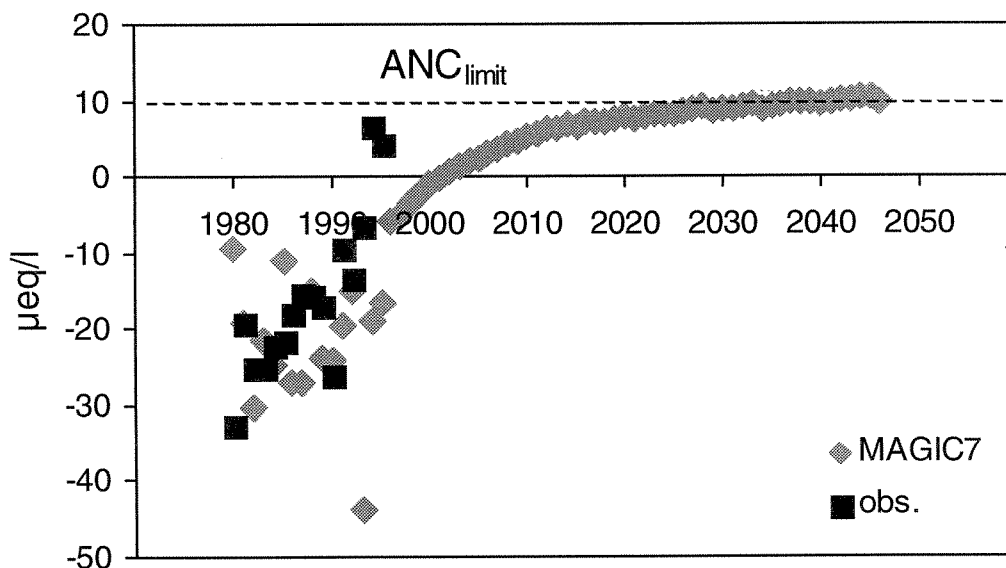


Gap closure: use of MAGIC model to predict time required to achieve steady-state following implementation of the Oslo protocol



Miljøverndepartementet
Fagrapport nr. 100

Tovdal ANC



Naturens Tålegrenser

Programmet Naturens Tålegrenser ble satt igang i 1989 i regi av Miljøverndepartementet. Programmet skal blant annet gi innspill til arbeidet med Nordisk Handlingsplan mot Luftforurensninger og til pågående aktiviteter under Konvensjonen for Langtransporterte Grensoverskridende Luftforurensninger (Genevekonvensjonen). I arbeidet under Genevekonvensjonen er det vedtatt at kritiske belastningsgrenser skal legges til grunn ved utarbeidelse av nye avtaler om utslippsbegrensning av svovel, nitrogen og hydrokarboner.

En styringsgruppe i Miljøverndepartementet har det overordnede ansvar for programmet, mens ansvaret for den faglige oppfølgingen er overlatt en arbeidsgruppe bestående av representanter fra Direktoratet for naturforvaltning (DN), Norsk polarinstitutt (NP) og Statens forurensningstilsyn (SFT).

Arbeidsgruppen har for tiden følgende sammensetning:

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Andre Kammerud - SFT
Else Løbersli - DN
Steinar Sandøy – DN
En representant fra Norsk Polarinstitutt**

Styringsgruppen i Miljøverndepartementet består av representanter fra avdelingen for naturvern og kulturminner, avdelingen for vannmiljø, industri- og avfallssaker og avdelingen for internasjonalt samarbeid, luftmiljø og polarsaker.

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Abstract

Reduction of sulphur deposition causes recovery of acidified surface waters. Processes in the catchment delay recovery. The acidification model MAGIC was applied to the Vikedal and Tovdal rivers in southern Norway. Water response is delayed by about 10 years at both rivers; delay is due to release of old sulphate at Vikedal, and cation exchange at Tovdal. Assuming that the Oslo protocol is fully implemented by the year 2010, about 60% of the increase of ANC will occur in the next 13 years (1997-2010) with a new steady-state reached by about 2046. Liming requirement for both rivers will be reduced to about 90% of present-day levels by the year 2010 and to about 70% by the year 2046. One of the main purposes of the Norwegian national monitoring programme is to provide documentation of changes in environmental quality due to long-range transported air pollutants. Modelling applications such as this clearly show that the data fill this purpose.

