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# Effects and quantification of acid runoff from sulfide-bearing rock deposited during construction of Highway E18, Norway.

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## ABSTRACT

The highway E18 between the cities of Grimstad and Kristiansand, southern Norway, constructed in the period 2006-2009, cuts through sulfide-bearing rock. The geology of this area is dominated by slowly-weathering gneiss and granites, and oxidation of fresh rock surfaces can result in acidification of surface water. Sulfide-containing rock waste from excavations during construction work in the period 2006-2009 was therefore deposited in three waste rock deposits off-site. The deposits consist of 630,000 – 2,360,000 metric tons of waste rock material. Shell sand and limestone gravel were added in layers in adequate amounts to mitigate initial acid runoff in one of the deposits. The shell sand addition was not adequate in the two others. The pH in the effluents from these two was reduced from 4.9-6.5 to 4.0-4.6, and Al concentrations increased from below 0.4 mg/L to 10-20 mg/L. Stream concentrations of trace metals increased by a factor of 25-400, highest for Ni, and then in decreasing order for Co, Mn, Cd, Zn and Cu. Concentrations of As, Cr and Fe remained unchanged. Ratios of Co/Ni and Cd/Zn indicate that the metal sources for these pair of metals are sphalerite and pyrite, respectively. Based on surveys and established critical limits for Al, surface waters downstream became toxic to fish and invertebrates. The sulfur release rates were remarkably stable in the monitoring period at all three sites. Annual sulfur release was 0.1-0.4 % of the total amount of sulfur in the deposit, indicating release periods of 250-800 years. Precipitates of Al-hydroxysulfates, well-known from mining sites, were found at the base of the deposits, in streams and also along the ocean shore-line. The effects of added neutralization agents in the deposits and in treatment areas downstream gradually decreased, as indicated by reduced stream pH over time. Active measures are needed to avoid harmful ecological effects in the future.

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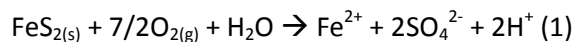
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## 1. Introduction

The devastating water quality and ecological effects of exposing sulfide-bearing bedrock to the atmosphere are widely known from mining areas all around the world (Nordstrom and Alpers, 1999; Nordstrom, 2011a). Most investigations of the complex hydrobiogeochemical processes that create acid rock drainage (ARD) are from these areas. Less frequently described are similar effects from other man-made disturbances of sulfide-bearing bedrock, e.g. road-construction.

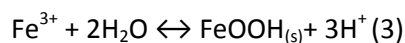
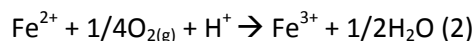
### 1.1. Sulfide oxidation

Oxidation of sulfide minerals (e.g. pyrite;  $\text{FeS}_2$ ) results in the production of ferrous and hydrogen ions according to equation 1:



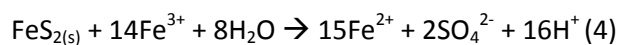
Availability of water and oxygen drives the reaction, and sulfuric acid ( $\text{H}_2\text{SO}_4$ ) is produced. This equation is a simplification of a much more complex process (Nordstrom, 1982; Nordstrom and Alpers, 1999). The acid is partly neutralized by the weathering of the host rock that liberates base cations, such as calcium and magnesium.

The  $\text{Fe}^{2+}$  is oxidized to  $\text{Fe}^{3+}$ , which may be precipitated as iron oxyhydroxides, such as goethite,  $\text{FeOOH}$ , according to equation 2 and 3.



The net effect of the oxidation and precipitation of iron is production of two moles of  $\text{H}^+$  for each mole of  $\text{Fe}^{2+}$  oxidized, hydrolyzed and precipitated (Nordstrom and Campbell, 2014). As the oxidation of iron may take place close to the source, iron may be removed from the leached water before the acid water from the source enters the surface water.

The oxidation of sulfides is accelerated by the resultant  $\text{Fe}^{3+}$ , which oxidizes  $\text{FeS}_2$  much faster than oxygen at low pH:



Hence, availability of  $\text{Fe}^{3+}$  is critical for the rate of this abiotic oxidation.

In the presence of S- and Fe-oxidizing bacteria of the type *Acidithiobacillus ferrooxidans* and *Leptospirillum ferrooxidans*, oxidation of  $\text{FeS}_2$  will proceed even faster. Availability of oxygen will be essential to replenish the supply of  $\text{Fe}^{3+}$  (Nordstrom and Alpers, 1999; Nordstrom, 2003; Herbert, 1999).

### 1.2. Al release

The acidity produced as described above, if not neutralized, dissolves Al of aluminosilicates. Al released from these minerals acts as a buffering agent against very low pH (Hindar and Lydersen 1994), but the dominating Al-species tend to be toxic to biota.

In the Lillesand area, base cation concentrations are low and aluminum ions tend to dominate affected stream-water together with sulfate (Hindar and Lydersen, 1994; Hindar and Iversen, 2006). These authors found that only 40-55 % of the sulfate was balanced by Ca and Mg from the host rock in this area of slowly weathering bedrock, known to be extremely sensitive for acidification due to acid rain. The rest of the anionic charge is almost totally balanced by Al-ions, which means that Al concentrations may be extremely high (several mg/L), in contrast to water acidified from acid rain (pH 4.4-6.0; 0.01-0.5 mg Al/L; Wright et al., 1980).

If buffering substances, such as calcite (calcium carbonate;  $\text{CaCO}_3$ ), are added to neutralize the acid produced by sulfide oxidation, the Ca concentration will increase and thereby contribute to the charge balance. The Al release from aluminosilicates will be reduced and Al-concentrations will be correspondingly lower. Adequate buffering by added carbonates will increase pH (lowering the  $\text{H}^+$  concentration). If calcite is mixed in a deposit of sulfide-bearing rock material, runoff pH may remain neutral for a few years (Strömberg and Banwart, 1999a), but the store of carbonates will gradually be reduced as the neutralization reactions proceed. Quantification of the acid-producing potential of the rock disposal, the inherent base production of the host rock, and the neutralizing capacity of added base is necessary to assess potential long-term ecological effects on the runoff.

### *1.3. Examples from construction works*

A few examples may illustrate the importance of pre-construction planning prior to blasting and disposal of waste rock.

A fish kill (brook trout) was reported following the completion of a highway in the Great Smoky Mountains National Park, USA, in 1963 and continues to be a problem (Hammarstrom et al., 2003). The roadbed material contained iron sulfide minerals, and the stream draining part of the area acidified and became devoid of fish for at least 10 years (Huckabee et al., 1975).

Road construction through sulfidic materials in Virginia, USA, threatened water quality, and sulfide hazard analysis was recommended by Orndorff and Daniels (2004) as an essential step in the pre-design phase of highway construction. Similar recommendations were given by Hammarstrom et al. (2005) after a highway construction project in Pennsylvania, USA, that exposed pyrite with high S-content (8 % S by weight). This material was used as road base and fill, and leached Zn, As, Co and Ni to the seepage water.

Another example comes from construction of The Halifax International Airport, Nova Scotia, Canada, from 1955 to 1960 (Hicks, 2003). Bedrock outcroppings of pyrite-bearing slate were blasted to create a level area for runways and taxiways, and several bog areas were filled with this material. Downstream the Johnson Brook was heavily acidified and had concentrations of aluminum in the range 0-20 mg Al/L in the period 1985-2000 (Hicks, 2003). The nearby McDowell Brook had numerous pH values near 2.5. The stream water was obviously still extremely toxic to fish and invertebrates 40-45 years after the airport construction. News releases in 2012 and 2013 report on recurring pothole sinks that might be related to continued acid generation.

In Norway ARD-problems are mainly associated with abandoned mining-sites (overview by Banks et al., 1997).

### *1.4. Highway E18*

Problems from blasting of sulfide-bearing bedrock in the Lillesand area, southern Norway were well-known prior to the onset of construction of the new line for Highway E18 (Hindar and Lydersen,

1994; Hindar and Iversen, 2006). The effect was first discovered in the area in the 1990's as a stream and a drinking water reservoir were acidified after construction of a new industrial area. Oxidation of sulfide minerals mobilized large amounts of aluminum that were reported to have toxic effects on brown trout (Hindar and Lydersen, 1994).

The new highway E18 in the Lillesand area was constructed in the period 2006-2009 and cuts partly through sulfide-bearing bedrock. During the construction of Highway E18 sulfide-bearing rock was identified by the construction joint venture company CJV E18 Grimstad-Kristiansand (CJV), and sulfide-bearing material was deposited off-site shortly after blasting.

Although large amounts of liming material were mixed into three waste rock deposits, data from the environmental monitoring program (EMP) revealed production of ARD and potential harmful effects (Hindar et al., 2007).

This study is based on weekly data from the environmental monitoring program (EMP; Hindar et al. 2007) and additional studies. The sites are well delimited waste rock deposits placed in small well-defined catchments. The purpose of the work was three-fold: 1) document ARD-effects on receiving surface waters, 2) calculate mass transport and S release rates, and 3) evaluate effects on water quality of passive and active countermeasures.

## **2. Material and methods**

The Landvik monitoring station 10 km east of Lillesand, southernmost Norway (Fig. 1), had annual mean temperatures of 7.5 °C and annual mean precipitation of 1400 mm in the period 2006-2010 (VIPS-landbruk.no). Mean specific runoff in the area for the period 1961-1990 was 25-27 L/s/km<sup>2</sup> (atlas.nve.no). Dominating bedrock composition is gneisses and granites (Maijer and Padget, 1987; Bjerkgård and Nordgulen, 2002), with highly varying content of sulfides (Bjerkgård and Nordgulen, 2002; Frigstad 2009). Pyrrhotite with small inclusions of chalcopyrite is the most abundant sulfides together with pyrite (Hindar and Lydersen 1994).

The highway E18 runs in parallel with the coast-line, and the sulfidic rock deposits are close to sensitive marine water bodies (Fig. 1).

### *2.1. Sulfide-rock identification and deposits*

The sulfide content of the rock was determined in a field laboratory by CJV in order to decide if the rock pile should be treated as waste after blasting. Hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) was added to drilling dust and the temperature increase measured. A temperature increase above 1.2 °C was used as an empirical criterion for waste rock, as calibrations had shown a corresponding sulfide S-content larger than 1.25 %.

Sulfide-bearing waste rock material with an assumed sulfide S-content in the range 1-5%, was placed off-site in the three waste rock deposits M15/16, M17 and M20 (Fig. 1, Table 1). Mass volumes reported from CJV were converted to weight by the factor of 1.9 ton/m<sup>3</sup>. This factor takes into account that the deposits consist of grain sizes from dust to large stones and boulders of up to one meter in diameter.

