

Highway E18 Grimstad-Kristiansand; effects and quantification of acid runoff from deposits of sulphide-bearing rock



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REPORT

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Abstract

The new highway E18 Grimstad-Kristiansand, constructed in the period 2006-2009, crosses through sulphide-bearing rock. Oxidation after blasting results in formation of sulphuric acid, which may enter the runoff. Blasted sulphide rock material was therefore deposited. Deposit M17 consisted of about 700.000 tonnes of rock material and covered 50.000 m² of terrain. Although a total of 11.000 tonnes of shell sand and limestone gravel was used, runoff from deposit M17 became acid and aluminium-rich during the construction phase. pH at the outlet of the downstream situated Lake Lomtjenn was reduced to pH 4.0-4.6, and aluminium concentrations increased to very high levels (10-20 mg/l) in 2008/2009. Countermeasures in 2009 increased pH, and the Al-concentration decreased to below 5 mg/l. The water quality stayed toxic to fish, however, and the lake did not recover. It was estimated that the initial annual sulphur transport probably was in the order of 0.5 % of the total sulphur content in deposit M17. This weathering rate will result in ongoing consolidation of the rock pile. A plan for new measures at the deposit, launched by CJV in May 2011, is described, and a procedure for approaching good ecological status is proposed.

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1. Vegbygging	1. Road construction
2. Sulfidstein	2. Sulphide-bearing rock
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Preface

E18 Grimstad-Kristiansand, constructed in the period 2006-2009, goes partly through sulphide-bearing rock. Blasting results in acidic runoff, and sulphide rock material was therefore placed in several deposits offsite.

As the water quality was reduced downstream one of these deposits (M17), the construction company CJV E18 Grimstad-Kristiansand asked NIVA to evaluate the water chemistry more closely. In April 2011 NIVA was also asked to suggest a procedure for approaching good ecological status in the downstream Lake Lomtjenn. A plan for new measures at the deposit, and the procedure is included in this report.

The water quality downstream of the nearby deposit M20 was acceptable throughout most of the construction period, and data from this deposit have been evaluated for comparison.

Data from the NIVA monitoring station in Urdalen was supplied by Rolf Høgberget. Liv Bente Skancke assisted with water sampling and quality check of the data until April 2007.

Dr. D. Kirk Nordstrom of the US Geological Survey visited both deposits in July 2009, on invitation from NIVA. Assistance with the geochemical part of the report and interpretation of data is highly appreciated.

Contact persons at CJV were Dr. Martin Schreck until September 2009, Naveed Hussain and others until January 2011, and Nico von der Hude at Bilfinger-Berger SE thereafter.

Grimstad, July 6, 2011

Atle Hindar

Contents

Summary	5
1. Background	6
1.1 Acid production from sulphide-bearing rock	6
1.2 Produced acid vs added base	7
2. Deposits M17 and M20	9
3. Data, sampling and analyses	14
4. Results	15
4.1 Monitoring results	15
4.2 Data at the deposits	24
4.3 S-transport	28
5. New measures at M17	31
5.1 Passive and active measures	31
5.2 Procedure	32
5.3 Design of new measures	34
5.4 Target water quality and anticipated effects of new measures	36
6. Conclusions	37
7. References	38

Summary

The new highroad E18 Grimstad-Kristiansand crosses through sulphide-bearing and acid-producing rocks. Oxidation following blasting, results in the formation of sulphuric acid, which may enter the runoff. Sulphide rock was identified by the construction company CJV and deposited offsite.

Runoff from the deposit M17 in the small valley Mannfalldalen and from the deposit M20 in Urdalen was examined.

Deposit M17 consisted of about 700.000 tonnes of sulphide-bearing rock material and covered 50.000 m² of terrain. 4.500 tonnes of shell sand was added to prevent acid runoff. The deposit has a flattened surface, and the surface water drainage system is not yet constructed to avoid entrance of runoff from the surrounding terrain. The limited amount of soil used as covering of the deposit, could not prevent oxygen and precipitating water from entering through the surface.

Runoff from deposit M17 became acid and aluminium-rich after the construction work started up. From August 2008 pH of the downstream Lake Lomtjenn was reduced from its background level of pH 4.8-7.0 to pH 4.0-4.6. At this time the aluminium concentration increased from relatively low to a very high level (10-20 mg/l), resulting in toxic conditions for fish. NIVAs fish net survey in Lomtjenn in mid May 2009 resulted in no fish.

In 2009 and 2010 6.500 tonnes of shell sand and limestone gravel were used downstream. These countermeasures, together with reduced sulphate concentrations resulted in increased pH, and the Al-concentrations at the outlet of Lake Lomtjenn decrease to below 5 mg/l. The water has stayed toxic to fish, and the lake did not recover, however.

Runoff from deposit M20 was not acid during most of the monitoring period due to other construction characteristics than M17. Acidic episodes have been documented, however, and we suspect acidic episodes to be more severe and more frequent in the future.

In order to get more quantitative information of the sulphur transport and the acid production, daily runoff from a nearby monitoring station was coupled to the water chemistry data. Assuming that the sulphur content is 2% in the deposits, the annual S-transport corresponds to 0.45 % and 0.12 % of the total sulphur in deposit M17 and M20, respectively. Linear increases in accumulated S-transport indicate very steady weathering of the deposits. Data from elsewhere show that acidic runoff may persist for decades.

Recovery of a healthy Lake Lomtjenn requires adequate measures at deposit M17. A plan for new measures, launched by CJV in May 2011, is presented and commented.

A procedure for how to approach good ecological status in Lake Lomtjenn is suggested. Costs for adequate measures might be high compared to the ecological benefits, and part of the procedure is that the environmental authorities and the construction company agree on an acceptable cost limit. In the case that costs for reaching target levels of pH and aluminium are above the limit, alternative use of money should be considered.

In line with the proposed procedure, the suggested new measures at M17 should be constructed and the effects of these attempts downstream of deposit M17 should be monitored. The monitoring period could be used to discuss and make decisions related to additional measures and costs if acceptable.

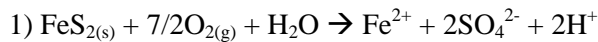
1. Background

The new highroad E18 Grimstad-Kristiansand goes partly through sulphide-bearing and acid-producing rocks. Oxidation following blasting, results in the formation of sulphuric acid, which may enter the runoff. Sulphide rock was identified by the construction company CJV based on temperature-increase of representative samples of drilling dust after addition of hydrogen peroxide. Blasted sulphide-bearing material was deposited offsite.

In the following the geochemical processes that produce acid runoff are described, and we also look at the potential effects of adding neutralising material.

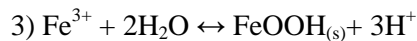
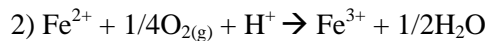
1.1 Acid production from sulphide-bearing rock

Oxidation of iron sulphides after blasting results in the production of ferrous ions and acid according to this equation:



Access to water/humidity and oxygen is essential, and sulphuric acid (H_2SO_4) is produced. This equation is a simplification – many sequential reactions in a row with production of several intermediate minerals may take place, see e.g. Nordstrom (1982) and Nordstrom and Alpers (1999). The acid is also partly neutralised by the weathering process that liberates base cations, such as calcium and magnesium.

Fe^{2+} from equation 1 is oxidised to Fe^{3+} , which may be precipitated as amorphous oxyhydroxides and eventually transformed to oxides:



The net effect of the oxidation and precipitation of iron is production of two moles of H^+ for each mole of Fe^{2+} . As the oxidation of iron may take place close to the source, iron may be stripped from the leached water before the acid water enters receiving surface waters. However, reactions 2 and 3 may be delayed, and hence also the acidification, as the Fe^{2+} may be transported over distances and eventually oxidise at higher pH and better availability of oxygen further downstream.

The precipitation of oxidised Fe-minerals results in clogging of all kind of surfaces and the particles themselves are toxic to fish due to clogging of fish gills.

The oxidation of sulphides may be accelerated by the resultant Fe^{3+} , which oxidises FeS_2 much faster than oxygen at low pH:



Availability of Fe^{3+} may thereby be critical for the rate of this abiotic oxidation.

In the presence of S and Fe oxidising bacteria of the type *Acidithiobacillus ferrooxidans*, oxidation of FeS_2 will proceed even faster. Availability of oxygen will therefore be the most essential limitation of the process, see several articles on this by Nordstrom and co-workers (e.g. Nordstrom and Alpers 1999; Nordstrom 2003) and a review article from the Swedish MiMi project (Herbert 1999).

Blasting by use of ammonium-nitrate based explosives will increase the availability of nitrate, which may be used as energy source for nitrate-reducing bacteria. These bacteria may thereby oxidise sulphides in an environment with limited access to oxygen. However, nitrate will be washed out from the rock material relatively rapid, probably before the nitrate-reducing bacteria are established, so this effect may be of little significance for acid production.

After oxidation of sulphides to sulphate, the sulphates may form a variety of secondary minerals. These metal-sulphate salts may be formed under different conditions and may thereby be of different composition. Available ions will be important, and as long as base cations are low, the hydrogen, aluminium, and iron ions will probably tend to dominate together with sulphate.

Sulphate minerals, formed preferentially in dry periods, will eventually dissolve. This dissolution is rapid. According to Jambor et al. (2000) "dissolution of Fe sulphates during storm runoff or after reclamation efforts can acidify streams, can rapidly increase metal loading to surface waters, and can lead to the development of acidic groundwaters." The metal loading and increase in toxic aluminium ions is relevant for the E18 area.

1.2 Produced acid vs added base

Undisturbed catchments of the Lillesand area are very poor in base cations due to the extremely low weathering rate of the dominating minerals (mostly gneiss and granites). This is why water acidified so easily in the 1950'ies and onwards with relatively modest input of acids from rain. Water qualities in the area are therefore characterised by low ion content, low/no buffering capacity and low pH (pH<6). Significant amounts of excess sulphates from blasted rock material must therefore be balanced by added base to avoid low pH and increased metal concentrations in surface waters.

Data from the events at Storemyr, Lillesand in August 2006 showed that the very high sulphate concentrations (up to over 1000 mg/l) were partly (40-50%) balanced by Ca and Mg from the rock itself (Figure 1; Hindar and Iversen 2006). This is a result of the weathering process, as the strong sulphuric acid dissolves minerals. Only close to half of the increase in sulphate downstream of the deposits can thus be regarded as a potential problem. This half is balanced mainly by toxic Al-ions, leaving 1-2 % for increase in H⁺. pH (-log [H⁺]) will typically be 3.5-4 close to the source and above 4 further downstream.

Based on this interpretation of water chemistry, sulphate is the driving force, and its concentration shows the potential for acid production. If aluminium is a significant part of the buffering, the water will also be acid and toxic. If buffering substances, such as calcite (calcium carbonate; CaCO₃), are added, the Ca concentration will increase, and Al concentrations will be correspondingly reduced. Adequate buffering will also increase pH (lower the H⁺ concentration).

Sources for neutralising the "excess" sulphate can be carbonates of marine shell sand, limestone gravel or other carbonate material. Hydroxides, such as caustic soda (NaOH) and slaked lime (Ca(OH)₂), with larger neutralising capacity, are used in mine areas. If mixed in a fill of sulphide rock material, the store of carbonates and hydroxides will gradually be reduced as the neutralisation reactions proceeds. The acid-producing potential of the rock pile vs the neutralising potential of the added base is essential for a long-lasting effect.

Experience with shell sand and limestone gravel in acid streams is that the surfaces tend to be covered by precipitates of a mix of aluminium, iron and organic material. It may thereby become inactivated. Extensive precipitation of iron has been observed downstream of sulphide rock material. Compared with sulphate, the concentrations of dissolved iron could be

relatively low in the runoff water, however, thus indicating massive iron precipitation inside such fills.

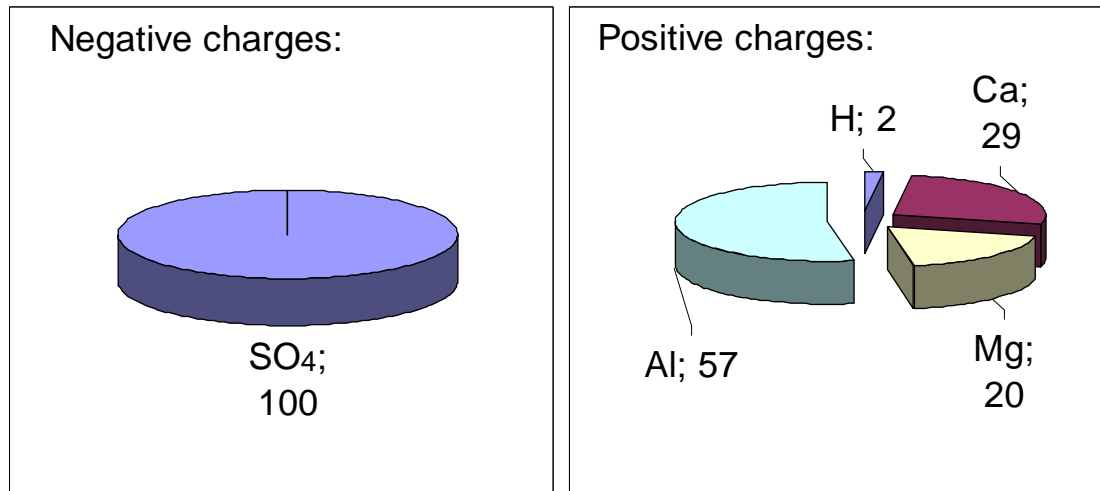


Figure 1. Charge balance (relative numbers; minor contributors to the balance not shown) for untreated runoff at Storemyr, Lillesand. Negatively charged sulphate is partly balanced and thereby partly buffered by calcium and magnesium from the rock itself, but aluminium is the dominating counter-ion. This water was very acid (significant concentration of H⁺) and extremely toxic to fish. Data from Hindar and Iversen (2006).