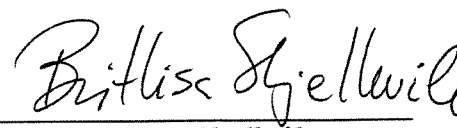
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**Gap closure: use of MAGIC model to predict time
required to achieve steady-state following
implementation of the Oslo protocol**

Richard F. Wright

Arne Henriksen

Preface

This work was supported by “Naturens tålegrenser” through contract number 981920 with SFT (Norwegian Pollution Control Authority). Arne Henriksen NIVA was project manager. Brit Lisa Skjelkvåle NIVA provided data files from the Norwegian monitoring programme, Arne Stuanes NISK provided the original soils data, Sverre Krog NVE provided annual discharge data, Øyvind Kaste and Atle Hindar NIVA provided information on liming. We thank Øyvind Kaste and Brit Lisa Skjelkvåle for useful comments on the manuscript.

Oslo, December 1998

Richard F. Wright

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Summary

International agreements on emissions of acidifying compounds to the atmosphere have resulted in decreased deposition of sulphate in Norway. When the Second Sulphur Protocol (Oslo protocol) is fully implemented by the year 2010, sulphate deposition will have decreased by about 60% relative to maximum levels in 1980. The recovery of surface waters from acidification is subject to delays due to processes in the terrestrial catchment such as retention and release of sulphate and cation exchange. As a result there is a gap between the time when emission controls and reductions in deposition are in place and when surface waters reach a new steady-state with respect to the new level of deposition.

Process-oriented dynamic models provide a tool by which these delays can be quantitatively evaluated. Here we apply MAGIC (Model for Acidification of Groundwater In Catchments) to the Vikedal and Tovdal rivers of southern Norway to demonstrate the lag times in recovery from acidification following implementation of the Oslo protocol in the year 2010. Both rivers are currently limed, and we use the model to estimate future liming requirement.

The model calibrations entailed compilation of deposition, runoff and soils data characteristic for the entire catchment. These data come largely from SFT's national monitoring programme for long-range transported pollutants. The data period is 1984-1997 for Vikedal, and 1980-1996 for Tovdal. Deposition data collected at one or two stations within the catchment were scaled such that Cl deposition equalled runoff for the entire data period, and SO₄ deposition equalled runoff for the first 3 years of the data period. Calibration proceeded by adjustment of model parameters until simulated agreed with observed concentrations of major ions in river water and soil for the first 3 years of the data period.

At Vikedal retention and release of sulphate is substantial and delays recovery by about 10 years of the river water from acidification. Acid neutralising capacity (ANC) is predicted to increase from present-day levels of about $-5 \mu\text{eq/l}$ to $+4 \mu\text{eq/l}$ in year 2010 and reach a new steady-state level of $+11 \mu\text{eq/l}$ by the year 2046. About 60% of the increase occurs during the first 13 years, with the remaining 40% coming during the following 37 years. At Tovdal change in base cation concentrations due to cation exchange appears to be the major cause of delay in recovery. Sulphate adsorption is minor compared to Vikedal. The delay in recovery of ANC at Tovdal is similar to that at Vikedal, with ANC predicted to increase from present-day levels of about $-5 \mu\text{eq/l}$ to $+5 \mu\text{eq/l}$ in year 2010 and reach a new steady-state level of $+10 \mu\text{eq/l}$ by the year 2046.

One of the main purposes of the Norwegian national monitoring programme is to provide documentation of changes in environmental quality due to long-range transported air pollutants. Modelling applications such as this clearly show that the data fill this purpose. The long-term monitoring provides a solid data base on which to calibrate the model. The data nicely constrain the model calibration such that predictions into the future can be made with confidence. The weakest part of the calibration lies in the paucity of soil data. These prognoses for the future assume that there is no change in deposition or leaching of nitrogen.