Construction of the three deposits started in autumn of 2007 and was finished in 2009. Material from the blasting was transported directly to the deposit sites. Liming material was placed as horizontal layers in the dumps. The deposits were not covered during the construction, but with a thin cover of un-sorted moraine soil after they were finished. Shapes, drainage and water treatment are described below.



**Fig. 1.** The new Highway E18 (yellow line) with the three deposits indicated in upper panel (Map source: The Norwegian Mapping Authority). Detailed sketches in lower panel of deposits (large rectangles), treatment basins (triangles), M17 treatment area (small rectangle), and monitoring stations (open squares).

**Table 1**

Data for deposits and catchments. Catchment data for M20 are from before and after the stream was redirected in June 2009. Mass weight was calculated from deposit volume by using the factor 1.9 tons/m<sup>3</sup>.

|                                                                       | M15/16    | M17     | M20       |       |
|-----------------------------------------------------------------------|-----------|---------|-----------|-------|
| Area of deposit, m <sup>2</sup>                                       | 129 000   | 47 000  | 47 000    |       |
| Tot. volume of deposit, m <sup>3</sup>                                | 1 270 000 | 340 000 | 646 000   |       |
| Volume of waste rock                                                  | 1 270 000 | 340 000 | 540 000   |       |
| Volume/area ratio, m                                                  | 9.8       | 7.2     | 13.7      |       |
| Weight of waste rock, tons                                            | 2 362 200 | 632 400 | 1 004 400 |       |
| Shell sand in deposit, m <sup>3</sup>                                 | 8 300     | 4 500   | 7 500     |       |
| Slaked lime in deposit, m <sup>3</sup>                                |           |         | 600       |       |
| Shell sand in treatment area/basin, tons                              |           | 3 500   |           |       |
| Limestone gravel (1-10 cm)<br>in treatment area/basin, m <sup>3</sup> | 3 400     | 2 000   | 310       |       |
|                                                                       |           |         | before    | after |
| Catchment areas, km <sup>2</sup>                                      | M15/16    | M17     | M20       | M20   |
| For the deposit                                                       | 0.31      | 0.29    | 0.72      | 0.19  |
| For 200 m downstream of the deposit                                   |           | 0.45    | 0.74      | 0.21  |
| For the main monitoring station                                       | 0.71      | 0.89    | 0.80      | 0.27  |

Because of the pollution potential and the need to protect sea trout (*Salmo trutta* L.) populations, the disposal sites were located in the small valleys Stordalen (M15/M16), Mannfallaldalen (M17) and Urdalen (M20) (Fig. 1). The Stordalen stream enters the Kaldvell fjord 0.8 km downstream of the deposit. The stream in Mannfallaldalen follows the valley to Lake Lomtjenn, and enters the sea at Kosvik, 2.5 km downstream of the deposit. The stream from M20 enters the small lake Urevann immediately downstream of the deposit, runs into four other small lakes in a row, and reaches the ocean at Vestre Vallesverd, 5 km downstream of the deposit.

The M15/16 waste rock deposit is the largest of the three (Table 1). It was constructed on solid bedrock, and filled parts of the small valley. The liming material shell sand was added as described above, and a small sedimentation and neutralization dam was constructed downstream of the deposit (Fig. 1) to level out runoff and increase the effluent pH. Neutralization was by shell sand placed as a layer on the dam bottom. The deposit is probably unsaturated with respect to water. Oxygen, although not measured, is likely available throughout the deposit due to the large stones and boulders. The Stordalen stream was piped through the deposit, and the outlet of the dam was led directly to the open stream at the end of the pipe. No ditches for rainwater were constructed around or across the deposit.

The volume of M17 waste rock deposit is substantially smaller than the two other deposits. This deposit was also built directly on bedrock surface, is relatively shallow and probably also unsaturated. The draining system was made of open ditches along parts of the deposit and across the deposit. The construction allowed water from all these ditches to enter the deposit, however. Shell sand was used in the construction and placed as layers on the bottom of two treatment basins immediately downstream (Fig. 1; Table 1).

A relatively flat marshland area further downstream of M17 was also part of the treatment, and in July 2009 about 2500 tons of shell sand and 1-2 cm gravel was spread out over the marshland. The idea was that the stream should be dispersed and neutralized after passing the area. Heavy flow changed the surface structure of the area, however, and the water was concentrated in one main stream. Rapid flow also restricted the contact time with the liming material. The treatment was therefore far from optimal.

The M20 waste rock deposit was built in a former marshland after removal of large parts of the marshland material. Slaked lime ( $\text{Ca}(\text{OH})_2$ ) was mixed with the remaining parts of the marshland to stabilize the deposit base, and shell sand was added in layers in the deposit. The main stream through the small valley was ditched and led outside of the deposit. In June 2009, this stream was redirected directly to Lake Urevann to reduce the water load on the treatment basin. This deposit may be water-saturated and depleted of oxygen at the base, but the main part is probably unsaturated and oxygen-rich.

## 2.2. Water sampling and analysis

Water chemistry was part of the environmental monitoring program (EMP). Sampling frequency was weekly from July 2006 until March 2011. This program was run by the Norwegian Institute for Water Research (NIVA) in the period July 2006-April 2007 and thereafter by the water chemistry laboratory Vannlaboratoriet in Kristiansand, both on contracts with the construction company CJV.

The water chemistry monitoring station at M15/16 was 120 m downstream of the confluence of the open stream and the effluent of the treatment basin. At M17 monitoring was at the outlet of Lake Lomtjenn (lake area  $0.011 \text{ km}^2$ ), 700 m downstream from the deposit. At M20 water samples of the EMP were collected in the Lake Urevann inlet, 200 m downstream of the deposit and immediately downstream of the treatment basin. A pH monitoring station, run by NIVA, was set up downstream of the treatment basin. Heavy metals were monitored only at M15/16.

All analyses were obtained by standardized methods at NIVA and Vannlaboratoriet. Water samples were normally not filtered prior to analysis, but turbid samples were filtered prior to analysis of Al. Particulate heavy metal species can therefore not be distinguished from dissolved species.

Total Al and the two fractions, reactive (RAI) and non-labile (NLAI) Al, were measured. RAI consists of dissolved species of inorganic and organic Al. The NLAI is measured after passing the water sample through an Amberlite IR 120 cation-exchange column (Røgeberg and Henriksen, 1985). The labile Al (LAI), which has been reported to be toxic for brown trout (*Salmo trutta* L.) at concentrations above  $0.1 \text{ mg/L}$  (Rosseland and Henriksen, 1990), is calculated as the difference between RAI and NLAI.

Aluminum data presented here are mainly RAI (reactive Al), the dissolved fractions of inorganic and organic Al, at low concentrations ( $< 0.5\text{-}5 \text{ mg/L}$ ) and total Al at high concentrations ( $> 1\text{-}5 \text{ mg/L}$ ). At  $\text{pH} < 5.5$  and Al concentrations above  $1 \text{ mg/L}$  almost all Al is normally as inorganic monomeric Al, i.e. the labile fraction (LAI).

During work at a nearby tunnel, drilling water was pumped from Lake Urevann, and drained back to the lake. The effluent entered the stream from M20 upstream of the monitoring station. This effluent had elevated pH, but the pH was reduced at site to pH 7-8 by mixing of  $\text{CO}_2$  into the water. In periods of very low stream flow the effluent dominated the stream water chemistry at the monitoring station downstream.

Continuously measured pH at the monitoring site of M20 was by a temperature-compensated Amagross Rexef pH electrode and reference electrode. Water from the stream was circulated through an in-door measuring cuvette. Water level was also measured on a continuous basis at this station. The water level meter scale was anchored at the stream bottom at a site with steep natural slopes. As the scale was buried periodically with erosion particles from the construction site, the water-level could in periods with high flow rise to levels above the meter scale, causing a cut-off at about 35 cm in the hydrograph. Relative water-level data were used to indicate flow periods. The measurements were disturbed on occasion by settled particulate matter.



NIVA evaluated the water chemistry of the runoff from M17 and M20 more closely in summer 2009. Water samples were collected and analyzed at two sites; immediately and 200 m downstream of both deposits.

Precipitates and particles downstream of M17 was observed at several occasions and analyzed. The material was collected in clean plastic bottles, and sent to the NIVA laboratory for analysis of the main or selected chemical constituents. At the lab one of the following three procedures were followed:

1) white, fluffy precipitates downstream of the treatment area; about 10 ml of the precipitate including water was freeze dried, dissolved in HNO<sub>3</sub> and then measured by inductively coupled plasma atomic emission spectroscopy (Perkin-Elmer Optima 4300 DV ICP-AES spectrometer and Perkin-Elmer Autosampler AS 93 plus).

2) grey, fluffy precipitate that settled in the treatment basin; the particles were filtered through 0.45 µm Millipore filter and rinsed with distilled water, then dried and analyzed as outlined above.

3) turbid stream water; raw and filtered water were analyzed for total and dissolved constituents with standardized methods.

### *2.3. Catchments and hydrology*

Catchment size upstream of the monitoring stations at all three sites was determined in order to calculate mass transport (Table 1). The size of the catchment area upstream of the monitoring station at M20 was unaltered until June 2009. At that time the upper two thirds of the catchment was cut off by diversion of the stream immediately upstream M20 directly to Lake Urevann through a blasted ditch. The flow at the monitoring station was correspondingly reduced.

Daily runoff data from the gauged weir at the Birkenes research site (Wright 2008) about 18 km north of the deposits were used for mass transport calculations after corrections for catchment area and differences in specific runoff. Measured daily runoff at Birkenes (catchment area of 0.41 km<sup>2</sup>) was adjusted to the catchment areas of the monitoring sites, e.g. for M15/16 (catchment area 0.71 km<sup>2</sup>, Table 1) by the factor 0.71/0.41. According to the database REGINE (Norwegian Water Resources and Energy Directorate) mean specific discharge for the period 1961-1990 differed between the Birkenes research site area (34 L/s/km<sup>2</sup>) and the three other catchments (26.6., 26, and 27 L/s/km<sup>2</sup> for M15/16, M17 and M20, respectively). Measured daily runoff was corrected for these differences, e.g. at M15/16 by the factor 26.6/34.

The redirection of the uppermost catchment of M20 was accounted for in the calculations by subtracting this catchment from the total catchment.