If not in adequate amounts, the buffering by added carbonates and hydroxides will be reduced over time. In a few years the non-neutralised excess sulphate will result in reduced pH and increased aluminium concentrations downstream.

An interesting example is Lake Langedalstjenn downstream of the Gaupemyr area, Lillesand. After blasting of 200 000 m³ sulphide-bearing rock in the late 1980's, the lake downstream acidified, the fish died and this former drinking water supply for 100 people was abandoned (Hindar and Lydersen 1994). Sulphate concentrations at the lake outlet have thereafter been reduced from 80 mg/l around 1990, via 44 mg/l in 2001 to 30 mg/l in 2008. In the same period pH increased from 4.4 to 5.1 and concentrations of toxic aluminium were reduced from 7 mg/l to 0.5 mg/l. The water quality was still toxic, however, and the lake was limed in September 2008. No investigations have been conducted to find the reason for the reduced sulphate, but capping of parts of the blasted area upstream with asphalt and roof surfaces might have been important.

2. Deposits M17 and M20

The two deposits M17 and M20 have been built over time and also changed their appearance over time. Both are built on relatively dry land and M17 is mostly unsaturated hydrologically. Part of M20 is soaked into a former swamp area. The pictures from the deposits in Figure 2 were taken in November 2009.



Figure 2. Deposit M17 (upper panel) with fill area, lower dam and downstream area with shell sand and gravel on November 17, 2009. Sampling point was at the dam outlet and 200 m downstream, at the base of the picture to the left. Deposit M20 (lower panel) with fill area, and outlet stream on November 17, 2009. The yellow-brown colour of the sulphide-bearing rock is clearly seen. Picture to the right shows the pH-measuring and sampling point 200 m downstream.

Deposit M17 was started up in May 2008 and finished in a late phase of the road project. The volume of the fill was intended to be 700-800 000 m³, but the actual volume was significantly less (Table 1). It was placed in the valley Mannfalldalen (Figure 2), as suggested by NIVA and Jordforsk (Hindar and Roset 2003). Mannfalldalen is parallel to the valley Fjelldal and River Fjelldalselva, an important sea trout river of the area. The small stream in Mannfalldalen follows the valley to Lake Lomtjenn, and enters the sea at Kosvik.

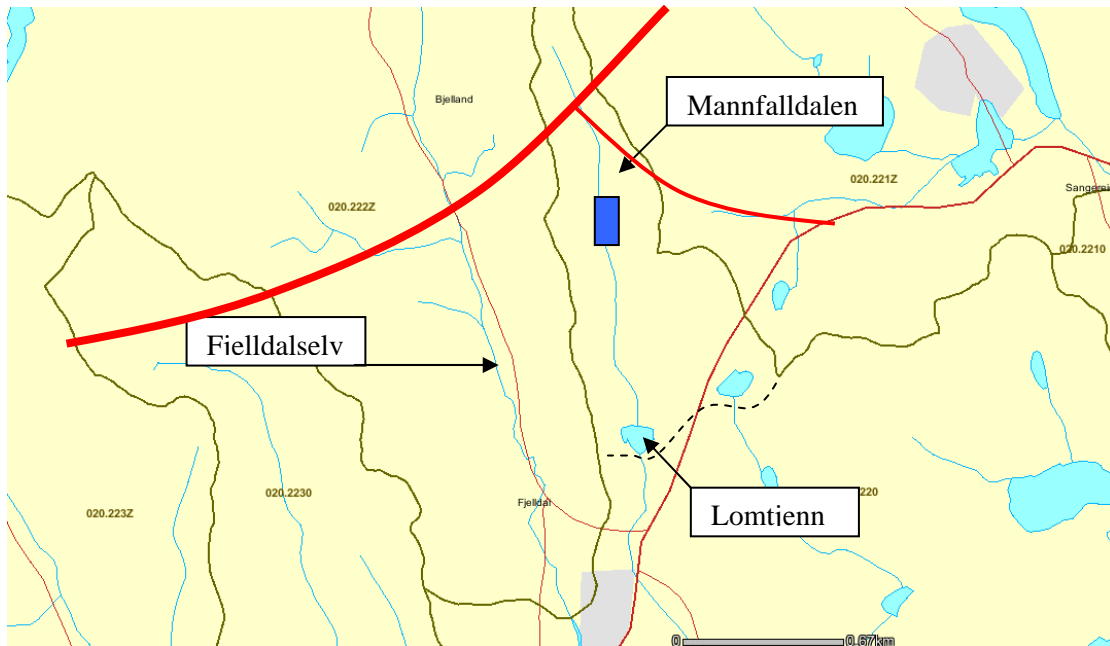


Figure 3. Situation of M17 (blue square) in the valley Mannfalldalen and Lake Lomtjenn. Black lines are delimitation of catchment areas according to the NVE database Regine. Stipled line is drawn after thorough inspection in the field, see text. The new E18, the access via Kjerlingland in the east and River Fjelldalselva are indicated. Source: NVE Atlas.

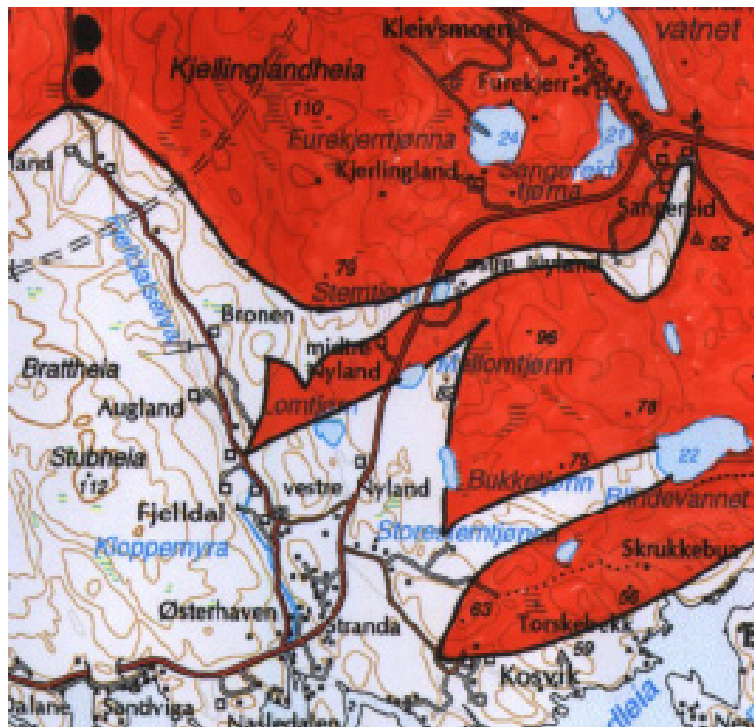


Figure 4. Geological map of the Lomtjenn/M17 area. Red colour indicates area with high risk of sulphuric bedrock. Black dots are areas with documented high concentrations of sulphur-containing bedrock. From Frigstad (2009).

The monitoring station of the environmental monitoring programme (Miljøkontroll-programmet) was at the outlet of Lomtjenn. After thorough inspection of the relatively complicated catchment, the southern delimitation was drawn as seen in Figure 3, and the catchment size calculated (Table 2). The outlet of the small lake Mellomtjønn is in north and not in south as indicated by the map. The stream runs together with two other small streams in the area of the small farm Midtre Nyland through ditches to Lomtjenn. The main blasted ditch connects this catchment to the catchment of Lomtjenn, thus increasing the total area by more than 10%. This has implications for calculations of the sulphur transport, see later sections.

Shell sand was used in the construction of deposit M17 and in treatment basins (Table 1).

Table 1. Data for the two deposits.

	M17 deposit	M17 downstream	M20 deposit	
Area of deposit	50 000		40 000	m ²
Volume of deposit	270 000		400 000	m ³
Mass weight	729 000		1 080 000	tonnes
Added shell sand	4 500	3 500	6 000	tonnes
Added slakes lime			600	tonnes
Added limestone gravel (1-10 cm)		2 000		tonnes
Construction start	May 2008		Autumn 2007	
Redirection of flow from			June 2009	

Table 2. Catchment sizes for the two deposits and monitoring stations.

Catchment areas, km ²	M17	M20
For the deposit	0.29	0.72
For the deposit after redirection of flow		0.19
For 200 m downstream the deposit	0.45	0.74
For 200 m downstream after redirection of flow		0.21
For the main monitoring station	0.89	0.80
For the monitoring station after redirection of flow		0.27

As can be seen from the geological map (Figure 4), parts of the Lomtjenn catchment and M17 area, especially the upper part, is in the area of sulphide-bearing rock. Any construction work in this area would therefore result in increased sulphate concentrations downstream.

Two dams were constructed at the foot of the mass deposit M17 to neutralise water. Heavy rainfall destroyed the lower dam shortly after construction, but reconstructed and filled with limestone gravel some months later. A relatively flat marshland area, filled with limestone gravel and shell sand, further downstream was also part of the treatment. The marshland area was meant to be part of the deposit area, but as the deposit was significantly smaller than planned, the area could be used for deacidification purposes.

In April 2009 a white, fluffy precipitate was produced downstream of M17, see later sections.

In early July 2009 about 2500 tonnes of shell sand and 1-2 cm gravel was spread out over the marshland downstream of the deposit and lower dam. The idea was that the stream water should be dispersed and neutralised after passing the flattened area. Heavy flow on July 18 changed the surface structure of the area and the water was concentrated in one main stream. Rapid flow seemed to prevent much contact time with the shell sand.

In late September 2009, 1-10 cm of limestone gravel was filled in the lower dam, and by the time of the site visit and sampling on November 5, 2009 the outlet of the dam was closed. All the gravel was below the water table, and this could probably have forced water to pass through the lower barrier of the dam at low runoff. On November 5 the water passed over the crest at moderate flow, however, probably due to both the large amount of water and the sealing of the inner dam wall with particles and precipitates.

The flow events, especially the extreme flow on October 24-25, partly destroyed the dam construction and the surface of the neutralisation zone downstream. Also, the stream bed and its nearest surroundings further downstream were filled with settled shell sand from upstream areas. Erosion of shell sand layers was noticed during sampling on October 24, 2009, and at this time the stream was grey and milky of particles.

In late autumn 2009 clay material was stored in piles on M17 for later use as a surface cover. These works were delayed, but will be carried out in summer 2011 according to recent plans.

Deposit M20 in Urdalen (Figure 5, Table 1 and Table 2) was started in autumn 2007, and it was the first in a series of several deposits for sulphuric rock material along the road line. It was, also as suggested by NIVA and Jordforsk (Hindar and Roset 2003), placed in a former marshland close to the western entering of the Steinsås tunnel. As can be seen from the geological map (Figure 6), most of the Urevatn catchment and M20 area is in the area of sulphuric bedrock, and any construction works in the area would result in increased sulphate concentrations downstream. Building of a forest road several years before building of the new E18 started up had obviously disturbed the bedrock. Thus water at the inlet of Lake Urevatn already showed clear-cut signs on pH and concentrations of sulphate and aluminium at the start of the monitoring period.

M20 was constructed in a way that should secure runoff from being acid. This included use of slaked lime ($\text{Ca}(\text{OH})_2$) in the base and layers with large amounts of shell sand within the deposit (Table 1). A treatment basin was built about 200 m downstream the deposit. Water samples for the monitoring programme (Miljøkontrollprogrammet) were collected immediately downstream this basin. Also, a pH monitoring station was set up at this site by NIVA on contract with the Norwegian Public Road Administration.

As the deposit was built, water from the surroundings was drained outside the fill area, and led back again upstream the monitoring station. The size of the catchment area upstream the monitoring station was thereby unaltered until June 2009. However, during tunnel works water from Lake Urevatn was pumped up and used during construction of the Steinsås tunnel. This water was sent back to the lake again via the inlet stream, and had elevated pH which was reduced to pH 7-8 by use of CO_2 before it entered the stream. In periods of low flow during summertime this water could dominate the stream water chemistry, but had negligible influence for the calculated sulphur transport.

In June 2009 the upper two thirds of the catchment upstream the monitoring station was cut off by diverting the stream immediately upstream M20 directly to Lake Urevann through a blasted ditch. The flow at the monitoring station was thereby significantly reduced.

A fluffy, white precipitate was found immediately downstream of M20 in winter 2010/spring 2011. This precipitate was not analysed, but probably very similar to the precipitate found at M17 two years earlier, see later sections.