Liming requirements for Vikedal and Tovdal will decrease in the future due to recovery of the water following reduced acid deposition. Because of the lag times, however, the liming requirement will still be 95% and 85% of 1996 levels in the year 2010 for Vikedal and Tovdal, respectively, and decrease to 70% for both rivers by the year 2046. These estimates assume that the pH (and ANC) targets for both rivers will be the same as present. Less ambitious targets can be used in the future as the rivers recover, levels of inorganic aluminium decrease, and the intensity and duration of acid episodes tapers off.

1. Background

Deposition of sulphur in Norway has decreased substantially since 1980 as a result of international agreements on reduction of sulphur emissions in Europe. The first sulphur protocol called for a 30% reduction by 1993 based on emissions in 1980, and the second protocol (Oslo protocol of 1994) calls for further reductions by the year 2010. The prognosis is about 60% reduction of sulphur deposition in southern Norway by the year 2010 relative to 1980 (UN/ECE 1994).

Measurements of wet and dry deposition conducted by NILU as part of SFT's national monitoring programme for long-range transported pollutants (Johannessen 1995) show that sulphur deposition has decreased by about 40% from peak levels in the late 1970's to the mid-1990's (Tørseth and Manø 1997).

Acidification of freshwaters in southern Norway has decreased as a result of this decrease in acid deposition. Since 1980 concentrations of sulphate and inorganic aluminium have decreased, and pH and acid neutralising capacity (ANC) have increased (SFT 1998). There has been a clear widespread improvement of water quality for fish and other acid-sensitive organisms.

The question now is what further improvement in water quality can be expected with the future reductions in sulphur deposition, and how rapidly will these improvements occur? Quantification of the temporal aspects of acidification requires the use of time-dynamic acidification models. Such models take into account the time lags between changes in deposition and changes in surface water quality caused by processes in terrestrial catchments and in the watercourses themselves. Such processes include simple storage of water, retention and release of anions by adsorption in the soil, oxidation-reduction processes, and cation exchange in soils.

MAGIC (Model for Acidification of Groundwater In Catchments) (Cosby et al. 1985a,b) is such a process-oriented dynamic model, and has been widely applied to a variety of catchments throughout the world. The credibility of the model has been built by applications of several types including comparison with historical acidification records derived from paleolimnologic studies (diatoms in lake sediments), trends from long-term monitoring, and catchment-scale manipulation experiments with acid addition and exclusion.

Here we calibrate MAGIC to data from the Tovdal and Vikedal Rivers, two acidified rivers in southern Norway (Figure 1), and then predict acidification status expected over the next 50 years given the expected reductions in sulphur deposition of the Oslo protocol. Further we estimate the future requirement for liming of these rivers compared with the present-day levels.