### *2.4. S release rates*

The procedure for calculations of S release rates was as follows:

First, S transport for each period between sampling occasions was calculated by multiplying the total estimated runoff for the period with the net mean concentration of sulfur from sulfate concentrations. Net concentrations were calculated by subtracting the mean background concentration for the pre-construction period from the measured concentrations. As the transport calculations were done for the whole dataset, the first, unaffected period appears with about zero net S transport.

Then, accumulated S transport (as metric tons of S) for the whole period was plotted along the time scale. The plotted curves were used for description of patterns and calculation of mean S

transport for specific periods. Mean S transport was determined by linear regression analysis for periods with stable increase in accumulated S transport.

### 3. Results and Discussion

#### 3.1. Major ions

Waters of undisturbed catchments in the Lillesand area are very poor in base cations due to the slow weathering of the dominating soil minerals. The water chemistry prior to the onset of construction of waste rock deposits is representative for the situation in the area. The mean pH at the three sites was 5.15 - 5.93 (Table 2) and pH could be as low as 4.9 (Fig. 2), mainly due to low inherent buffering capacity, dissolved organic acids (mean TOC of 7-10 mg/L, Table 2) and acid rain effects in the area (Wright 2008). Pre-construction  $\text{SO}_4$  concentration of 4-7 mg/L indicates contribution of acidity from sulfidic rock weathering of the relatively unaltered catchment of M17 and from minor excavations in the other two. The Al concentrations at all sites were moderate (200-300  $\mu\text{g}$  RAl/L), and LAI was close to and above the critical limit for brown trout of 100  $\mu\text{g}$ /L (Rosseland and Henriksen, 1990) at M17 and M20, respectively. Relatively high concentrations of Na and Cl reflect influence of sea-salts derived from atmospheric deposition.

**Table 2**

Pre-construction water chemistry at the stream monitoring stations for the three deposits.

| Monitoring at deposit: | pH   | ALK<br>mmol/L | $\text{SO}_4$<br>mg/L | Ca<br>mg/L | RAI<br>$\mu\text{g}$ /L | NLAI<br>$\mu\text{g}$ /L | LAI<br>$\mu\text{g}$ /L | $\text{NO}_3\text{-N}$<br>$\mu\text{g}$ /L | $\text{NH}_4\text{-N}$<br>$\mu\text{g}$ /L | Cl<br>mg/L | Na<br>mg/L | TOC<br>mg/L |
|------------------------|------|---------------|-----------------------|------------|-------------------------|--------------------------|-------------------------|--------------------------------------------|--------------------------------------------|------------|------------|-------------|
| M15/16 (n=12)          |      |               |                       |            |                         |                          |                         |                                            |                                            |            |            |             |
| Mean                   | 5.93 | 0.12          | 7.28                  | 3.18       | 232                     | 178                      | 54                      | 107                                        | 75                                         | 11.3       | 7.4        | 10.3        |
| St.dev.                | 0.62 | 0.06          | 1.65                  | 1.02       | 68                      | 42                       | 37                      | 130                                        | 71                                         | 1.8        | 1.6        | 1.8         |
| M17 (n=21)             |      |               |                       |            |                         |                          |                         |                                            |                                            |            |            |             |
| Mean                   | 5.53 | 0.06          | 4.36                  | 1.66       | 209                     | 138                      | 80                      | 67                                         | 20                                         | 12.4       | 7.2        | 7.2         |
| St.dev.                | 0.52 | 0.02          | 0.63                  | 0.18       | 80                      | 29                       | 78                      | 82                                         | 22                                         | 1.9        | 1.4        | 1.8         |
| M20 (n=8)              |      |               |                       |            |                         |                          |                         |                                            |                                            |            |            |             |
| Mean                   | 5.15 | 0.04          | 6.10                  | 0.86       | 299                     | 160                      | 140                     | 4                                          | 6                                          | 7.7        | 5.2        | 9.7         |
| St.dev.                | 0.24 | 0.02          | 1.86                  | 0.16       | 106                     | 131                      | 135                     | 5                                          | 3                                          | 0.8        | 0.2        | 8.2         |

Blasting and oxidation of sulfide-containing bedrock inevitably results in production of sulfuric acid if no base is added. This had occurred in the examined area on several occasions prior to construction of the new highway E18 line, and resulted in massive fish kills of brown trout (*Salmo trutta* L.) in receiving surface waters (Hindar and Lydersen, 1994; Hindar and Iversen, 2006).

After construction of the deposits there was a marked increase in  $\text{SO}_4$  concentrations downstream of all three deposits relative to pre-construction values (Fig. 2, Table 2). An increase to above 10 mg/L started in March 2007, July 2007 and September 2006 at M15/16, M17 and M20, respectively. Relatively high concentrations (> 100 mg/L) were reached after 4, 12 and 9 months, respectively. At M15/16 the initial increase could be related to blasting for the road line rather than deposit construction. A rapid increase to a new level between 300 and 800 mg/L was observed after 14 months (in May 2008). At this time the construction of the deposit was finished.

A rapid  $\text{SO}_4$  concentration increase from 50-150 mg/L and up to about 400 mg/L at M20 took place in summer 2009, and the concentrations stayed at that level. The main reason for this increase is the re-direction of flow upstream of M20, which reduced the catchment area at the monitoring station by two thirds, resulting in a corresponding increase in concentrations.

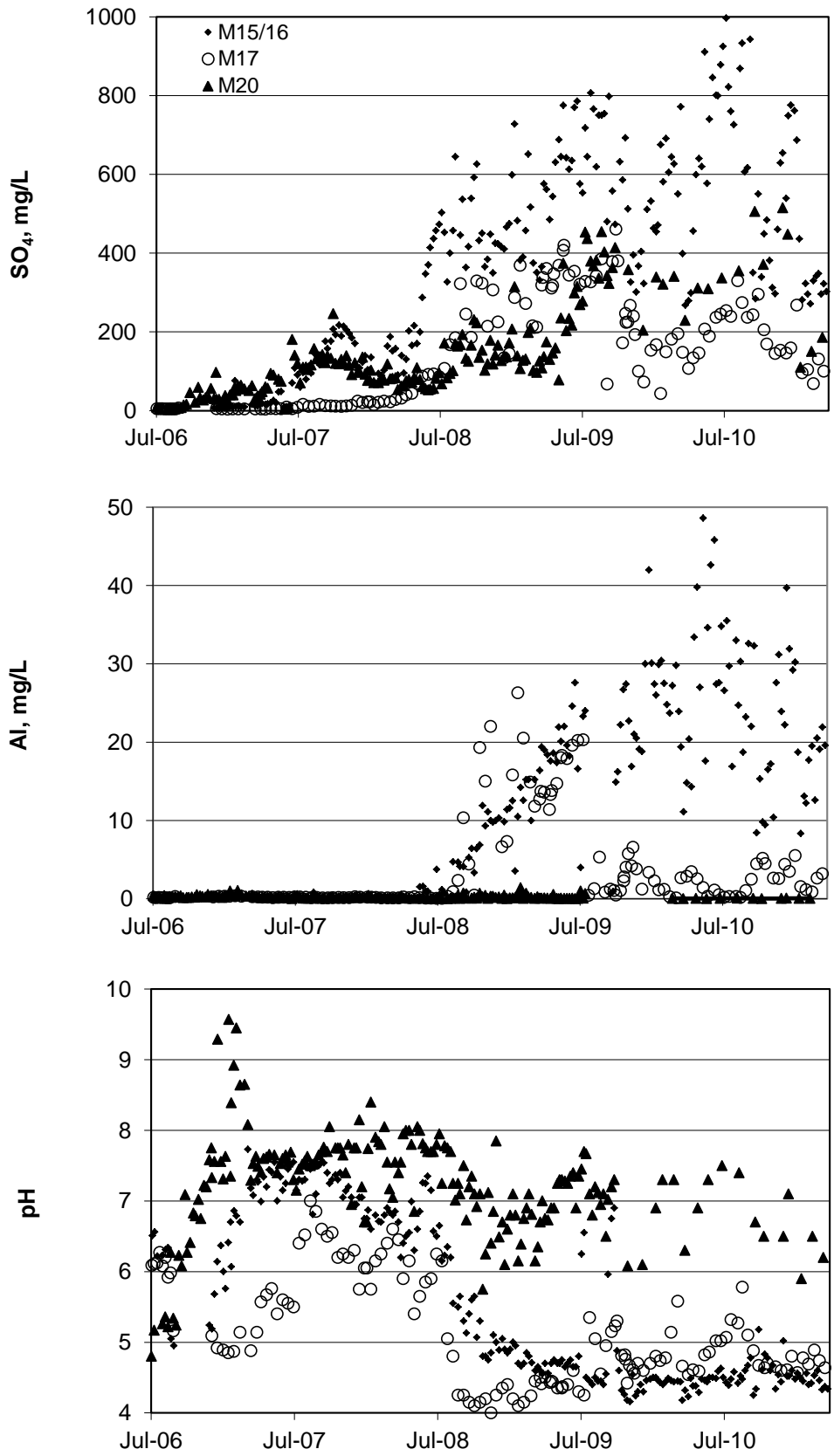
The increases in  $\text{SO}_4$  concentrations were accompanied by substantial reductions in pH at M17 and after a short time also downstream of M15/16. The pH values at M20 stayed above pH 6.0 at low to moderate flow, but dropped to below 6.0 on heavy rain episodes, indicated by increases in runoff (Fig. 3).

The Al concentrations started to increase to high levels ( $> 1$  mg/L) at the same time as pH decreased to below about 4.8 at M17 and below 5.5 at M15/16 (Fig. 2). At M15/16, Al concentrations were 10-50 mg/L from January 2009. At the outlet of Lake Lomtjenn downstream of M17 Al concentrations were 10-20 mg/L from autumn of 2008 until summer 2009. From summer 2009 and until 2011 Al concentrations were markedly lower (0.2-5.5 mg/L). The critical limit of Al for brown trout referred to above was thus exceeded.