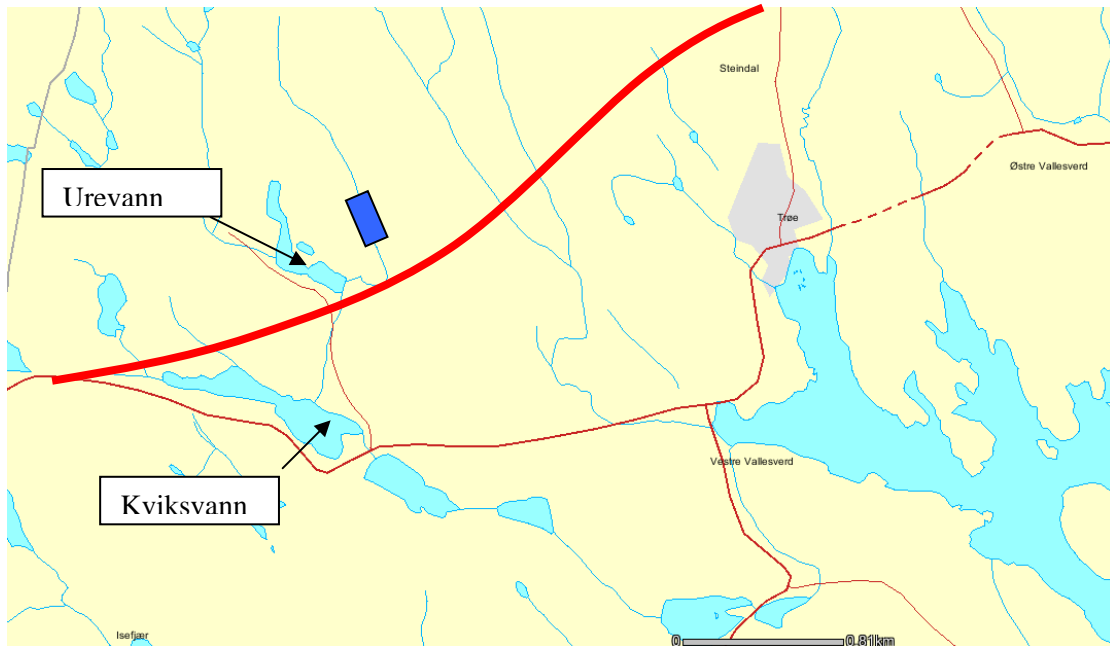


Figure 5. Situation of M20 (blue square), Lake Urevann and Lake Kviksvann. The watercourse enters the sea at Vestre Vallesverd. Source: NVE Atlas.

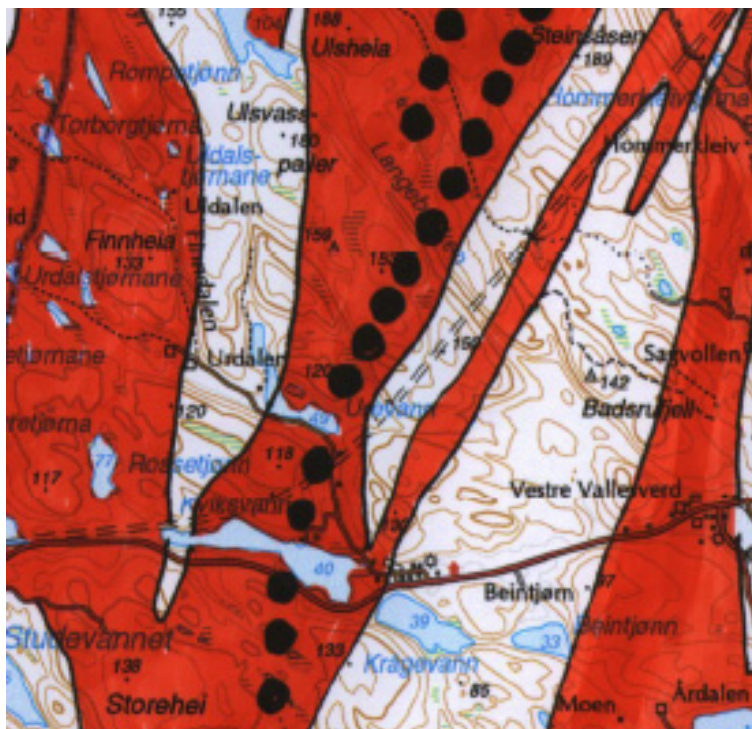


Figure 6. Geological map of the Urevatn/M20 area. Red colour indicates area with high risk of sulphuric bedrock. Black dots are areas with documented high concentrations of sulphide-bearing bedrock. From Frigstad (2009).

3. Data, sampling and analyses

Data mainly from the monitoring programme (Miljøkontrollprogrammet) have been used to characterise the water chemistry downstream both deposits over time and to calculate transport of sulphur from the deposits. Monitoring stations at M17 and M20 were located at the outlet of Lake Lomtjenn and at the inlet of Lake Urevann, respectively. NIVA measured these data from July 2006 until April 2007, and Vannlaboratoriet in Kristiansand thereafter. Data from NIVAs monitoring from 2009 are also included. Aluminium data in the figures are mainly as reactive Al (RAI; dissolved fractions of inorganic and organic Al) at low concentrations and total Al of unfiltered samples at high concentrations. At $\text{pH} < 5.5$ and Al-concentrations above 1.0 mg/l almost all Al is as dissolved, inorganic Al, i.e. the toxic, labile fraction (LAI).

To document the differences of the water chemistry downstream of the two deposits further, water samples were collected at two sites (immediately and 200 m downstream of the deposits) at two occasions in summer 2009. The first was on May 18, 2009 and the second on July 22, 2009. Sampling at M17 continued thereafter, whereas only one additional sample was collected at M20. On November 17, 2009 a sample was also taken from the redirected stream north of M20 to document its water chemistry.

The fluffy, white material that was formed immediately downstream of M17 and its sedimentation basins, was observed during a trip on April 23, 2009. Examination at NIVA showed no bacteria, and thus the material was analysed for the most likely chemical constituents. The material (see Figure 7 for pictures) was grabbed with a clean plastic bottle on May 18, 2009, and sent to the NIVA laboratory. At the lab, about 10 ml of the precipitate including water was freeze dried, dissolved in acid (HNO_3) and then measured by ICP-AES (Perkin-Elmer Optima 4300 DV ICP-AES spectrometer, Perkin-Elmer Autosampler AS 93 plus, Hewlett Packard LaserJet 100). Data are given as μg per gram dry weight of the material.

A similar white precipitate was also formed at M20 in winter 2010/spring 2011, but was not analysed.



Figure 7. White precipitate at the dam barrier of M17, and attached to submerged plants and stream bed about 200 m downstream (May 18, 2009).

On November 17, 2009, after the closing of the dam outlet at M17, a grey and very fluffy precipitate had formed on the surfaces of the coarse gravel in the lower dam. This precipitate

was sampled after careful disturbance of the gravel (see pictures in Figure 8), and concentrated by repeated sampling and sedimentation of water. At the lab, the particles were filtered through 0.45 µm Millipore filter and rinsed with distilled water, then dried and analysed as outlined above.



Figure 8. Fluffy, grey precipitates in lower dam at M17 before (left) and after careful disturbance of the limestone gravel.

Runoff data from the Birkenes research site (BIE01; NVE), about 18 km north of the deposits, have been used for sulphur transport calculations. Measured runoff at Birkenes was downscaled (if smaller catchment area than the 0.41 km² at Birkenes) or upscaled (if larger than Birkenes). Also, daily water flow was corrected for differences in mean specific runoff for the period 1961-1990 between Birkenes (34 l/s/km²) and the two other catchments (26 and 27 l/s/km² for Lomtjenn and Urdalen, respectively). In summer 2009 the uppermost two thirds of the catchment at M20 was redirected, and this was taken care of in the calculations.

All data for deposit areas and volumes, and also of added shell sand and gravel were given by CJV.

4. Results

4.1 Monitoring results

Data from the monitoring programme 2006-2011 show a remarkable increase in sulphate concentrations downstream of both M17 (outlet of Lake Lomtjenn) and M20 (inlet of Lake Urevatn) over time (Figure 9 and Figure 11).

At the outlet of Lomtjenn sulphate started to increase in July 2007, but stayed below 100 mg/l until August 2008. Following the rapid increase in August, pH decreased from background concentrations of 4.8-7.0 down to 4.0-4.6. At this time the aluminium concentration increased to very high levels, but data for the period September 2008 – February 2009 show large temporal variation.

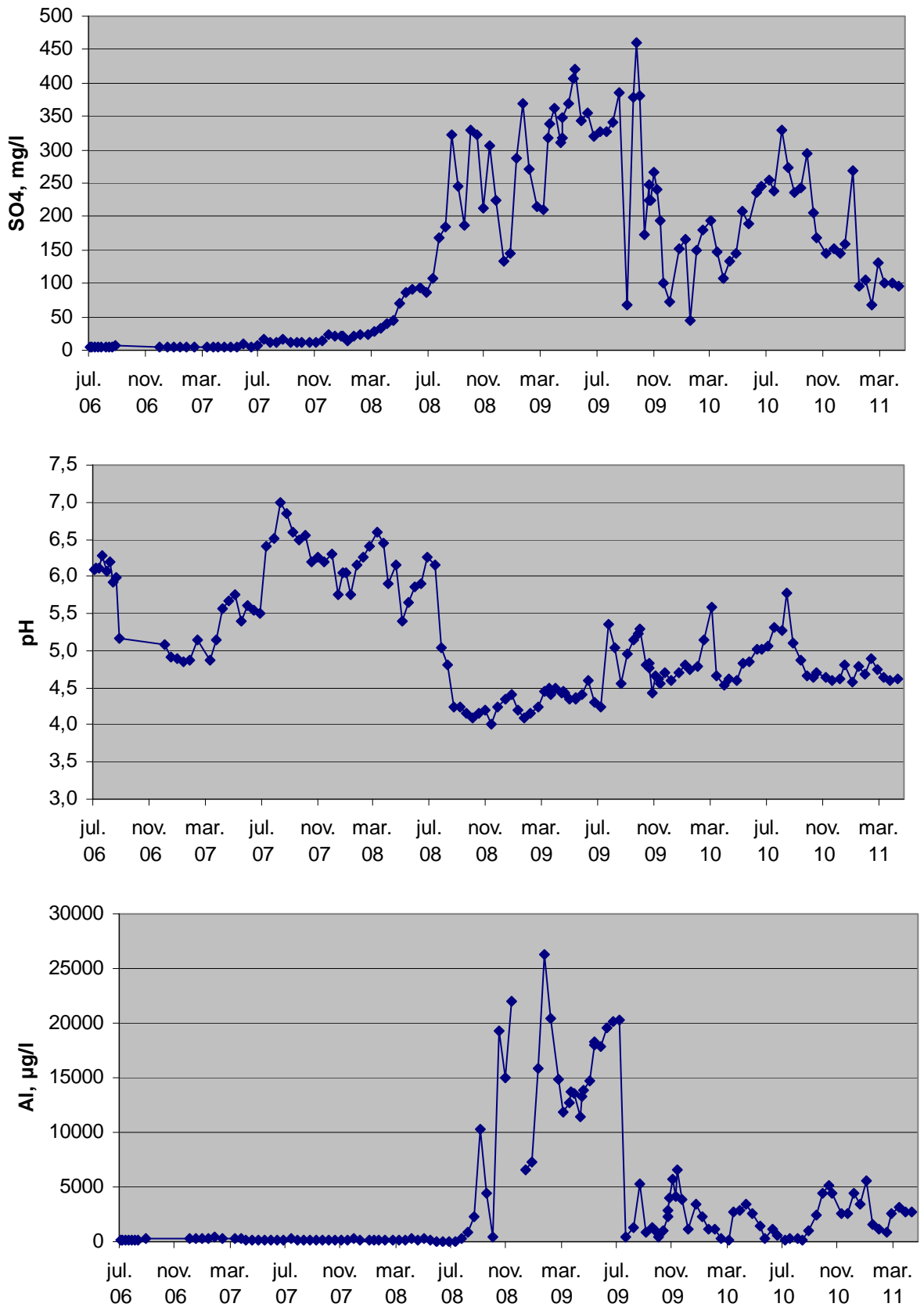


Figure 9. Outlet of Lomtjenn downstream of M17; sulphate, pH and reactive (at low concentrations) or total (at high concentrations) aluminium.

