensation to 25°C |
| Chloride | Precipitation titration in KNO ₃ (0.50M) + HNO ₃ (0.0105M) with Ag ⁺ (0.01M) and indicating the inflexion point with the Ag/AgCl/HgSO ₄ electrode pair |
| Sulphate | Indirectly by ionexchangeable cations until Dec. 84 and thereafter directly by the Ba-methylthymole blue method (Flow injection technique) |
| Dissolved oxygen | Winkler method |
| Metal constituents | Atomic absorption spectrophotometry (Perkin-Elmer 5000/Zeeman instrument). Direct calibration of the instrument (flame or furnace) after control of non-specific absorbance at the selected wavelengths. |
| Phase separation | Pressure filtration using 0.47 mm(Ø) polycarbonate filters (Nuclepor. Corp.), pore size 0.40 µm. |

atmosphere. Water samples were always collected from the center of the stream to avoid resuspending stationary sediment phases.

The analytical program included major constituents of the waters (Na, K, Mg, Ca, Cl⁻, SO₄²⁻, O₂(aq) and pH) as well as trace metals (Al, Fe, Mn, Cu, Zn, Cd and Pb). Separation of solids and the operationally defined soluble phases was made by pressure filtration through a polycarbonate filter. Analytical procedures are summarized in Table 2.

Phase separation and chemical analysis of non-metal constituents were usually completed within 8 hours after the samples had been collected. Metal analysis was performed on acidified samples as well as on acidified filtrates and were completed within 48 h. The use of automatic samplers did not allow for accurate determinations of the solid and soluble phases of trace metals since precipitation obviously occurred already at moderate pH during the storing in the field. Only major constituents were determined in such samples.

Mine Tailings – Hydrological Background

Table 3 – Composition of the water in the Bersbo area.

| Constituent ^a | Groundwater (n=18) | | Leachate (site 2a) (n=80) | | Surface water ^b (site 1) (n=120) | |
|--|-----------------------|------|---------------------------------|-------|---|-------|
| | Mean | S.D. | Mean | S.D. | Mean | S.D. |
| Na | 601 | 117 | 290 | 90 | 254 | 62 |
| K | 165 | 30 | 118 | 34 | 45 | 32 |
| Mg | 2115 | 1600 | 1881 | 1424 | 355 | 711 |
| Ca | 1900 | 240 | 512 | 288 | 207 | 149 |
| Al | 16 | 36 | 1875 | 946 | 37 | 21 |
| Fe | 36 | 50 | 160 | 240 | 57 | 73 |
| Mn | 23 | 5 | 111 | 44 | 7 | 16 |
| Cu | 0.3 | 0.5 | 132 | 67 | 0.6 | 1 |
| Zn | 3.9 | 5 | 1066 | 550 | 8 | 17 |
| Cd | c | c | 1.790 | 0.824 | 0.028 | 0.051 |
| Pb | c | c | 0.130 | 0.066 | 0.003 | 0.003 |
| Cl ⁻ | 145 | 157 | 62 | 23 | 204 | 83 |
| SO ₄ ²⁻ | 3 | 2 | 7701 | 3134 | 367 | 354 |
| CO ₃ ²⁻ ^d | 1.58 | 0.28 | 0 | – | 0.23 | 0.36 |
| O ₂ ^e | – | – | 8.4 | 2.00 | 4.9 | 2.9 |
| pH | 7.18 | 0.18 | 3.42 | 0.20 | 5.33 | 0.41 |

a Concentrations in $\mu\text{mol/l}$

b Cf. Figs. 1 and 2

c Less than 3 nmol/l

d Concentrations in mequ/l (not determined when pH < 5.4)

e Concentrations in mg/l

General Hydrochemistry

The shallow groundwater in the area is of $\text{Ca}^{2+}\text{-SO}_4^{2-}\text{-CO}_3^{2-}$ -type (Table 3) with an alkalinity of 2.5-3 meq/l and pH 7.1-7.5 (max. 7.8). The total salinity is fairly high (< 500 mg/l) but without any obvious marine origin (low $\text{Na}^+\text{-Cl}^-$ -concentrations). The alkalinity/pH-properties reflect the presence of calcium carbonate (as shells) in the ground. There is no significant contamination of the groundwater with effluents from the deposit at the location of the groundwater sampling, some 20 m east of the deposit at 4-6 m below the soil surface. Evidently, the clay layer acts as an effective barrier that prevents direct exchanges between leachates and groundwater.