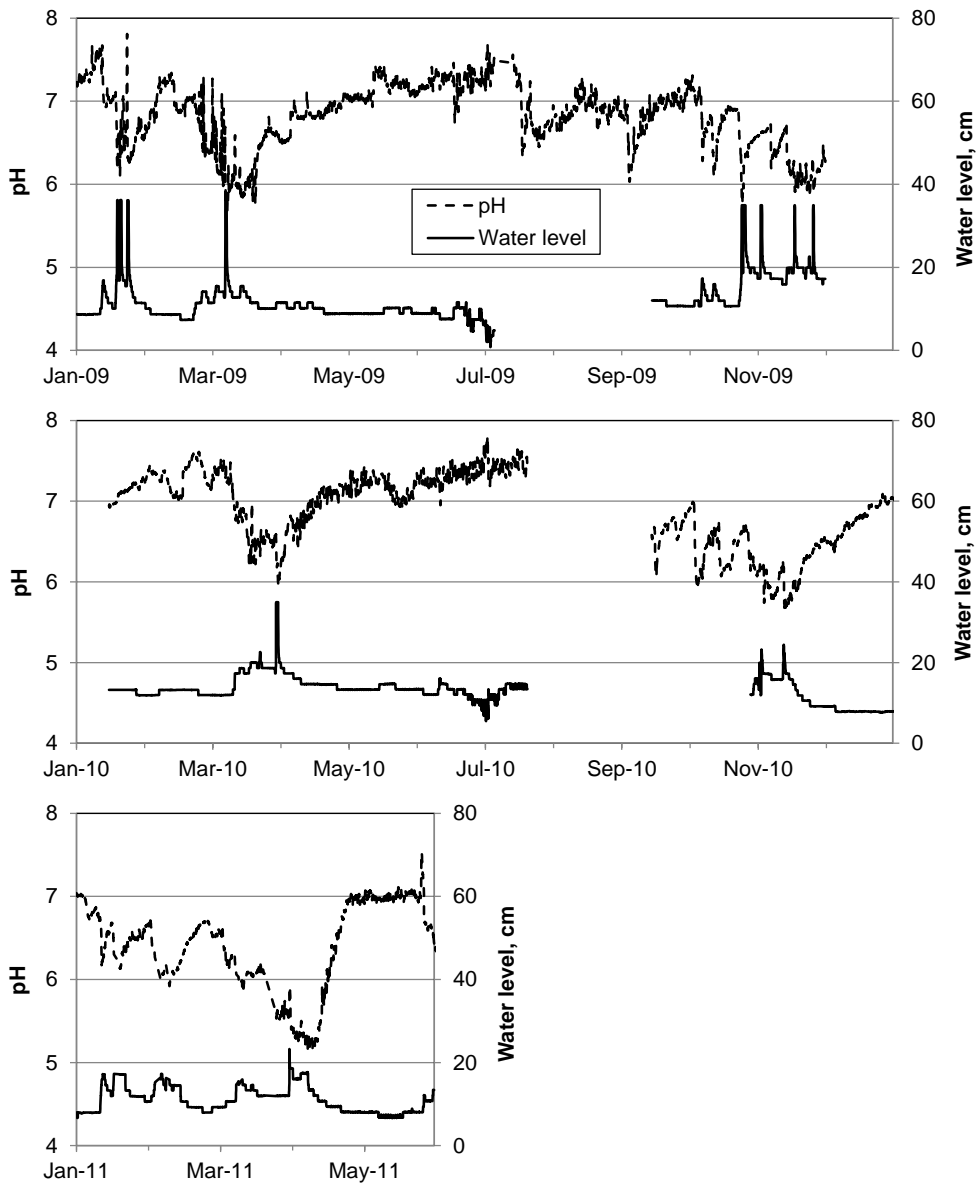
At M20 few water samples with pH below 6 exist from the construction period, and Al in the labile fraction was low ( $< 0.1$  mg/L) also for these few samples, except for the April 2011 sample (1 mg/L as LAI). Sampling during the pH decreases at flow events documented by the continuous water-level monitoring in the stream (Fig. 3) would probably have resulted in more data showing high Al concentrations. Mobilization of Al during such episodes, typically with pH around 5.5, may explain why a white, fluffy precipitate had formed in the stream during winter 2010/spring 2011 (see below). The mobilizing effect on Al when  $\text{SO}_4$  concentration increased and pH dropped below 5.5-6 is also seen in Fig. 4. Significantly higher Al/ $\text{SO}_4$  weight ratios were found below pH 6 than above. Concentrations of Al are very sensitive to pH and every increase or decrease in Al seen in Fig. 2 is correlated inversely with a change in pH. A corresponding decrease in buffering effects of the added neutralizing agents, reflected by the Ca/ $\text{SO}_4$  ratio (Fig. 4), was observed at low pH.

Excursions of decreased pH at M20 are related to flushing events as seen by the relation between pH and water level for M20 in Fig. 3. Acidity is continuously generated within the rock pile and stored to some extent, later to be flushed out with a major rain event. This phenomenon is similar to the "first flush" seen at mined sites (Nordstrom, 2011b). Repeated flow situations resulted in increasingly lower pH downstream of M20, strongly indicating lowered de-acidification capacity over time. Rainwater follows preferential flow paths through deposits, and added liming material will gradually be eroded along these paths. Deactivation of the shell sand surfaces by precipitates will reduce the neutralizing effect further. It is suggested here that reduced effect of the added liming material at M20 also will result in increased S release rates, and thus change the water quality further towards what has been documented here for the two other deposits.

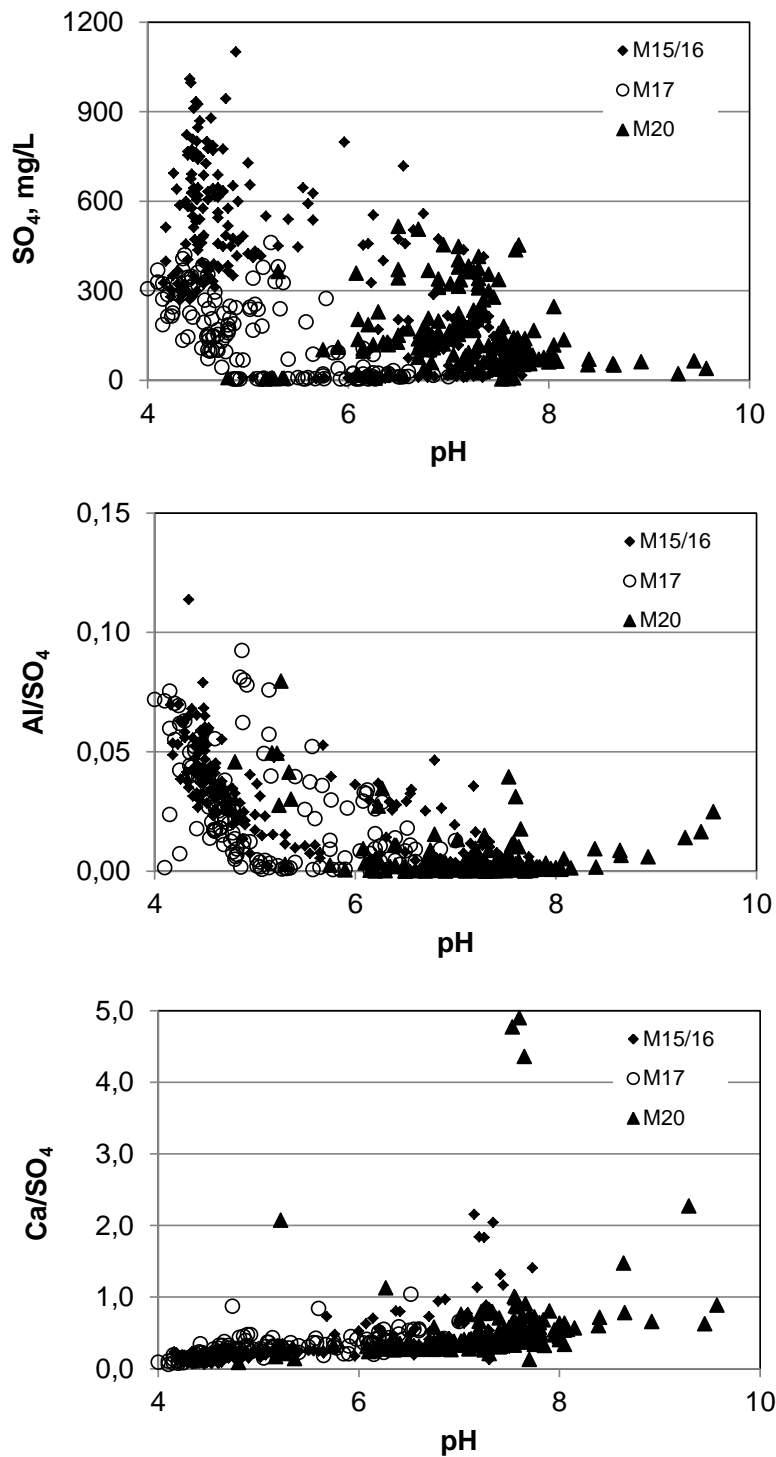
As the climate of this area changes in the direction of drier summers and more intensive rainfalls, one might expect periods with increasing toxic potential, such as documented after a long dry period at the nearby River Moelva in 2006 (Hindar and Iversen, 2006). Here, accumulated weathering products at a construction site were flushed out during a rain event after drought, which destroyed the fish populations of the river. Climate change is thought to be an additional factor that might affect the long-term acidification potential of mining sites (Nordstrom, 2009).



**Fig. 2.** Stream concentrations of SO<sub>4</sub> and Al, and pH at the monitoring stations of the three deposits. Al was not measured at M20 in the period August 2009- medio February 2010.



**Fig. 3.** Data from the continuous measurement of stream pH (upper lines) and water level (lower lines) at deposit M20 for the period 2009-2011. The measuring device for water level had a cutoff at 35 cm, see text, which means that some of the water level spikes were actually higher than 35 cm.



**Fig. 4.** Plots of SO<sub>4</sub> concentrations against pH (upper) together with the Al/SO<sub>4</sub> (middle) and Ca/SO<sub>4</sub> (lower) ratios (by weight) against pH in runoff from the three deposits.

### 3.2. Trace metal behavior

Even though pH in the Stordalen stream was as low as 5.0 prior to the construction of M15/16, heavy metal concentrations were low (Table 3). As soon as the construction started, concentrations

of Cd, Co, Cu, Mn, Ni and Zn increased, but as long as sulfate concentrations were below about 300 mg/L and pH above 6.0 concentrations stayed at relatively moderate levels. At the time when the deposit was finished in summer 2008, sulfate concentrations increased to above 300 mg/L, pH decreased to below 5.5, and trace metal concentrations increased to very high levels (Table 3, Fig. 5). The increase relative to pre-construction levels was 25 - 400 times, highest for Ni and then in decreasing order for Co, Mn, Cd, Zn, Cu. There were no substantial changes in As, Cr and Fe.

**Table 3**

Mean pre-construction (second half of 2006) and post-construction (from August 2008) metal concentrations in the Stordalen stream downstream of M15/16. Standard deviations are given.