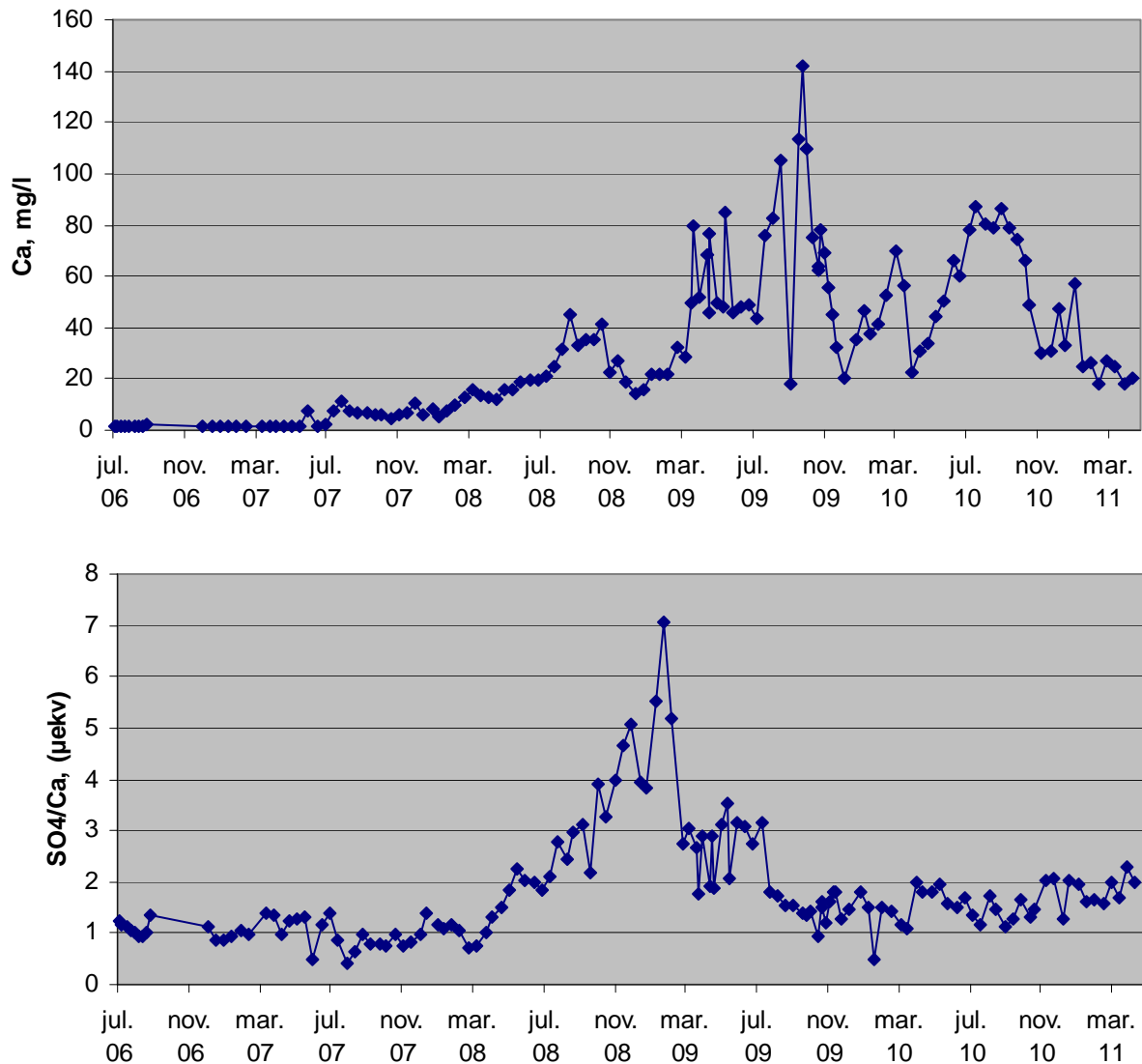


Figure 10. Outlet of Lomtjenn downstream of M17; calcium and the ratio sulphate/calcium on charge basis. A ratio close to one after the onset of the construction works indicates that sulphate is balanced by calcium due to addition of shell sand or other liming material.

From autumn of 2008 until summer 2009 Al-concentrations were 10-20 mg/l. From summer 2009 and until present Al concentrations have been markedly reduced (0.1-5.5 mg/l). This water quality is still extremely toxic for fish, however. NIVAs fish net survey in Lomtjenn in mid May 2009 resulted in no fish.

As shown in Table 4, high concentrations of six of eight heavy metals (not As and Cr) are associated with the leaching from deposit M17.

The lake was very turbid with a greenish appearance in 2009, obviously affected by particles transported to the lake. Shell sand particles, but also Al-precipitates may have caused this turbidity, see results in section 4.2.

Lake Lomtjenn has a theoretical water retention time (the mean time needed to fill up an empty lake with water from the catchment) of only 10-15 days if the mean water depth is 2-3 meters. This means that a major change of the inlet water chemistry will change the water chemistry of the lake totally within a month or two at mean flow conditions. In periods with high flow this will happen even faster. The rapid reduction in Al-concentrations in summer 2009 was therefore a response to measures at the deposit. High Al-concentrations until present, show that this was not sufficient for a recovery of the lake.

Reduction in sulphate concentrations, as observed in 2010 and 2011, together with a substantial effect 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. The liming effect can be seen from the charge balance for the sample at Lomtjenn outlet on 16 October 2010 (Table 3). 76 % of the sulphate was balanced with calcium, compared with typically 29 % without liming, and pH was 4.64. Without treatment pH would probably be 4.0 and the Al-concentration 22 mg/l.

Table 3. Measured data and charge balance for Lake Lomtjenn outlet on October 16, 2010 (upper panel). Magnesium was not measured, but estimated on basis of sulphate concentration and the relation between the main ions as seen in Figure 1. The charge balance in the lower panel is constructed. It is based on the same measured sulphate concentration and the typical distribution of Ca, Mg and Al in relation to sulphate in Figure 1. The difference in pH and Al-concentration between the two panels can thus be ascribed to a liming effect at M17.

Lomtjenn outlet 16102010			
	Measured		% of SO4
	mg/l	µeqv/l	
pH 4.64		23	1
Ca	65,9	3288	76
Al	4,97	552	13
<i>Mg</i>	10,5	864	20
SUM		4727	110
SO4	205	4305	

Constructed; no liming effect			
	Measured		% of SO4
	mg/l	µeqv/l	
pH 4.00		100	2
Ca	25	1248	29
Al	22	2442	57
<i>Mg</i>	10,5	864	20
SUM		4653	108
SO4	205	4305	

At the inlet of Urevatn sulphate started to increase in late 2006 (Figure 11), and was at relatively high levels from the summer of 2007. A rapid increase from 50-150 mg/l and up to about 400 mg/l took place in summer 2009, and the concentrations have stayed at that level since then. The main reason for this increase is the re-direction of flow upstream M20, which reduced the catchment by two thirds, resulting in a corresponding increase in concentrations.

In contrast to Lomtjenn, pH at Urevatn has been at acceptable levels (above 6.0) during most of the construction period. Al-concentrations were 0.2-0.4 mg/l (200-400 µg) before the construction period due to the low pH, and have, with some exceptions, been close to or below this level throughout the period. Some of the high concentrations may be due to formation of the aluminate ion ($\text{Al}(\text{OH})_4^-$) at high pH, but some are also associated with acidic water, e.g. the high value on April 14, 2011.

The high Al value in April 14, 2011 coincided with pH 5.3, and a flow situation was recorded at NIVAs monitoring station. About 1 mg/l of the Al was as labile Al (LAl), and the water was extremely toxic to fish during this episode. Mobilisation of Al during such episodes may explain why the white, fluffy precipitate had formed in winter 2010/spring 2011.

At both sites shell sand and other liming materials have been used in order to improve the water quality. Such material consists of calcium carbonate (CaCO_3), which might be traced by the calcium concentration. However, calcium also follows the sulphate from the blasted minerals, see Figure 1. In this report the ratio sulphate/calcium on a charge basis is used to identify influence of added liming materials.

Calcium concentrations at both sites increased as sulphate increased (Figures 10 and 12). But the SO_4/Ca ratio reveals large differences in liming effects. At Lomtjenn, the ratio was close to one before the construction period and throughout until early 2008. It then increased to 2, and showed a remarkable increase to very high levels starting in August 2008. At this time the buffering system of the lake collapsed and pH decreased to 4.0-4.6. Addition of shell sand reduced the ratio to 1-2 from late 2009.

At Urevatn the SO_4/Ca ratio started at 2-5 before the construction period, and the water was acid at the time. Then the ration decreased to below two in early autumn 2006 due to the addition of liming material at the deposit. From summer 2008 until the end of the calcium monitoring period the ratio stayed at 1-2. In the same time the alkalinity was high and pH acceptable.

Unfortunately, calcium was excluded from the approved monitoring programme at this site in October 2009, and the direct liming effect at M20 could not be followed further.

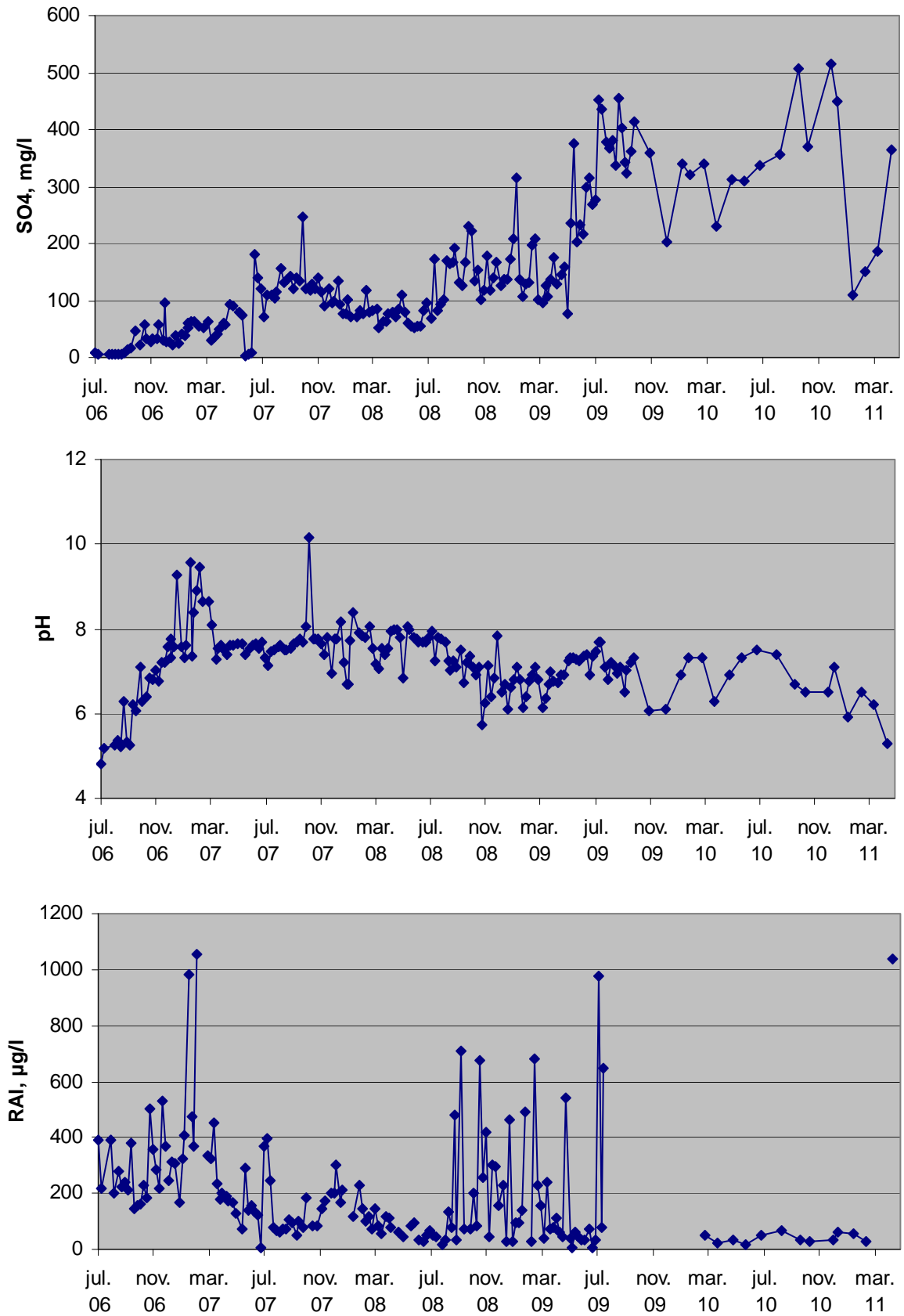


Figure 11. Inlet of Urevatn downstream of M20; Sulphate, pH and reactive aluminium.