The surface waters close to the deposit typically have total salinities of 70-75 mg/l, Tables 1 and 3, with $\text{Ca}^{2+}\text{-Na}^+$ as major cations, sulphate as dominating anion and pH around 4.9-5.7. Water coming from bog areas has large contents of organics (humic and fulvic acids) which can be as high as 50 mg/l.

Oxygenated water percolating through the tailings will lead to a severe weathering of the sulphide minerals (Stumm and Morgan 1981). The final products would eventually be metal ions, sulphate and hydrogen ions (a yield of totally 2-4 moles of

H⁺ and 1-2 moles of SO₄²⁻ per mole of Fe, Cu, etc.). The released hydrogen ions could partly be consumed in the weathering of amphiboles, micas and feldspars of the leptite-amphibolite host rock. The released metals may form sparingly soluble compounds, e.g. hydroxides, hydroxysulphates, etc.

Estimated yearly maximum amounts of metals that are leached from the deposit in the western and eastern directions, respectively, are ca. 3,000 kg aluminium, 6,000 kg zink, 700 kg copper, 25 kg cadmium and 2 kg lead, based on data from 1983-86 (in the Kuntebobäcken and in the outlet from Lake Gruvsjön). The corresponding produced amount of sulphuric acid is of the order 100,000 kg. There is however large variations in the transported amounts between different years, related to hydrological factors. Annually transported metal loadings 1983-86 for the easterly drainage are 1,300 kg aluminum, 1,600 kg zink, 340 kg copper, 6 kg cadmium and 1,5 kg lead.

The leachates from the tailings are mixed with precipitation and alkaline groundwater in the system of streams that are draining the deposit. Thus, several different processes will lead to a decreasing acidity and depletion of metals from the aqueous phase in the streams:

- Dilution with surface water and groundwater
- Neutralization with alkaline groundwater
- Precipitation of sparingly soluble metal hydroxides, notably of iron and aluminium
- Coprecipitation or adsorption of dissolved hydrolysable trace metals
- Adsorption of trace components on any exposed surfaces
- Sedimentation of metal rich particle fractions.

The result, in terms of gradual changes in surface water chemistry, is illustrated in Table 1 (cf. Table 3). The variations caused by the hydrological conditions and the subsequent chemical responses are further discussed in Sandén *et al.* (1987).

Perspectives on Metal Distribution in the Area

The present database consists of time series with field measurements of hydrological parameters (e.g. water flow at site 3 and precipitation) and the chemical composition of the stream water with its suspended matter at the respective locations. During the three years of sampling the variations in flow conditions as well as chemical conditions have been large (cf. Tables 1 and 3). Changes in chemical characteristics are, however, not confined to any specific location but rather related to seasonal variations as well as more rapid responses to single precipitation events (cf. Fig. 2). Thus, the area can serve as an excellent model system. Effects of pH on metal speciation and distribution, solid phase formation, sorption, sedimentation etc. are demonstrated. The remarkable buffering capacity of the system is

briefly discussed since it causes a rapid increase in pH with increasing distance from the deposit. Data from the field site are presently used for modelling of water flow in the stream and the impacts of hydrological conditions on pH and alkalinity (Brandt *et al.* 1987). These parameters are likely to influence the chemical state of metals in the stream, especially those originating from the leachates (Sandén *et al.* 1987). The partitioning of metals between suspended solid phases as a result of changes in chemical conditions (e.g. pH, saturation state and distribution of dissolved species) is discussed in Karlsson *et al.* (1987).

The present series of papers consider phenomena related to the eastern drainage of the deposit. Measurements have recently started to include the more slowly reacting western drainage as well as the lake system downstream the area.

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