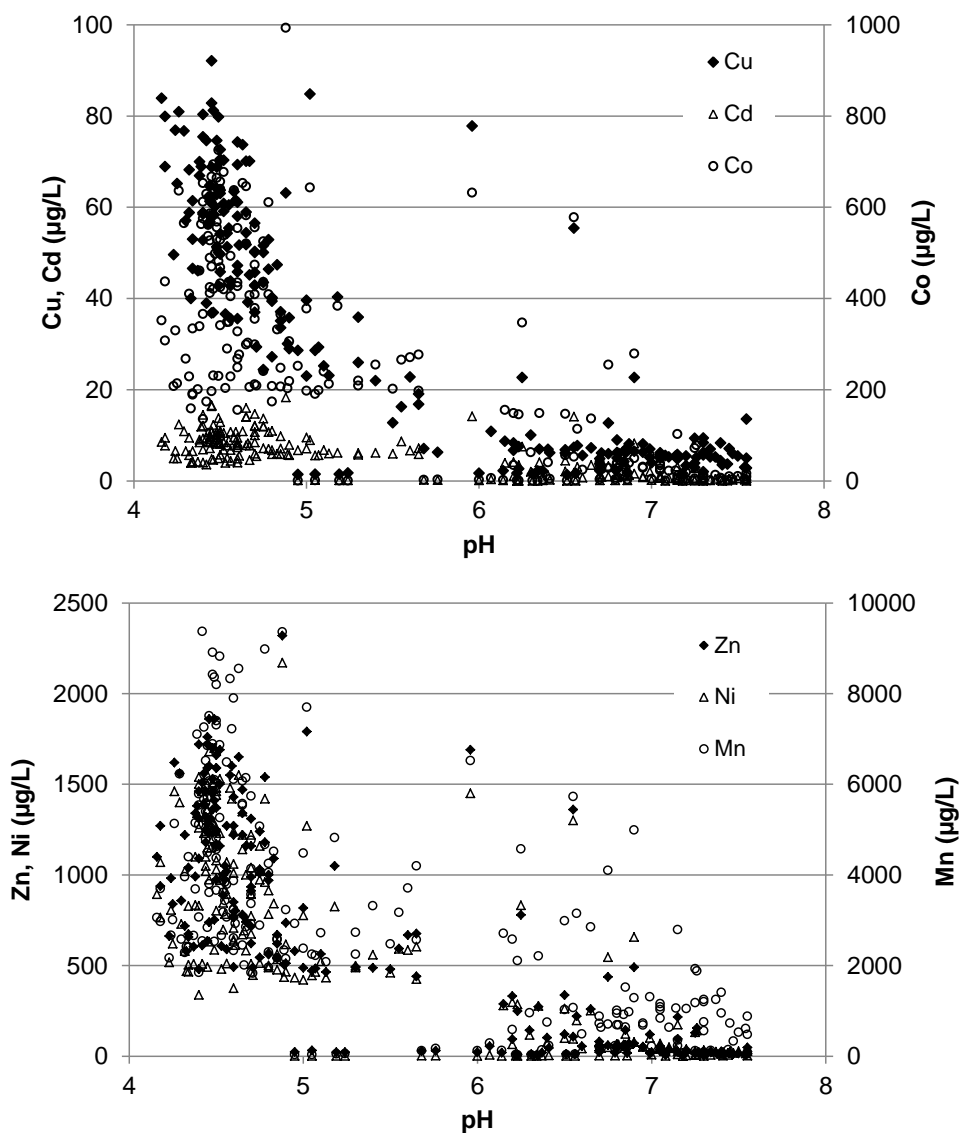
|                    | Pre-construction |         | Post-construction |         |
|--------------------|------------------|---------|-------------------|---------|
|                    | Mean             | St.dev. | Mean              | St.dev. |
| As $\mu\text{g/L}$ | 0.82             | 0.21    | 0.72              | 0.68    |
| Cd $\mu\text{g/L}$ | 0.121            | 0.072   | 8.5               | 3.1     |
| Co $\mu\text{g/L}$ | 1.49             | 0.66    | 401               | 169     |
| Cr $\mu\text{g/L}$ | 0.82             | 0.43    | 0.97              | 0.68    |
| Cu $\mu\text{g/L}$ | 2.2              | 1.5     | 52                | 18      |
| Fe $\mu\text{g/L}$ | 1351             | 683     | 712               | 760     |
| Mn $\mu\text{g/L}$ | 58               | 42      | 4472              | 1963    |
| Ni $\mu\text{g/L}$ | 2.3              | 0.4     | 924               | 383     |
| Zn $\mu\text{g/L}$ | 18.4             | 7.5     | 1054              | 422     |

The dataset reflects the sources and processes that give rise to this water chemistry. The relation between Zn and Cd for runoff from M15/16, shown in Fig. 6, is positively correlated but with considerable dispersion. The reason for some of this dispersion can be seen with reference to Fig. 7a where the Cd/Zn ratio is plotted over time for M15/16 runoff. Between mid-winter of 2006-2007 and July of 2008 the pH values for M15/16 varied between 5 and 7.5 because of the neutralization effect of added shell sand. Shortly following the pH increase, the Cd/Zn ratio more than quadrupled. Such an effect could be explained by selective and competitive adsorption of Zn by hydrous ferric oxides or by carbonates. With Zn concentrations about 2 orders of magnitude higher than Cd concentrations, the Zn would tend to swamp reactive surface sites relative to Cd and the resultant Cd/Zn in solution would increase. At high sulfate concentration and low pH the Cd/Zn ratio (Fig. 7b,c) can be seen to converge to a fairly constant value of about 0.007, a common value for the mineral sphalerite, as shown below. The same convergence is seen in the time series data of Fig. 7a after July, 2008.

Indeed, sphalerite (ZnS) has been the main ore mineral for Cd production. Sphalerite is the most common source of Zn in sulfidic crystalline rocks. This mineral nearly always contains Cd because of the similarity in ionic radius and the ZnS crystal structure is the same as that for CdS which allows for easy substitution of Cd for Zn. Most sphalerites have Cd concentrations in the range of 0.1-0.5 wt.% (Fleischer, 1955; Barrie et al., 2005; de Oliveira et al., 2011), Zn concentration in the range of 58-63 wt.% (de Oliveira et al., 2011; Hammarstrom et al., 2005), and thus a common Cd/Zn weight ratio in the range 0.0016-0.0086. Under acidic conditions, the rates of release of Cd and Zn from sphalerite dissolution are essentially the same (Stanton et al., 2008) and there are no mechanisms for attenuation such as sorption or mineral precipitation under low-pH, oxidizing conditions. Hence, the ratio of Cd/Zn in solution reflects that of the mineral.

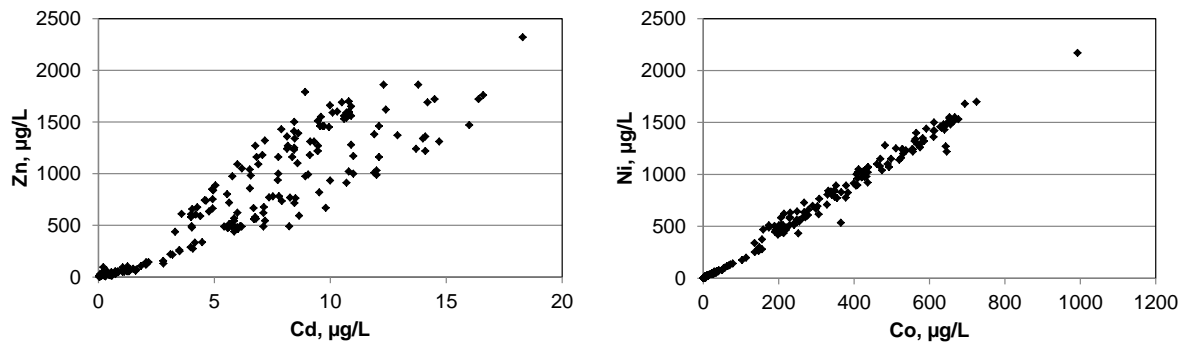
Another correlation that clearly shows up is that for Co and Ni (Fig. 6). The strong linear correlation suggests that both elements are derived from dissolution of the same mineral. It has long

been known that  $\text{CoS}_2$  and  $\text{NiS}_2$  have the same crystal structure as pyrite and the solid substitution of Co and Ni in pyrite is common and covers a wide range, usually with  $\text{Co} > \text{Ni}$  but with many exceptions (Fleischer, 1955). Lab studies on the leachability of Co and Ni are not known to the authors but in a study on natural acid rock drainage in New Mexico a similar strong linear correlation was found between Co and Ni with a ratio of 0.42 (Nordstrom, 2008). Further, it was possible to show that the Co/Ni ratio in these acid waters was similar to the Co/Ni ratio in the pyrite. Similarly, Moncur et al. (2009) were able to show that the main source of mobile Co and Ni were derived from oxidizing pyrite. Consequently, it seems most likely that the elevated Co and Ni concentrations in the M15/16 runoff are derived from pyrite oxidation. As shown with Cd/Zn ratios, Co/Ni ratios converge to a constant value after July, 2008 of about 0.44 (Fig. 7d). Similarly, the same convergence is seen with high sulfate concentration (Fig. 7e) and with low pH (Fig. 7f). The close similarity of this ratio with that in the New Mexico study supports the contention that pyrite is the source of Co and Ni.