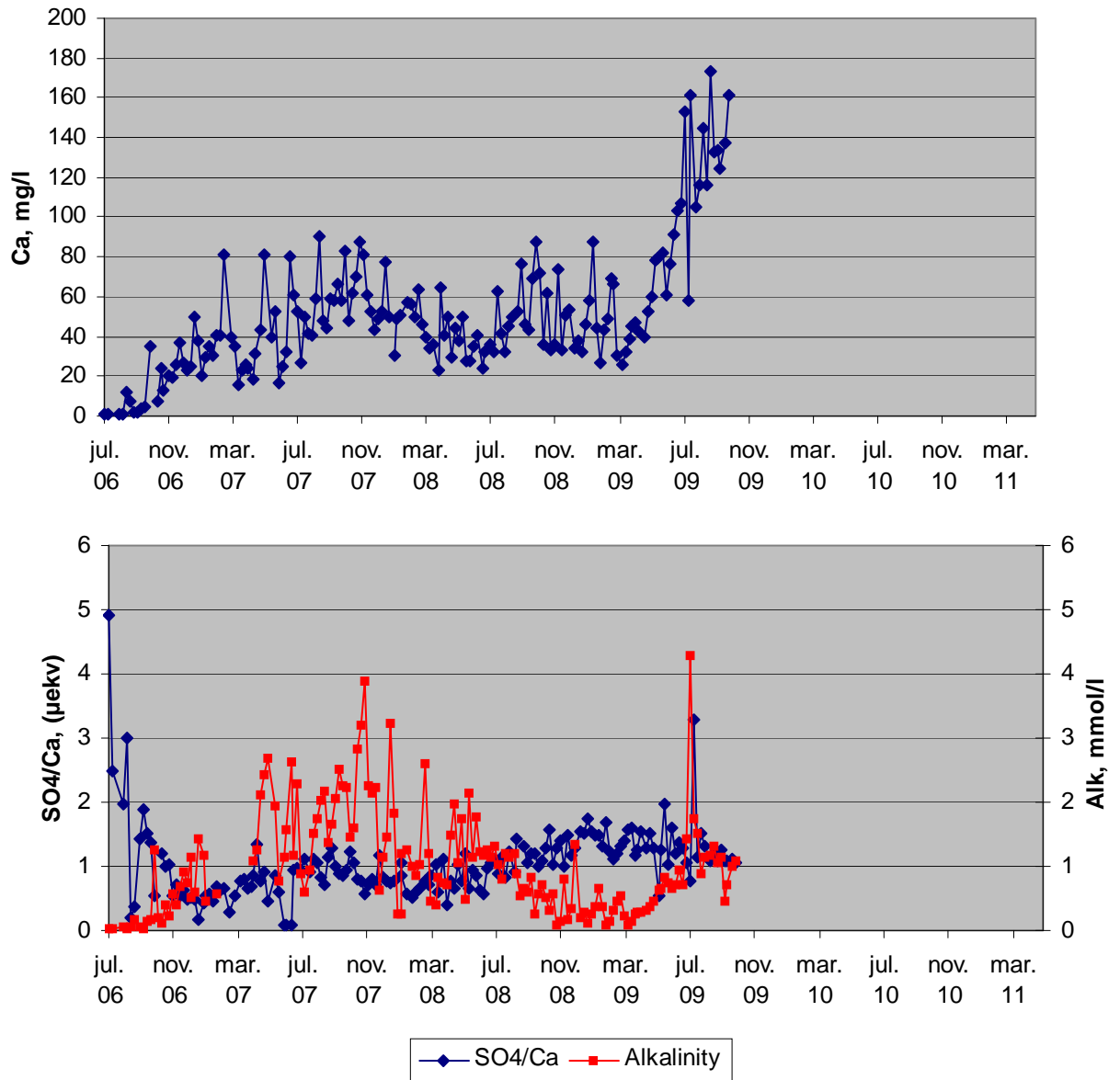


Figure 12. Inlet of Urevatn downstream of M20; Calcium and the ratio sulphate/calcium on charge basis. A ratio close to one after the onset of the construction works indicates that sulphate is balanced by calcium due to addition of shell sand or other liming material. Ca and alkalinity was excluded from the monitoring programme in October 2009.

Table 4. Water chemistry data for sites immediately and 200 m downstream of M17 at Lomtjenn, together with NIVA-data from the monitoring at the outlet of Lomtjenn. *Samples from May 18, 2009 were not filtered before analysis. All other samples were filtered, except for analysis of pH, conductivity and alkalinity.

	UTM NS meter	UTM ØV meter	Date	pH	KOND mS/m	ALK mmol/l	STS mg/l	Tot-P µg/l P	Tot-N µg/l N	NH4-N µg/l N	NO3-N µg/l N	TOC mg/l C	Cl mg/l	SO4 mg/l	Al µg/l	Ca mg/l	K mg/l	Mg mg/l	Na mg/l	Fe µg/l
At/M17*	64549	4592	18.05.09	4,42	176			7	6120	2400	2500	4,3	77,9	919	37400	209	7,2	64,5	59,5	1390
At/M17	64549	4592	22.07.09	3,95	141			2	6490	1250	4050	5,8	26,1	817	44500	140	6,2	47,6	27,2	1040
At/M17	64549	4592	29.09.09	6,56	212	0,949		3	4515	2250	1500	4,2	92,9	1040	300	304	8,0	76,7	59,5	10,6
At/M17	64549	4592	24.10.09	6,88	144	1,635	221	2	2870	980	1450	3,4	51,3	620	6260	173	5,6	41,8	34,7	243
At/M17	64549	4592	05.11.09	4,35	102			2	2560	710	1500	4,0	30,2	512	25100	105	4,8	29,3	23,4	305
At/M17	64549	4592	17.11.09	4,28	69,8			2	1740	500	890	4,1	20,5	329	16800	63,7	3,2	18,1	14,8	840
At/M17 + 200 m*	64545	4593	18.05.09	4,39	160			5	4840	2300	1700	4,3	73,0	807	22900	187	6,7	56,8	54,4	2420
At/M17 + 200 m	64545	4593	22.07.09	4,34	157			2	5440	1050	3750	4,8	85,7	776	27000	178	7,2	50,9	56,3	379
At/M17 + 200 m	64545	4593	29.09.09	7,81	215	3,803		3	3560	720	2050	4,4	94,2	957	56	375	7,8	71,2	61,8	26,2
At/M17 + 200 m	64545	4593	24.10.09	6,49	111	0,575	295	1	2420	780	1200	3,2	39,4	489	4290	139	4,7	32,4	27,9	322
At/M17 + 200 m	64545	4593	05.11.09	4,46	93,2			2	2270	590	1350	3,5	27,5	443	17300	102	4,1	25,9	21,9	195
At/M17 + 200 m	64545	4593	17.11.09	4,42	70,7			1	1680	480	930	3,2	21,9	336	15300	69,8	3,2	18,8	16,7	626
At M17, acid digested			24.10.09												41700	200				2850
At M17 + 200 m, acid digested			24.10.09												33900	149				9710
At M17, small stream	64549	4591	05.11.09	6,86	117	1,542		1	2270	370	1500	3,4	36,3	483	71	166	5,1	30,2	36,6	5,1
Lomtjenn, outlet*	6453968	459412	26.02.09	4,24	53,0			12	3230	700	2100	5,6	19,1	215	14900	32,6	2,3	12,5	16,0	
Lomtjenn, outlet	6453968	459412	25.03.09	4,41	80,7			6	4250	840	2650	3,7	32,4	338	13700	79,4	3,8	22,4	26,4	270
Lomtjenn, outlet	6453968	459412	16.04.09	4,43	75,5			6	3180	780	2050	3,5	33,7	311	11400	68,3	3,1	21,1	27,4	
Lomtjenn, outlet	6453968	459412	22.04.09	4,44	83,1			3	3480	870	2150	3,4	36,1	347	13800	76,5	3,4	24,1	29,4	
Lomtjenn, outlet	6453968	459412	18.05.09	4,37	93,1			4	4190	1050	2500	2,8	31,9	420	18300	85,2	3,8	28,1	30,0	
Lomtjenn, outlet	6453968	459412	29.09.09	5,23	105	0,054		2	2595	740	1400	3,4	44,4	461	1050	142	4,9	33,2	31,1	115
Lomtjenn, outlet	6453968	459412	24.10.09	4,82	63,8	0,040		2	1530	380	760	5,4	29,7	247	2270	63,7	2,3	15,5	17,9	184
Lomtjenn, outlet	6453968	459412	25.10.09	4,76	56,4	0,037		2	1380	340	750	4,9	22,2	224	2790	62,1	2,1	14,7	15,3	206
Lomtjenn, outlet	6453968	459412	05.11.09	4,67	63,6	0,033		1	1530	380	850	4,3	22,3	267	5750	69,1	2,5	16,4	17,0	135
Lomtjenn, outlet	6453968	459412	17.11.09	4,56	48,6	0,011		3	1160	300	630	4,3	19,8	193	6580	44,7	1,8	11,5	14,0	252
Lomtjenn, outlet	UTM NS meter	UTM ØV meter	Date	As µg/l	Cd µg/l	Co µg/l	Cr µg/l	Cu µg/l	Mn µg/l	Ni µg/l	Zn µg/l									
Lomtjenn, outlet	6453968	459412	25.03.09	1,3	3,94	220	0,73	40,5	1870	441	746									

Table 5. Water chemistry data for sites immediately and 200 m downstream of M20 at Urdalen, together with NIVA-data from the monitoring at the inlet of Urdevann. *Samples from May 18, 2009 were not filtered before analysis. All other samples were filtered, except for analysis of pH, conductivity and alkalinity.

	UTM NS meter	UTM ØV meter	Date	pH	KOND mS/m	ALK mmol/l	STS mg/l	Tot-P µg/l P	Tot-N µg/l N	NH4-N µg/l N	NO3-N µg/l N	TOC mg/l C	Cl mg/l	SO4 mg/l	Al µg/l	Ca mg/l	K mg/l	Mg mg/l	Na mg/l	Fe µg/l
At deposit M20*	64520	4540	18.05.09	7,60	121	1,192		7	27010	840	23500	5,0	16,6	469	472	175	6,1	39,4	33,8	279
At deposit M20	64520	4540	22.07.09	6,79	129	0,540		<1	29900	490	29000	4,4	17,1	523	27	185	8,6	42,3	35,9	50,7
At deposit M20	64520	4540	17.11.09	5,96	77,2	0,129		1	7250	250	6650	3,2	12,1	326	418	95,9	6,2	22,5	19,1	40,8
Urdevann inlet*	6451912	453984	18.05.09	7,22	73,8	0,762		5	10330	300	9400	3,0	15,8	271	261	97,9	5,3	20,6	19,6	400
Urdevann inlet	6451912	453984	22.07.09	7,16	117	0,873		<1	20900	200	20000	3,8	18,7	468	23	175	8,3	37,9	31,0	6,3
M20, stream in north	64523	4538	17.11.09	4,89	5,71	0,020		1	425	4	96	4,3	8,83	4,51	287	0,87	0,30	0,72	5,29	118

* not filtered before analysis

Table 6. Data for the white precipitate (including about 10 ml of water, see text) sampled on May 18, 2009 downstream of basin at M17 and of the grey precipitate sampled inside the dam at M17 on November 17, 2009.

Date	TC µg C/mg	TOC µg C/mg	Al µg/g	As µg/g	Ca µg/g	Cd µg/g	Co µg/g	Cr µg/g	Cu µg/g	Fe µg/g
Downstream basin at M17	19,8	20,8	218000	27	10500	0,6	22,2	11,7	129	12000
In basin at M17	66,3	57,4	79900	20	24900	0,6	21,2	35,9	165	38200

Date	K µg/g	Mg µg/g	Mn µg/g	Na µg/g	Ni µg/g	S µg/g	Si µg/g	Zn µg/g
Downstream basin at M17	1280	2750	219	1020	58	53200	4790	227
In basin at M17	7920	8720	344	565	52,1	9250	3720	288

4.2 Data at the deposits

Water chemistry from immediately and 200 m downstream of the two deposits were significantly different (Table 4 and Table 5). At M17 sulphate was twice as high as at M20, even though the amount of rock material is smaller at M17. This indicates a more rapid weathering rate and thereby sulphate production at M17. Smaller amounts of rock material are also reflected in significantly lower concentrations of nitrogen at M17. Nitrogen comes from about a 50/50 distribution of ammonium-N/nitrate-N in rest-explosives of the blasted material. Interestingly, the rate of oxidation of ammonium to nitrate seems to be higher at M20, indicated by the significantly lower amount of ammonium relative to nitrate at M20. This is probably due to the higher pH at this site.

The charge balances typically show that the sulphate and nitrate ions at the deposits are balanced mainly by calcium, magnesium and aluminium ions (Table 7). At M17 (Lomtjenn) on July 22, 2009 79 % of the sulphate was balanced by Ca and Mg. By being significantly higher than 50%, this clearly shows that liming material had an effect on the water. But the liming effect was insufficient, resulting in pH 4.34 and 27 mg/l of aluminium, corresponding to 18% of the balance. At M20 (Urdalen) Ca+Mg balanced the sum of SO₄ and NO₃ entirely, and aluminium was low (0.023 mg/l) and pH high (7.16).

These two pairs of samples clearly indicate a significantly higher weathering rate at M17 than at M20. We therefore decided a more quantitative approach to this issue by calculating and comparing the sulphur transport from the two fills, see next section.

At M20 data from the NIVA monitoring station 200 m downstream of the deposit show that pH may change very quickly as response to changes in water flow (i.e. water level), see Figure 13. During the heavy flow in late September 2009, pH was reduced to 5.8, indicating limited neutralising ability within the deposit during this episode. The flow episode on April 14, 2011 was even more severe, as already ascribed.

On heavy rainfall occasions we expect that the rainwater follows preferential flow-paths through the fill, and that the liming material gradually will be eroded and more easily degraded here than in surrounding parts of the fills. Also, increased deactivation of the carbonate surfaces by precipitates will reduce the neutralising effect. Although the pH seldom was reduced to below 6.0 in 2008-2011, we expect it to do so in the years to come.

Table 7. Measured concentrations and charge balances for samples from 200 m downstream of deposit M17 and M20 on July 22, 2009. Ions with positive charges are on top of each panel. Measured concentrations in mg/l are converted to microequivalents per liter (charge). Hydrogencarbonate (HCO_3^-) is the main contributor to measured alkalinity at $\text{pH}>7$, and is included for the M20 balance.