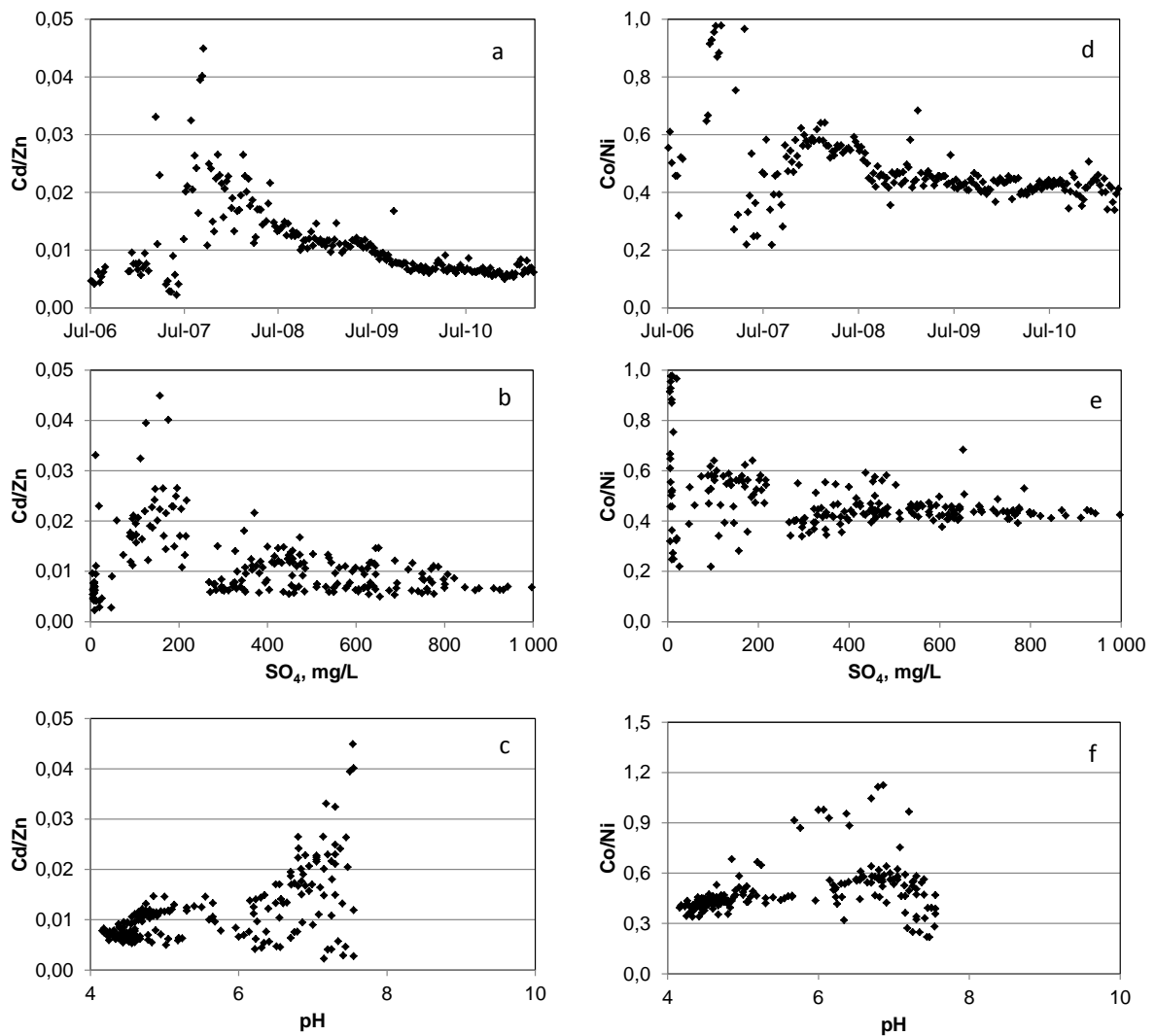
**Fig. 5.** The trace metals Cu, Cd, Co, Zn, Ni and Mn against pH for runoff at deposit M15/16.





**Fig. 6.** Plots of Zn against Cd (left) and Ni against Co (right) for runoff at deposit M15/16.

Concentrations of As, Cr and Fe downstream of M15/16 were unchanged relative to background levels during the monitoring period. Part of this (unchanged As and Cr) may be due to insignificant sources in the waste rock. Precipitation of  $\text{Fe}^{3+}$  from sulfide-oxidation as Fe oxy-hydroxides at the source and co-precipitation of As and Cr may have also attenuated these metals.



**Fig. 7.** The ratios Cd/Zn and Co/Ni plotted against time,  $\text{SO}_4$  concentration and pH for runoff at deposit M15/16.

### 3.3. Deposit S release rates

Estimates of metal and sulfur flux from mining activities are severely hindered by lack of sufficient data (Nordstrom, 2011a). This study is based on an unusually detailed dataset, with high frequency of EMP-data, exact delimitation of deposits and catchments, and available pre-construction data from the sulfide-containing deposit areas. The soil-cover is sparse, and loss of water to groundwater is probably negligible. These conditions ensure rapid responses to rain events with sufficient access to oxygen and rain/moisture, and rapid transport of weathering products from the deposits.

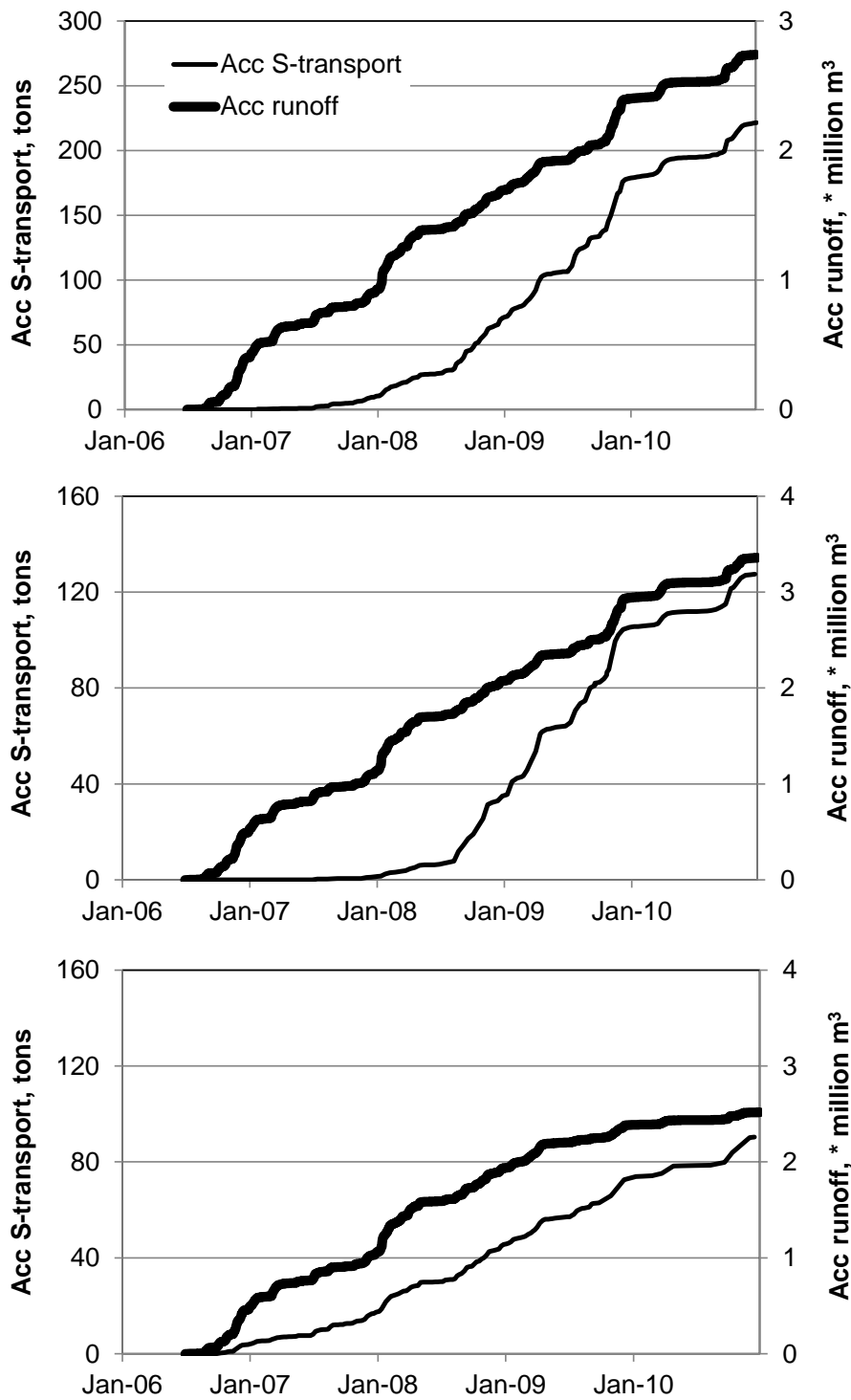
To get an idea of the size and pattern of sulfur release rates from the waste rock deposits, sulfate concentrations at the EMP monitoring stations and daily runoff data were used to calculate net S transport from the deposits.

The increase in net S transport started at the same time as the construction activities, and after a short, initial phase came a period of a remarkably stable S transport at all three deposits (Fig. 8). The initial phase coincided with the construction phase.

Patterns of the S transport follow runoff patterns, as can be seen from the similar forms of the accumulated runoff curve and S transport curve for all sites (Fig. 8). The redirection of the stream through M20, indicated by the flattening of the accumulated runoff curve in 2009-2010, had no apparent effect on the S transport. Long, dry periods, as the warm summer of 2010, delayed the S transport, seen as a parallel displacement of the steady increase in accumulated S transport for M17, but probably did not affect the weathering rate following such periods. The linear increase in accumulated S transport clearly indicates constant release rates and thereby constant weathering of the sulfides over the last part of the monitoring period.

The S transport after the initial phase was 69-225 kg S/day, corresponding to annual S transport of 25-82 tons for the three deposits (Table 4). The mean S-content of the waste rock could, for obvious reasons, not be measured. A content of 2 % S by weight might be a conservative estimate, as the limit for disposal was 1.25 % S. If based on 2 % S, the annual S transport corresponds to 0.12-0.40 % of the total amount of S in the deposits (Table 4). If the S-content of the waste rock was 4 % by weight, these percentages would be halved. By inverting percent mass per year we have the years it would take to deplete all the sulfides at current weathering rates. It would take 250 years to deplete the rock sulfide at M17 and 800 years at M20, with M15/16 in between (580 years). As the release rates are expected to be non-linear over such a long time period, these estimates should be regarded as indications of the time frame for acid runoff.

The estimated S transport gives release rates of 1.4-4.6 mg SO<sub>4</sub>/kg rock/week (Table 4). When comparing this number with release rates from other sources one should remember that the waste rock in the deposits consist of all particle sizes from boulders to dust. The surface area per weight is thus magnitudes lower than in a sample of drilling dust.



**Fig. 8.** Accumulated net S transport (tons) and accumulated runoff (million m<sup>3</sup>) from deposits M15/16 (top), M17 (middle) and M20 (lower). Note different scales.

**Table 4**

Sulfur transport and release rates.

|                                                      | M15/16 | M17   | M20   |
|------------------------------------------------------|--------|-------|-------|
| Sulfur in deposit (2 % S by weight), tons            | 47200  | 12650 | 20100 |
| S-transport from deposit, tons/yr                    | 82     | 50    | 25    |
| S-transport from deposit, % of mass S/yr             | 0.17   | 0.40  | 0.12  |
| Release period, yrs                                  | 580    | 250   | 800   |
| Release rate, mg SO <sub>4</sub> /kg waste rock/week | 2      | 4.6   | 1.4   |

Weathering of exposed sulfide-bearing rock will last for centuries. Data from this work and from elsewhere, e.g. from Aitik, a Swedish Cu mine (Stromberg and Banwart, 1999a,b), indicate almost constant oxidation rates during the period of measurement. An initial lag phase due to establishment of Fe and S oxidizing bacterial populations has been documented (Kuipers et al., 2006). The lag phase is consistent with the low oxidation rate resulting from abiotic processes as compared to bacteria-supported oxidation (Nordstrom and Alpers, 1999). Small particles will contribute relatively more than large particles to oxidation and acid production. One might therefore also anticipate high initial S release rates as these particles are easily oxidized and broken down.

Decreasing effects of weathering of calcitic and other buffering minerals of the rock itself may delay the timing of net acid production (Stromberg and Banwart, 1999a). In the Lillesand area no lag-time related to this calcite-effect is possible, as about 50 % of the sulfate immediately after blasting was balanced by Al and H<sup>+</sup> ions, rather than Ca and Mg.

If the whole monitoring period presented here is part of a lag-phase, a non-linear accumulated S transport pattern should have been seen over time. That was not the case. A short lag-time during the construction period was found, however.

Redirection of runoff upstream of M20 and substantially reduced flow through the deposit did not affect the constant release rate. This stream passed by the deposit at the base without much contact with sulfides or neutralizing material. Redirection was thereby of little significance for the processes in the deposit, but might have been important for neutralizing countermeasures downstream by reducing the water load to the sedimentation basin.

The S release rates of 1.4-4.6 mg SO<sub>4</sub>/kg rock/week are in the same order of magnitude as the 3-91 mg SO<sub>4</sub>/kg rock/week found by Hollings et al. (2001) for small particles (up to 25 mm) and the release rate of 4.8 mg SO<sub>4</sub>/kg rock/week found by Wagner et al. (2006) for low-sulfuric (<0.5 % sulfide) material. However, the S release rates, corresponding to 0.15-0.47 · 10<sup>-9</sup> mequiv/kg rock/s, are lower than the acidity flux from column experiments (0.93 · 10<sup>-9</sup> mequiv/kg rock/s) and more than one order of magnitude lower than the acidity flux at the waste rock site (5 · 10<sup>-9</sup> mequiv/kg rock/s) of the Aitik mine (Stromberg and Banwart, 1999a). Such figures are encumbered with large differences in specific surface area, sulfide-mineral composition, particle sizes of the studied material, ambient air temperature, microbiology and oxygen content inside the sulfide-containing material. Many of these important regulating factors are often not accounted for, and comparison is therefore difficult. The exothermic reaction of pyrite oxidation may also result in a significant temperature increase in poorly aerated material of high sulfide-content, thus increasing the oxidation rate significantly. No such temperature effects were noted in the present study.

If we assume constant S transport at the rates found here (0.1-0.4 % of deposit S/year) these waste rock deposits will gradually be broken down and probably change their shapes. A 10% reduction in mass weight may be the result after 40-100 years.

The environmental problems already associated with the waste rock deposits will last for many decades if not resolved by adequate countermeasures. Estimates here and for other waste rock dumps indicate time periods for S-leaching from a few hundred up to many thousand years (e.g. Linklater et al., 2005). Significantly reduced oxidation rates may probably not occur in the near future if the deposits stay exposed to water and oxygen. But as the sulfides are gradually depleted in the deposit, lower sulfate-concentrations and thereby reduced acidification may be expected.

Data from Hindar and Lydersen (1994) and the EMP for the E18 project for Lake Langedalstjenn, close to M15/16, indicate significantly reduced  $\text{SO}_4$  and Al concentrations over 15 years. The reduced concentrations coincided with asphalt and roof capping over much of the blasted material. Capping certainly has reduced the oxidation rates of the sulfide material.

Annual S release rates may characterize the construction of the three deposits. The rate was about three times higher at M17 than at M20, with M15/16 in between of the two. There is no reason to believe that the waste rock of M17 is especially rich in sulfide content, and none of the deposits are covered as recommended by Fröberg and Höglund (2004), which make them open for incoming water and oxygen. The reason for the large difference between M17 and M20 may be due several features of the deposits.

The volume/area ratio for M17 is only half of that for M20. The relatively steep slopes of M20 may have favored a more rapid runoff of incoming rainwater and possibly less access to oxygen for the innermost parts of the deposit. The base of M20 fills up a former marshland area, and some of the organic material may have been lifted upwards during construction and mixed with the deposit. Oxygen consumption from decomposition of this organic pool may have reduced the sulfide oxidation. As slaked lime was mixed into the residual marshland masses, this hydroxide may have been relatively important for increasing the neutralizing processes at the base.

Draining of water from the surrounding terrain was by open and highly permeable ditches. At M17 rainwater, diffuse runoff from the surrounding terrain and water that was captured in shallow and non-continuous ditches on the side of the deposit were allowed to enter the deposit. Diffuse runoff that was directed into a ditch across the deposit infiltrated back to the fill due to the highly permeable character of the ditch. At M15/16 and M20 runoff from the terrain was more successfully led around the deposit masses due to the placement of the deposit on dry land and the more favorable shape (volume/area ratio; Table 1). At M15/16 the small stream at the base of the deposit was led through a pipe and the stream-water had no contact with the waste rock material.

### *3.4. Al-precipitates*

Dilution and mixing of acid waters with waters of higher pH result in on-going precipitation downstream of the deposits. If the mixed water reaches a pH of about 4.5 or higher, the dissolved Al can hydrolyze and precipitate because the first hydrolysis constant for Al is  $\text{p}K_1 = 5.0$  (Nordstrom and May, 1996). The effect of pH on Al precipitation can be easily seen in Fig. 4 in which Al/ $\text{SO}_4$  weight ratios have been plotted against pH. For pH values below 4.5 the ratios are consistently high and above pH 4.5 the ratios are much lower.

Precipitates were found at all three sites on several occasions. The white precipitate at M17 was dominated by aluminum and sulfur (Table 5), and the molar ratio for Al/S is slightly above 5. Because there is no mineral with this ratio, it was probably composed of a mix of some amorphous aluminum hydroxide ( $\text{Al}(\text{OH})_3$ ) and amorphous aluminum hydroxysulfate, probably hydrobasaluminite ( $\text{Al}_4(\text{SO}_4)(\text{OH})_{10} \cdot 12-36 \text{H}_2\text{O}$ ). Hydrobasaluminite is a white, fluffy and clay-like mineral with Al/S molar

ratio of 4, which dehydrates to felsöbányaite (earlier called basaluminite;  $\text{Al}_4(\text{SO}_4)(\text{OH})_{10}\cdot 5\text{H}_2\text{O}$ ) if settled on surfaces that dry up. The minerals are known to form at similar sites elsewhere where acid, Al-rich and non-acid waters mix (Nordstrom and Ball, 1986; Nordstrom and Alpers, 1999; Bigham and Nordstrom, 2000), e.g. as at the site called Paradise Portal in the San Juan Mountains, CO, USA (Nordstrom, 2011a).

**Table 5**

Composition of particles at deposit M17, analyzed on freeze dried samples (May and November samples) and on water samples (October; obtained as the difference in concentrations between total and dissolved fractions).

|                                                              | Date      | Al     | Ca    | Fe    | K    | Mg   | Na  | S     | Si   |
|--------------------------------------------------------------|-----------|--------|-------|-------|------|------|-----|-------|------|
| White particles, $\mu\text{g/g}$                             | 18-May-09 | 217626 | 8410  | 11986 | 1208 | 2105 | 425 | 50137 | 4790 |
| Grey particles from dam, $\mu\text{g/g}$                     | 17-Nov-09 | 79900  | 24900 | 38200 | 7920 | 8720 | 565 | 9250  | 3720 |
| Particles in runoff at M17, $\mu\text{g/L}$                  | 24-Oct-09 | 35440  | 27    | 2607  |      |      |     | 18    |      |
| Particles in runoff 200 m downstream of M17, $\mu\text{g/L}$ | 24-Oct-09 | 29610  | 10    | 9388  |      |      |     | 12    |      |

On October 24, 2009 the runoff from M17 was grey and very turbid, with concentrations of total solids of 221 mg/L and 295 mg/L at the deposit and 200 downstream, respectively. Erosion of shell sand was seen and shell sand obviously affected the water chemistry resulting in pH of 6.88 and 6.49, respectively. Each sample was analyzed for dissolved and total fractions of Al, Ca, Fe and S and the concentrations of the particles were found by difference (Table 5). These particles consisted mainly of precipitated Al and Fe, probably as amorphous Al hydroxides and Fe oxyhydroxides, whereas Ca and S were almost entirely as dissolved ions. The flow may have mobilized already precipitated metal hydroxides at the disposal site.

On November 17, 2009 the limestone gravel in the lower treatment basin at M17 was covered by a grey, fluffy precipitate, which could be made either by sedimentation of particles from the deposit or precipitation directly on the limestone surfaces. The particles were collected by a bottle after careful disturbances of the gravel and concentrated by repeated grabbing and sedimentation of the material in the bottle. In the laboratory the particles were filtered and analyzed for metals and organic carbon. The inorganic part was dominated by Al (Table 5). Ten times more Al than S on a molar basis indicates precipitation of mainly Al hydroxides rather than Al-hydroxysulfate. The concentrations of Fe and Ca were relatively high, indicating both iron precipitation as iron hydroxide and presence of  $\text{CaCO}_3$ . The concentration of total organic C was 57 mg/g, about 70 % of the Al by weight. We interpret this as co-precipitation of dissolved organic matter from the surrounding terrain on the Al and Fe precipitate rather than an organic complex with Al and Fe. The relatively large contribution by organic matter may explain the grey color.

The high concentrations of inorganic monomeric Al at the outlet of Lomtjenn indicate that the precipitation was far from complete. A mix of ongoing precipitation and received precipitates from the inlet stream made the lake turbid with a greenish appearance. The color might be due to very small particles (clay size) or colloids at an early phase of in-lake precipitation. In-lake metal precipitation is undesirable for several reasons. The process (hydrolysis) is acid-producing, precipitates may clog important respiratory organs of aquatic organisms and it might affect the nutrient availability of the lake by co-precipitation of inorganic phosphate.