M17 22072009		Measured	
Deposit+200m	mg/l	$\mu\text{ekv/l}$	% of NO_3+SO_4
pH 4.34		46	0
Ca	178	8882	54
Al	27	2997	18
Mg	50,9	4187	25
Na	56,3	2449	
K	7,2	184	
$\text{NH}_4\text{-N}$	1,05	75	
Fe	0,38	21	
SUM		18841	97
SO₄	776	16296	
NO₃-N	3,75	268	
Cl	85,7	2418	
SUM		18981	
M20 22072009		Measured	
Deposit+200m	mg/l	$\mu\text{ekv/l}$	% of NO_3+SO_4
pH 7.16		0	0
Ca	175	8733	78
Al	0,023	3	0
Mg	37,9	3118	28
Na	31	1349	
K	8,3	212	
$\text{NH}_4\text{-N}$	0,2	14	
Fe	0,01	1	
SUM		13428	105
SO₄	468	9828	
NO₃-N	20	1428	
Alk		840	
Cl	18,7	528	
SUM		12624	

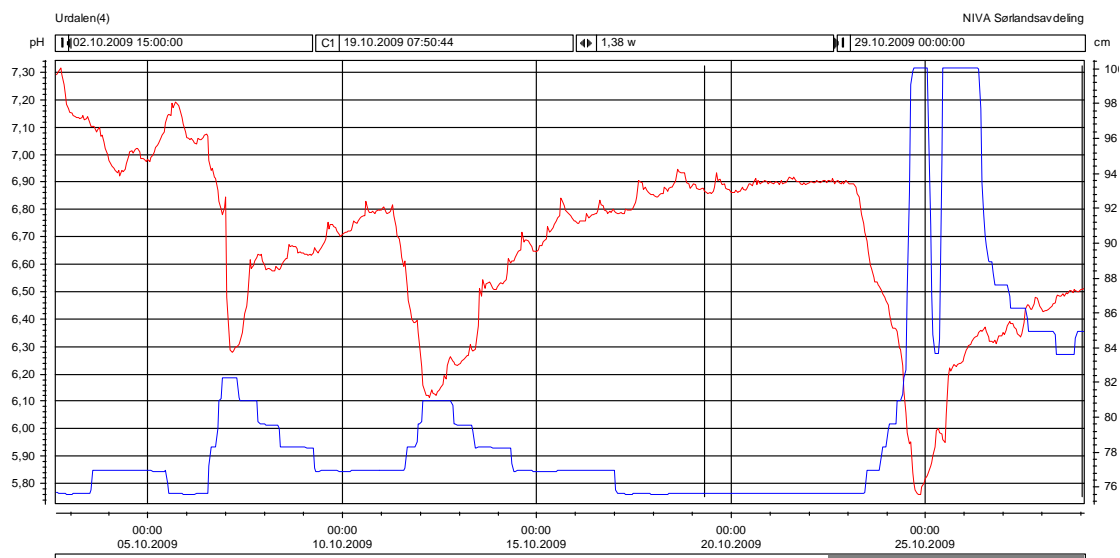


Figure 13. Continuous pH-readings (red line) and water level (step-wise changes) at the NIVA monitoring station in the inlet stream of Lake Urevatn in October 2009. The pH scale is from 5.8 to 7.3. pH drops when the water level, and thereby the water flow, of the stream increases.

Data for precipitates at the deposits

Results from analysis of the white, fluffy precipitate from downstream of M17 (Table 6) is consistent with monitoring data and the runoff data at the deposits. The freeze drying procedure before analysis included some water, probably around 10 ml (not measured). The composition of the water is thereby included in the result for the precipitate. However, by taking into account the chemical composition of the water sampled at the same site and the analytical results for the combined material, the molar composition of the precipitate is as follows in $\mu\text{mol/l}$:

	Date	Al	Ca	Fe	K	Mg	Na	S	Si
M17; white particles	18.05.2009	8066	210	215	31	87	18	1564	170

The precipitate was totally dominated by aluminium and sulphur, and the molar ratio for Al/S is slightly above 5. This may correspond to what we believe is a mix of some amorphous aluminium hydroxide ($\text{Al}(\text{OH})_3 \cdot n\text{H}_2\text{O}$), but mainly amorphous aluminium hydroxysulphate, probably basaluminite ($\text{Al}_4(\text{SO}_4)(\text{OH})_{10} \cdot 5\text{H}_2\text{O}$) or hydrobasaluminite ($\text{Al}_4(\text{SO}_4)(\text{OH})_{10} \cdot 12\text{-}36\text{H}_2\text{O}$), a white precipitate known to form at similar sites elsewhere (D. Kirk Nordstrom, pers. com.). These freshly precipitating hydroxysulphates typically form at pH of 5-5.5 when acid, aluminium-rich water meets water with higher pH.

The concentration of organic carbon was $20.8 \mu\text{g/mg}$, about 10% of the Al by weight. Organic carbon was probably precipitated from dissolved organics of the incoming water from the surrounding catchment. As the amount of Ca, Fe, K, Mg, Na and Si was low, this precipitate was very clean, as also the white colour indicates.

During the flow on October 24, 2009 the runoff from M17 was grey and very turbid, with a concentration of total solids of 221 mg/l and 295 mg/l at the deposit and 200 downstream of the deposit, 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 analysed for Al, Ca, Fe and

S after filtration to get the dissolved ions and after digestion of the unfiltered sample by strong acid to get the total ion content. The content of the particles was found by difference, and the molar composition ($\mu\text{mol/l}$) was as follows:

	<u>Date</u>	<u>Al</u>	<u>Ca</u>	<u>Fe</u>	<u>S</u>
At M17	24.10.2009	1314	0,67	46,7	0,57
200 m downstream of M17	24.10.2009	1097	0,25	168,1	0,37

These particles obviously consisted mainly of precipitated aluminium and iron, probably as amorphous Al-hydroxides and Fe oxy-hydroxides, whereas Ca and S were almost entirely as dissolved ions. This was surprising because erosion of shell sand and the high pH indicated shell sand particles, mainly CaCO_3 .

At the site visit on November 5, 2009 the white, amorphous aluminium hydroxysulphate was again found, but this time only in a tiny stream at the outer side of the dam barrier. A water sample was taken in order to examine the water chemistry characteristics for this formation. Interestingly, pH was 6.86, and only $71 \mu\text{g Al/l}$ was dissolved in the water. All sulphate was balanced by Ca+Mg. It could be that the precipitate had been formed under other circumstances than at this high pH and then been transported out by the high-pH stream.

On November 17, 2009 the limestone gravel which was filled in the lower dam was covered by a gray, very fluffy sediment, which could be made either by sedimented particles transported to the dam during flow or precipitates formed on the limestone surfaces. The particles were collected by a bottle and concentrated after repeated careful disturbances of the gravel, and sedimentation of the stuff in the bottle. In the laboratory the particles were filtered and analysed for metals and carbon, see Table 6.

The inorganic part of the precipitate was dominated by aluminium, as can be seen from the following composition (as $\mu\text{mol/l}$):

	<u>Date</u>	<u>Al</u>	<u>Ca</u>	<u>Fe</u>	<u>K</u>	<u>Mg</u>	<u>Na</u>	<u>S</u>	<u>Si</u>
M17; lower dam	17.11.2009	2961	621	684	203	361	25	288	132

Ten times more Al than S on a molar basis indicates precipitation of mainly aluminium hydroxide rather than aluminium hydroxysulphate. The concentration of Fe and Ca was relatively high, indicating both iron precipitation as amorphous iron hydroxide and presence of CaCO_3 . Some of these ions may also have co-precipitated with the organic material. The concentration of organic carbon was $57 \mu\text{g/mg}$, about 70 % of the Al by weight. The relatively large contribution by organics may explain the grey colour.

Analysis of water chemistry and particles at M17 shows that Al is released from the fill as a result of weathering of the sulphide-bearing rock and precipitates in several ways, probably depending on the pH. The condition for preferred formation of the aluminium hydroxysulphate seems to be pH close to 5.0-5.5, as also seen elsewhere (D. Kirk Nordstrom, pers. com.), whereas the aluminium hydroxide is known to form at pH 6-7.

The high concentrations of dissolved Al both at the fill and at the outlet of Lomtjenn shows that the precipitation is far from complete in the fill area. The very fluffy particles found inside and outside the lower dam, show together with the Al-particles of the turbid water that the precipitated Al-particles are transported downstream. This may be the main reason for the turbidity of Lomtjenn, as also indicated by the lower level of Al-concentrations at the outlet of Lomtjenn after filtering of samples in 2009.

The precipitates described above mixed with microbial slime have probably decreased the permeability in parts of the deposit pile and helped to create less reactive preferential flowpaths.

4.3 S-transport

In order to get an idea of the size and pattern of the weathering rate and sulphur release from the two deposits, sulphate concentrations at the monitoring stations and up- or downscaled daily water flow were used to calculate the net S-transport from the deposits.

Calculations were done as follows: First, transport for each period between sampling occasions was calculated by multiplying the total runoff for the period prior to sampling with the net concentration of sulphur (from measured sulphate). 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 tonnes of S) over the whole period was calculated. The plotted curves were used for description of patterns and calculation of mean S-transport for specific periods.

Increase in net S-transport started at the same time as the construction activities, and after a short lag came a period of a remarkably stable S-transport over time (Figure 14). The stable period lasted for 1.5 years at M17. A significant decrease started in March 2010, but the increase in autumn 2010, after a very dry summer, resembled the pattern of the stable period. It might be that the dry summer resulted in a parallel displacement of the steady increase in accumulated S-transport, as indicated in the figure, but only more data will show if this interpretation is correct.

At M20 the stable period started in May 2007 and lasted throughout the monitoring period, about four years. One might have expected that the redirection of water from the uppermost part of the catchment in summer 2009 should result in reduced weathering. This was not the case, as the S-transport was unaltered by this measure. The steady S-transport over four years strengthens the interpretation of the M17 graph.

Patterns of the S-transport follow runoff patterns, as can be seen from the similar forms of the runoff curve and S-transport curve for both sites in Figure 14. Low water flow in summer resulted in low S-transport, whereas high runoff in autumn and winter gave high transport. This pattern together with the linear increase in accumulated S-transport clearly indicates constant weathering. As the climate of this area changes in the direction of dryer summers and more intense rainfalls, one might expect periods with an extremely toxic potential downstream, such as documented for a similar site at River Moelva, Lillesand, in 2006 (Hindar and Iversen 2006).

The S-transport at M17 in the stable 1.5 years period was 180 kg S/day. At M20 the S-transport in the stable 4-years period was 70 kg S/day. Annual release of S was 65.9 tonnes and 25.7 tonnes, respectively. If the mean S-content of the fill masses was 2 %, these annual release rates correspond to 0.45 % and 0.12 % of the total amount of S in deposits M17 and M20, respectively (Table 8).

Table 8. S in the rock material and S-transport from the deposits.

	M17	M20	
Sulphur in deposit (2% S)	14580	21600	tonnes
S-transport from deposit	65.9	25.7	tonnes/year
S-transport from deposit	0.45	0.12	%/year

The high release rates for sulphur indicate high weathering rates for the sulphide-bearing rock masses. According to D. Kirk Nordstrom, these rates are certainly very reasonable. The rate will probably slow down over time, however.

High weathering rates will result in ongoing consolidation of the rock pile and unstable masses. For M20 stable S-release is a warning sign, as the amount and effect of the liming material in the deposit is supposed to decrease over time. The significance of the treatment basin downstream will therefore increase.