In 2011 a white precipitate was found on bedrock surfaces in a little water-fall upstream of the treatment basin at M20. This precipitate accumulated over time. White precipitates also formed on bedrock surfaces along the ocean shore-line at the outlet to the ocean downstream of M15/16 and M17. As at M20 these precipitates were strongly attached to the bedrock and hard to remove. The precipitates are assumed to first settle as hydroxysulfates and then gradually convert to more crystalline forms.

At M15/16 Al and probably some of the heavy metal ions precipitated on arrival in the Kaldvell fjord 500 m downstream of the deposit. Based on the mass transport of sulfur (Table 4) and the relation between mean concentrations of sulfate and Al, the annual sludge production (as Al-hydroxysulfate) in this fjord is estimated to be about 40 tons (10 tons as Al). Sedimented metal-rich material may make the fjord sediments closest to the stream outlet toxic to bottom-dwelling organisms, and the particles may be resuspended by wave-action and transported to marine areas further away from the original sources. Clogging after polymerization and precipitation of metals on vital biological structures, e.g. fish gills (Muniz and Leivestad, 1980; Rosseland et al., 1992), although not documented here, may be a characteristic feature of these waters.

### *3.5. Effects of Countermeasures*

In the Stordalen stream at M15/16, pH increased from low pre-construction values during the first part of the construction period due to added shell sand in the deposit and gravel in the treatment dam. In summer 2008, pH decreased to below 6.0 and remained at 4-5 thereafter (Fig. 2). The amount of added neutralizing agents and their neutralizing ability was obviously inadequate to cope with the weathering rate of waste rock in the deposit.

Lake Lomtjenn at M17 has a theoretical water retention time of only 10-15 days. This means that a major change of the inlet water chemistry will change the water chemistry of the lake totally within 2-3 months at mean flow, and even faster at high flow. The rapid reduction in Al concentrations in summer 2009 (Fig. 2) was therefore a response to countermeasures at the deposit. Effects of added shell sand may explain why Al concentrations have been at a lower level during the last part than in the middle of the monitoring period.

Water chemistry in samples from downstream of deposits M17 and M20 at the visits in 2009 were substantially different (Table 6). The charge balances typically show that the sulfate and nitrate ions at the deposits are balanced mainly by Ca, Mg, and Al ions. At M17 (200 m downstream) 79-83 % of the  $\text{SO}_4$  was balanced by Ca and Mg. By being substantially higher than 40-55 %, typical for samples close to exposed bedrock in the area (Hindar and Lydersen, 1994; Hindar and Iversen, 2006), this clearly shows that added liming material had an effect on the water chemistry. But the liming effect was insufficient, resulting in pH 4.3 and 23-27 mg/L of aluminum. The Al concentration corresponded to 15-18 % of the ion balance. At M20 (Urdalen) Ca+Mg balanced the sum of  $\text{SO}_4$  and  $\text{NO}_3$  entirely, the Al concentration was relatively low (23-260  $\mu\text{g/L}$ ), and pH circumneutral (7.1-7.2).

Substantially lower total N concentrations at M17 in 2009 (Table 6) partly reflects the smaller amounts of rock material at M17 than at M20 (Table 1). Nitrogen in the construction phase comes from about a 50/50 distribution of  $\text{NH}_4$  and  $\text{NO}_3$  in explosives used for blasting. These explosives contained 80 %  $\text{NH}_4\text{NO}_3$ , and typical doses were 0.65 and 2.5 kg explosives/ $\text{m}^3$  of solid rock for earth works and tunnel works, respectively (Hindar et al., 2007). Interestingly, the rate of oxidation of  $\text{NH}_4$  to  $\text{NO}_3$  seems to be higher at M20, indicated by the substantially lower amount of  $\text{NH}_4$  relative to

NO<sub>3</sub> at M20. This result is probably related to the higher pH at this site. High NO<sub>3</sub> concentrations at M20 are a result of the dominance of waste rock from tunnel works at this deposit.

**Table 6**

Water chemistry data for sites immediately and 200 m downstream of M17 and M20 at two sampling occasions in 2009.

|                 | Date      | pH   | Cond.<br>mS/m | Alk.<br>mmol/L | Tot P<br>µg/L | Tot N<br>mg/L | NH <sub>4</sub> -N<br>mg/L | NO <sub>3</sub> -N<br>mg/L | TOC<br>mg/L | Cl<br>mg/L | SO <sub>4</sub><br>mg/L | Al<br>µg/L | Ca<br>mg/L | K<br>mg/L | Mg<br>mg/L | Na<br>mg/L | Fe<br>µg/L |
|-----------------|-----------|------|---------------|----------------|---------------|---------------|----------------------------|----------------------------|-------------|------------|-------------------------|------------|------------|-----------|------------|------------|------------|
| At M17*         | 18-May-09 | 4.42 | 176           | -              | 7             | 6.12          | 2.40                       | 2.50                       | 4.3         | 78         | 919                     | 37400      | 209        | 7.2       | 65         | 60         | 1390       |
| At M17          | 22-Jul-09 | 3.95 | 141           | -              | 2             | 6.49          | 1.25                       | 4.05                       | 5.8         | 26         | 817                     | 44500      | 140        | 6.2       | 48         | 27         | 1040       |
| At M17 + 200 m* | 18-May-09 | 4.39 | 160           | -              | 5             | 4.84          | 2.30                       | 1.70                       | 4.3         | 73         | 807                     | 22900      | 187        | 6.7       | 57         | 54         | 2420       |
| At M17 + 200 m  | 22-Jul-09 | 4.34 | 157           | -              | 2             | 5.44          | 1.05                       | 3.75                       | 4.8         | 86         | 776                     | 27000      | 178        | 7.2       | 51         | 56         | 379        |
| At M20*         | 18-May-09 | 7.60 | 121           | 1.19           | 7             | 27.0          | 0.84                       | 23.5                       | 5.0         | 17         | 469                     | 472        | 175        | 6.1       | 39         | 34         | 279        |
| At M20          | 22-Jul-09 | 6.79 | 129           | 0.54           | <1            | 29.9          | 0.49                       | 29.0                       | 4.4         | 17         | 523                     | 27         | 185        | 8.6       | 42         | 36         | 51         |
| At M20 + 200m*  | 18-May-09 | 7.22 | 74            | 0.76           | 5             | 10.3          | 0.30                       | 9.40                       | 3.0         | 16         | 271                     | 261        | 98         | 5.3       | 21         | 20         | 400        |
| At M20 + 200 m  | 22-Jul-09 | 7.16 | 117           | 0.87           | <1            | 20.9          | 0.20                       | 20.0                       | 3.8         | 19         | 468                     | 23         | 175        | 8.3       | 38         | 31         | 6.3        |

\* not filtered before analysis

Adequate countermeasures for treating acid-producing waste material in sulfide-ore mining areas are described and recommended in reports from the Swedish MiMi-program (Mitigation of the Environmental Impact from Mining Waste; Fröberg and Höglund, 2004), a similar program in Australia (Australian Government, 2007), and from several disposal sites for sulfidic rock elsewhere. Although the sulfate and metal concentrations may be lower, the environmental problems described here resemble those described for mining areas. These recommendations are therefore of great value.

According to Fröberg and Höglund (2004) countermeasures to reduce the possibilities for biological damage may be grouped in passive and active measures. Passive measures have relatively low costs and demand a minimum of operation over time. An important aim of passive measures is to avoid/reduce oxidation and thereby the sulfate production. Shortly after the waste rock is deposited, the main focus should therefore be on reducing the availability of water, moisture and oxygen. Oxygen and water infiltration may be mitigated by establishing the deposits under a water-table or cap waste rock dumps with adequate low-permeable clay and protective masses on top. A top covering of e.g. 75 cm of clay with a hydraulic conductivity of less than 10<sup>-6</sup> cm/sec (NS regulations, 1995) or a corresponding clay layer of 20-60 cm covered with 1-2 meters of protective moraine (Fröberg and Höglund, 2004) has been recommended. Active measures, such as water treatment, have high technical complexity and costs. They demand a long-term commitment for operation.

Part of the Highway E18 construction project was to identify sulfide-bearing rock material and transport this material to the three disposal sites described here. The lower 1.25 % S limit for waste is lower than a corresponding limit of 0.4 % set by the Nova Scotia sulfide-bearing material regulations (NS Regulations, 1995) and may have resulted in spreading of acid-producing rock material to areas outside the deposits.

The disposal sites were located in small watersheds to avoid potential harmful effects on e.g. valuable sea trout (*Salmo trutta* L.) populations in rivers crossing the new road line. Inadequate construction and capping, however, resulted in acid and metal-rich runoff from deposits M15/16 and M17 during the road construction period. Active measures are therefore necessary to avoid harmful ecological effects in the future. The remarkably stable S release rates vs a gradually decreasing effect of added neutralization agents at M20 calls for active measures also at this site in the future.



Added liming material has balanced  $\text{SO}_4$  to some extent. The charge balances at Lomtjenn downstream of M17 showed that the Ca increase was far from adequate. Further reduction of Al (LAI) to the critical value for brown trout (*Salmo trutta* L.) of about 0.1 mg/L would require another 10-12 mg/L of Ca at the time, and a final Ca-concentration close to 80 mg/L (4 meq/L). This would have resembled the situation at M20, where  $\text{SO}_4$  most of the time was balanced with added Ca. Mass balances based on such calculations should be the basis for design of effective countermeasures.

A major challenge for proper treatment is the different behavior of metals in response to increasing pH, and also the character of the precipitates. A pH of 5.5 may be sufficient to precipitate Al, but insufficient for Mn and other metals. Precipitation of Mn as oxy-hydroxides at higher pH may also facilitate precipitation of other metals, as they may be adsorbed to the oxides.

Fluffy Al-precipitates, as described here, may be hard to settle, and one might consider use of inorganic chemicals or organic polymers to coagulate the precipitates. Once precipitated the metal sludge has to be deposited, probably as potentially harmful waste in dedicated areas.

Establishment of the deposits in other areas in the region would not have “solved” the problem. Rivers with buffering capacity of any significance do not exist in the area, and the catchment areas should have been larger than 300 km<sup>2</sup> for proper dilution to avoid the critical concentrations of LAI for brown trout. Rivers in catchments of that size may support the genetic and economically valuable Atlantic salmon (*Salmo salar* L), which is even more sensitive to Al than the brown trout (Kroglund et al., 2008).

Simple countermeasures will probably not be ecologically adequate for receiving fresh and marine waters, and facilities for multi-step treatment will be expensive. The important question for the environment and the people living in this area will be if there is sufficient commitment and willingness to pay what it costs to establish and run such facilities.

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