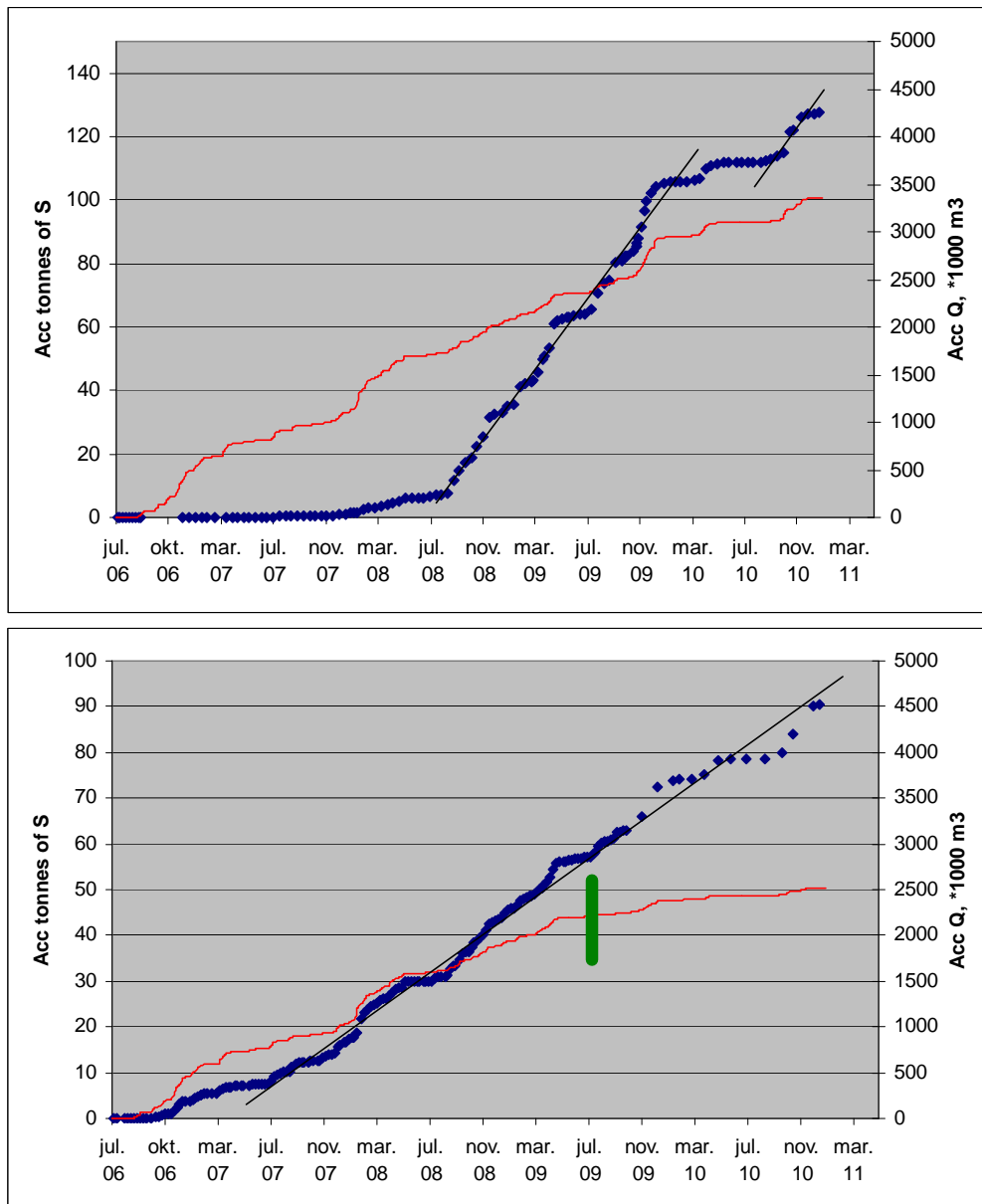


Figure 14. Accumulated net S-transport from M17 (upper) and M20 (lower). Lines are drawn for stable periods. Mean daily S-transport is calculated from the slope of these lines. Accumulated runoff for the catchments of the monitoring sites is also shown. The redirection of water from the upper catchment of M20 in summer 2009 is indicated with the green bar. The result is seen as a decrease in estimated runoff, but not in S-transport.

S-transport in terms of percent of S in the masses per year is about four times higher at M17 than at M20. There is no reason to believe that the masses of M17 are especially rich in sulphide content, and none of the deposits are covered as recommended by Fröberg and Höglund (2004), which make them open for incoming water and air. Relatively more shell sand has been used at M17 according to the figures in Table 1. About half of the shell sand at M17 was used in the dam construction, however, so the actual neutralising capacity of the rock pile itself has been as for M20. The reason for this large difference may therefore be due to other features, as suggested below.

First of all, the surface of M17 is relatively flat in cross-section, and it is shallow with a volume/area ratio for the fill of 5.4 m. M20 has significantly more volume per area, and the ratio is 10 m. M20 therefore has relatively steep slopes in several directions, which may favour a more rapid runoff of incoming rainwater and possibly less access to oxygen for the innermost parts of the pile.

Second, much of the rock in M20 probably fills up the former, deep swamp, and the swamp material may have been lifted upwards during construction of the fill and mixed with other parts of the fill. As slaked lime was mixed into the swamp, this hydroxide may have been relatively important for reducing weathering processes and for increasing the neutralising processes. The lower hydraulic conductivity and probably increased anoxic conditions for part of M20 may have reduced the weathering rate.



Figure 15. Open ditch across M17 on November 17, 2009. The stream in the picture to the left was supposed to cross the fill and enter the pipe in the picture to the right. Instead, it runs into the fill halfway over.

Third, although draining of water from the surrounding terrain was not very well carried out at either of the two deposits, draining at M20 is probably the most successful. Re-direction of 2/3 of the flow through the fill in summer 2009 had no apparent influence on the S-transport, however. At M17, all runoff that should go in open ditches on the sides and in an open ditch across the fill infiltrated back to

the fill (Figure 15). This resulted in good conditions for transport of weathering products to the stream water. Also at M20 a little stream that was supposed to run from a 0.1 km² side-catchment in east across the fill in an open ditch found its way down in the open fill halfway over the fill area.

5. New measures at M17

5.1 Passive and active measures

Adequate measures are described in reports from the Swedish MiMi-programme (Mitigation of the environmental impact from Mining waste; Fröberg and Höglund 2004), a similar programme in Australia (Australian Government 2007), and from several disposal sites for sulphuric rock material elsewhere.

Measures to reduce the possibilities for biological damage may be grouped and seen as five steps in a ladder you shouldn't climb too far up. The first steps in the ladder are passive measures. They have relatively low costs and demand a minimum of operation over time. The last steps are active measures with high technical complexity and costs. They demand a long-term commitment for operation.

An important aim of passive measures is to avoid/reduce weathering and thereby the sulphate production. Short after the sulphuric rock material is deposited, the main measure is therefore to reduce the availability of water, moisture and oxygen. Use of shell sand and other carbonates is also recommended. In theory several possibilities are at hand:

- Soak the deposit in fresh water or the sea to reduce availability of oxygen from the air. Wave dampening to prevent erosion of the deposited material, and liming to facilitate precipitation of metals is recommended.
- Increase the ground water table artificially to ensure that the material is permanently in water-saturated environment with reduced access to oxygen.
- Cover land deposits fully with adequate masses; clayey till masses with low permeability covered with protective masses.
- Cover land deposits fully with impermeable membranes to prevent rain from entering and reduce oxygen availability.
- The slope of deposit surfaces should be constructed for rapid runoff of rain and snowmelt water.
- Clean water from coverings and unaffected water should be drained away from the deposit and treatment areas.

If passive measures are inadequate, active measures, i.e water treatment, have to be considered to avoid adverse effects in streams and lakes. Water treatment of runoff from deposits implies long-term operation of a treatment plant, which tend to be costly. Operation of a treatment plant also demands for a long-term commitment and agreement between involved parties.

Several possibilities exist for water treatment. Addition of carbonates reduces the acidity and results in precipitation of metals. The advantage of adding calcite (CaCO₃) is that too high pH is prevented due to the buffering character of carbonates. But very acid water may demand for use of slaked lime (Ca(OH)₂). Dosing has to be controlled to avoid very high pH.

Addition of dissolved sodium silicate (NaSi; mol-ratio $\text{SiO}_2:\text{Na}_2\text{O}$ of 3.4) has been used in Norway (Teien et al. 2009). This is due to its rapid and strong binding to Al-ions and therefore rapid detoxification of aluminium. If active water treatment is necessary, this method should be considered. Water treatment, if based on precipitation of metals, will produce a sludge that has to be deposited properly. Experience with NaSi is that dissolved Al binds to Si, but that most of the Al stay in solution. Si-metal particles may also be formed, however.

Treatment basins, as already in function at M17 and M20, have elements of both passive and active measures. The basins are shallow, filled with shell sand or limestone gravel, and are dimensioned to deacidify runoff from the deposits over time. As the shell sand and gravel is dissolved due to the neutralisation reactions, but also clogged due to precipitation on the surfaces, the basins must be maintained. Precipitates have to be removed and used reactive material has to be replaced by fresh material with a frequency that depends on the chemical processes in the basins.

5.2 Procedure

The construction of M17 and measured runoff-water chemistry shows a potential and need for improvement of the water quality. Experiences from Lake Langedalstjenn are that sulphate concentrations decreases over time. This might be partly due to coverings of asphalt and roof constructions, but additional measures than at the deposit were needed to reach low target Al-concentrations.

According to recommendations from the Swedish MiMi-project, active measures, such as water treatment, will be needed if the design of the deposit is inappropriate, and/or if the passive measures at the deposit fail to give a sustainable water quality. The costs of water treatment might be substantial, partly due to the long-term commitment to such measures.

By April 2011, CJV suggested new measures at M17. They can be classified as mainly passive, see description below, but as outlined above, they also have elements of active measures. In order to facilitate a process towards a cost-efficient solution, NIVA was asked to document the present relation between pH and the concentration of toxic aluminium and suggest a procedure for achieving, if possible, good ecological status of Lake Lomtjenn.

Before we turn to this procedure, it should be pointed out that M17 was suggested (by NIVA and Jordforsk; Hindar and Roseth 2003) and later placed in Mannfalldalen, rather than in the neighbouring valley in the west with the sea-trout river Fjellaldalsbekken. This was partly because Lomtjenn and its outlet stream represented a relatively small catchment with anticipated less value than Fjellaldalsbekken. Low-quality runoff from M17 would therefore be less harmful. However, according to the EU Water Framework Directive (EU 2000; Directive 2000/60/EC), all water bodies should have or reach good ecological status.

On the other hand, and according to the same directive, "Member States may designate a body of surface water as artificial or heavily modified". It can be argued that Lake Lomtjenn is a candidate. Arguments are that E18 and deposition of sulphuric rock in deposits is a substantial alteration of the catchment hydromorphological characteristics (Article 4(3)(a)). Also, according to Article 4(3)(b), technical feasibility or disproportionate costs for countermeasures could be basis for this designation. Such designation and the reasons for it shall be specifically mentioned in the river basin management plans required under Article 13 of the EU WFD.

Based on the EU WFD or not, we suggest a principal sketch as basis of the procedure (Figure 16). The sketch is based on:

- the documented Al vs pH relationship at the site

- relative costs for reducing the Al-concentrations from one level to the next
- an acceptable cost limit

The sketch shows the effect on Al of increasing pH. The target Al-concentration could be the critical concentration for e.g. the sensitive brown trout (*Salmo trutta*), which is at 0.05-0.1 mg/l for the labile Al fraction (LAI). It could also be the Al-concentration which allows measures directly in the recipient. Lake Langedalstjenn is a good example here. NIVA did not recommend liming directly in the lake in 1992 (Hindar et al. 1992) due to very high Al-levels (several milligrams per liter), and fear for a green lake after substantial Al-precipitation. As the Al-concentrations decreased significantly over 15 years and down to about 0.5 mg/l, NIVA suggested liming. Successful liming was carried out. In this case Al at 0.5-1 mg/l represented a target concentration, which made it possible to take the last step towards the critical Al concentration for brown trout.

Costs for achieving a specific pH and thereby a target Al concentration might be hard to calculate, but could be based on experience at the specific site.

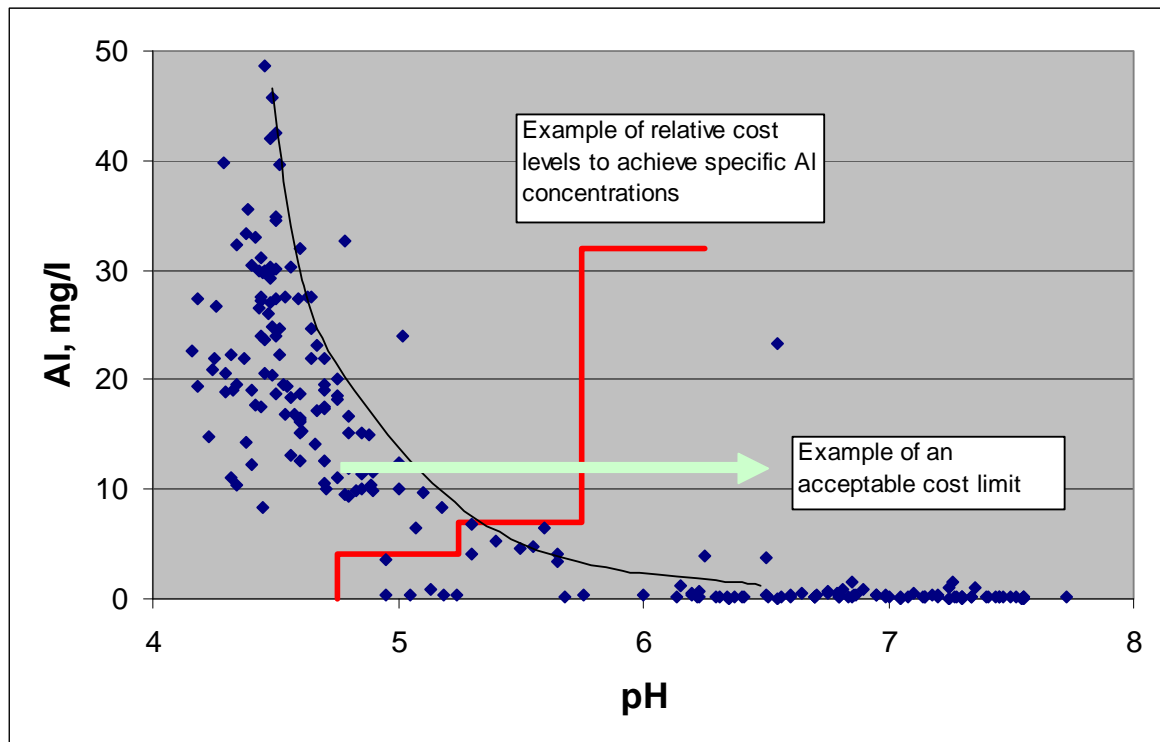


Figure 16. Principal sketch showing the relation between measured pH and aluminium concentrations (data not from M17/Lomtjenn). The black line, drawn by hand, is the operational Al-pH relationship, which will be the basis for cost evaluation. The red line is an example of relative cost levels for reducing pH, and thereby Al to specific concentrations. An acceptable cost limit should be established.

An acceptable cost limit might be difficult to agree upon for both the constructor and the environmental authorities. Alternative use of money in this case could be in the form of a fund paid by the constructor and owned by e.g. Lillesand municipality. The fund could be used to improve the environment elsewhere with higher benefit/cost ratio. The steps of the procedure itself are relatively straight-forward, as shown in Figure 17.

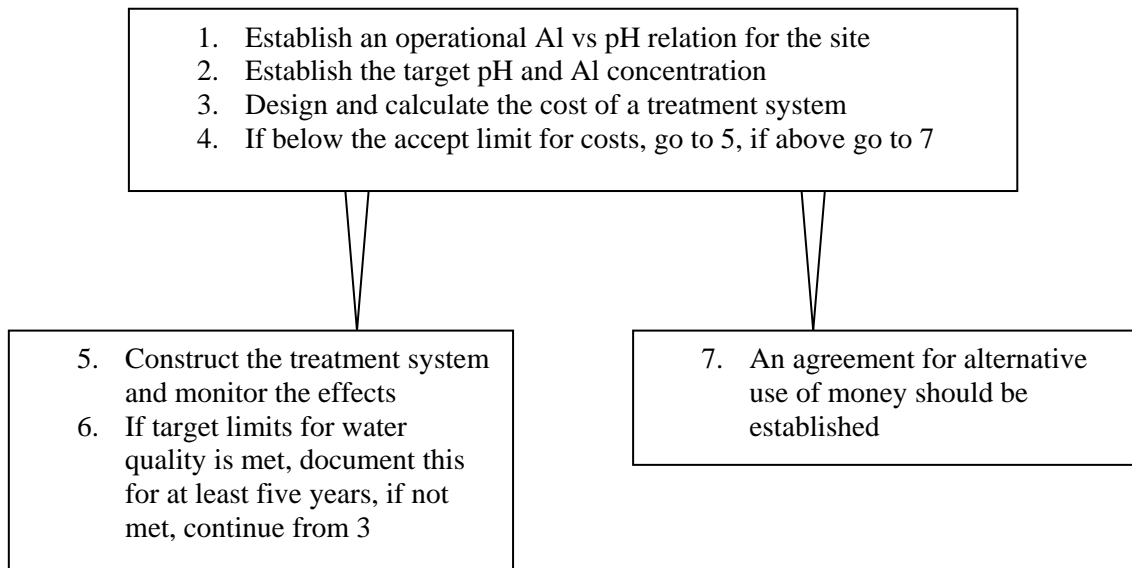


Figure 17. Procedure for achieving good ecological status, if possible, within an acceptable cost limit.

5.3 Design of new measures

By May 2011 CJV presented a new design for M17 (Figure 18). CJV suggested to re-direct unaffected water from the catchment through ditches along the deposit. Also, overflow from the northern deposit area will be collected in pipes and directed to these ditches during heavy rainfall. This will significantly reduce the water entering the deposit and significantly improve the possibility to treat the water from the deposit. Crosswise, near to the foot of the deposit, an active barrier with shell sand will be built. This barrier will force water through the shell sand. Two basins, one for water retention and one for water treatment, will be constructed immediately downstream of the deposit, and the deposit itself will be covered with a thin layer of recultivation soil.

The construction of treatment basin 2 (grey coloured basin in Figure 18) in row with treatment basin 1 was suspended for the following reasons given by CJV:

- Laboratory tests made by Prof. Dr. Winfried Schmidt of coarse-grained lime stone material showed good and quick results by increasing the pH-value from about 4 to 6 in less than 10 minutes. These tests were confirmed with already used limestone material.
- Studies of mine-water treatment showing that longer retention time of acidic water in the lime stone cause more extensive precipitation on the grain surfaces, thereby reducing the reactivity.
- Results from on site bucket tests at another deposit along E 18 (M 15/16) and M17 with “fresh” and “used” reactive material also showed quick reaction (S. Winter and M. Adams at CJV).

Following these works pH was expected by CJV to be above the present accept limit of pH 4.7 from the discharge permit. If the monitoring results after reconstruction of the deposit are not acceptable, basin 2 will be constructed.

5.4 Target water quality and anticipated effects of new measures

In the period elapsed from peak sulphate concentrations in 2008-2009, the sulphate concentrations have been reduced to 100-300 mg/l (Figure 9), indicating that the weathering rate of the deposit has decreased. The combination of less sulphate concentration and the documented liming effect (Table 3) has resulted in a decrease in Al to below about 5 mg/l (Figure 9).

The new measures at M17 may reduce the sulphate transport even further, due to less water through the deposit. But the sulphate concentrations might stay at present level or even increase. This depends on the relationship between the weathering rate and the concentration of weathering products, following re-direction of water. Based on the data for M20 after re-direction of water, reduced weathering rate can not be guaranteed.

The effects of deacidification at the barrier and in the treatment basin will decide the resulting pH and Al-concentration downstream. Based on experience with measures at M17 in 2009 and 2010, it is not easy to calculate or foresee the resulting pH, but pH will increase and Al decrease further.

We suggest a critical Al concentration for brown trout in Lake Lomtjenn of about 0.1 mg/l, as labile Al (LAl). This is the target Al concentration, which may be reached at a relatively high pH. Before the construction period, pH could be down to 4.8 and LAl close to this target. The change in Al vs pH relation, as seen in Figure 19, is due to the change in sulphate concentrations from 4-5 mg/l in 2006 to 100-400 mg/l in 2008-2011, see also paragraph 1.2 and Figure 1.

According to Figure 19, pH below 4.5 may result in Al higher than 15 mg/l. This is not sustainable. At the present accept limit of pH 4.7 from the discharge permit, Al may still be up to 6 mg/l. 1 mg Al/l has been measured for water with pH 5.2.

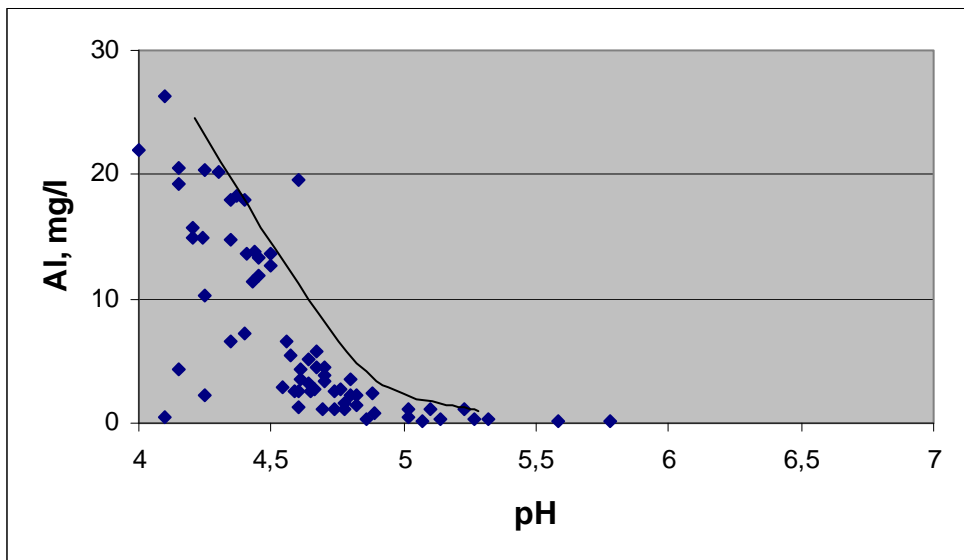


Figure 19. The Al-pH relationship at the outlet of Lake Lomtjenn during and after the construction period. The black line is drawn by hand, and indicates an operational Al-pH relationship. If a certain pH is reached, Al will probably not be higher than indicated by the line.

Increase of pH to 5.5 may result in target Al-concentrations in the lake. Few data exist at this pH level during and after the construction period, and the basis for estimation of the resulting Al-concentrations is poor, however. Data from deposit M15/M16 at Stordalen east of Lillesand city show that pH should

be above 6.0 to reach the target Al concentration. The size of M15/M16 is about twice that of M17, and the sulphate concentrations mainly above the 100-400 mg/l range and up to 1000 mg/l. For the time being, we suggest use of the existing relationship at M17.

If approaching the target Al concentration of 0.1 mg/l as labile Al (LAl), Al-speciation is important. LAl is the sum of toxic, inorganic monomeric Al-species, typically the cations Al^{+++} , $Al(OH)^{++}$ and $Al(OH)_2^+$, which mainly exist at pH below 6. LAl is operationally defined as Al ions retained in a specific cationic exchange resin. It is calculated as the difference between reactive Al of the water and reactive Al of the ion-exchanged water. Non-labile Al-ions contributes to the reactive Al and total Al concentration.

At Al-concentrations over time below 1 mg/l at the outlet of Lake Lomtjenn, one might consider direct water treatment of the lake to reduce LAl to target levels. As the water retention time is very short, probably only a couple of weeks, lake liming has to be repeated several (5-10) times each year to give stable water quality. For the time being, this is not recommended as a solution, due to logistics, labour and to the fact that a relatively large fraction of ordinary limestone powder (e.g. Miljøkalk NK3, particle diameter: 95 % < 0.12 mm) will sediment and accumulate at the lake bottom.

However, as experience with lake liming under similar circumstances (acid and high Al concentrations) are very limited, the effects of lake liming at high Al concentrations could be worth while to test. Lake liming, with commercially available slurry of very fine-ground limestone powder (e.g. Hustadmarmor Biokalk 75, particle diameter: 95% < 0.01 mm), could be used in order to significantly reduce the sedimentation of particles. In order to optimize the dose, depth at several sites should be measured in order to calculate the lake volume. The present (by May 2011) situation at Lake Lomtjenn will probably not be worsened by a test treatment.

An alternative to lake liming is adding limestone powder or liquid sodium silicate on a continuous basis to the inlet stream. Precipitation of Al in the stream-bed will probably not be a problem, as the Si-Al complexes are small and tend to stay in solution. These complexes will be transported to the lake, however, and might result in a greenish colour due to spreading of light from the particle surfaces. To avoid sedimentation of limestone particles, the same slurry as mentioned above should be considered.

The slurry or the sodium silicate may be stored in a tank, and could be mixed directly into the stream. Dosing should be on a volume proportional basis, however, and this requires both a weir in the stream, a dose control system, and access to electricity.

In line with the proposed procedure, the suggested new measures at M17 should be constructed and the results at the outlet of Lake Lomtjenn monitored. The monitoring period could be used to discuss and make decisions related to additional measures and costs if acceptable.

6. Conclusions

Runoff from two deposits for sulphide-bearing rock along the highway E18 Grimstad-Kristiansand has been examined. The monitoring period for water chemistry was from July 2006 until April 2011, including one year before start of the construction works.

Runoff downstream of the deposit M17 in the small valley Mannfalldalen became acid (pH 4.0-4.6) and had high concentrations of dissolved aluminium (10-20 mg/l) in 2008-2009. No fish were caught during net fishing in the downstream situated Lake Lomtjenn in May 2009. The runoff from deposit

M20 in Urdalen did not acidify as much during the monitoring period. Differences were partly due to construction characteristics.

Attempts to neutralise the acid runoff from M17 in 2009-2010 were not sufficient to secure acceptable water quality of Lake Lomtjenn. Water chemistry at the outlet was characterised by pH down to 4.5 and concentrations of dissolved aluminium in the range 0.1-6.5 mg/l from summer 2009 until April 2011. Although pH increased and Al decreased in 2010 and 2011, the water is still toxic to fish and the lake has not recovered.

Runoff data from the nearby monitoring site Birkenes made it possible to quantify the net mass transport of sulphur from M17 and M20 based on the water chemistry data and up- or down-scaled data for daily runoff. By accumulating the S-transport for each period between the sampling occasions, it was found that the initial net S-transport was linear over time. This indicated that the weathering rates within the deposits were relatively constant.

By assuming that the mean S-content of the rock in the deposits is 2 %, we found that the annual release rate of S could be about 0.45 % of the total S-content of the deposit at M17 and 0.12 % at M20. These weathering rates will result in ongoing consolidation of the rock pile and unstable masses.

Recovery of a healthy Lake Lomtjenn will require new measures at the deposit site, such as suggested by May 2011. The pH limit of the discharge permit of pH > 4.7 will probably be reached, but the Al-target of 0.1 mg/l, as suggested here, probably not. The water will still be toxic for fish.

In line with the proposed procedure for achieving good ecological status, the suggested new measures at M17 should be constructed and the effects of these attempts downstream of deposit M17 should be monitored. The monitoring period could be used to discuss and make decisions related to additional measures and costs if acceptable.

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