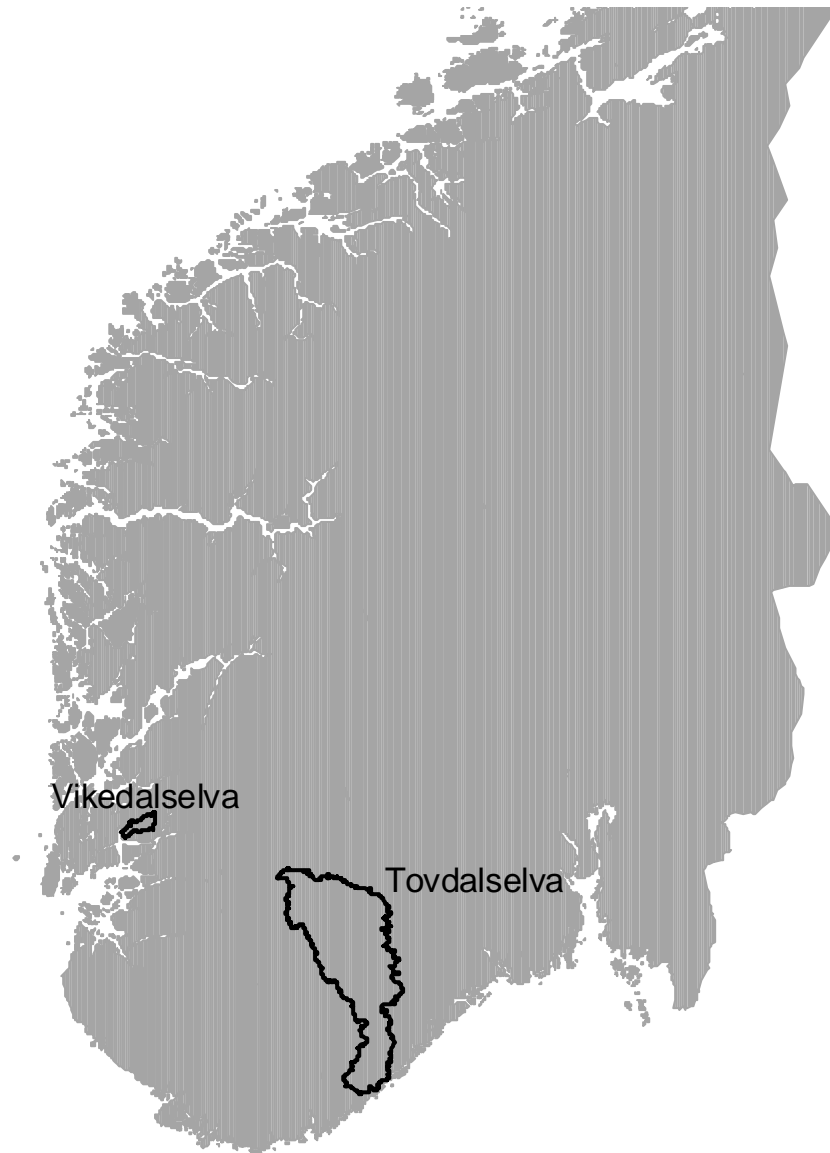


Figure 1. Map of Norway showing location of Vikedal and Tovdal river basins.

2. The MAGIC model

MAGIC is a lumped-parameter model of intermediate complexity, developed to predict the long-term effects of acidic deposition on surface water chemistry (Cosby et al. 1985a; Cosby et al. 1985b). The model simulates soil solution and surface water chemistry to predict average concentrations of the major ions. Time step is month or year. MAGIC calculates for each time step the concentrations of major ions under the assumption of simultaneous reactions involving sulphate adsorption, cation exchange, dissolution-precipitation- speciation of aluminium and dissolution-speciation of inorganic carbon. MAGIC accounts for the mass balance of major ions in the soil by bookkeeping the fluxes from atmospheric inputs, chemical weathering, net uptake in biomass and loss to runoff.

At the heart of MAGIC is the size of the pool of exchangeable base cations in the soil. As the fluxes to and from this pool change over time owing to changes in atmospheric deposition, the chemical

equilibria between soil and soil solution shift to give changes in surface water chemistry. The degree and rate of change of surface water acidity thus depend both on flux factors and the inherent characteristics of the affected soils.

Data inputs required for calibration of MAGIC comprise input and output fluxes for water and major ions, soil chemical and physical characteristics, and net uptake fluxes for vegetation.

There are two major mechanisms by which the response in surface water can be delayed following changes in acid deposition. Retention and release of sulphate is the first. Since sulphate is the major strong acid anion in most acidified surface waters (apart from chloride from seasalts in near-coastal areas) delays in response of sulphate concentrations in surface waters to reductions in sulphate deposition will result in lag times in response of ANC, pH and inorganic Al concentrations. Adsorption/desorption and reduction/oxidation of sulphate are 2 processes active in most soils to a greater or lesser degree.

Sulphate adsorption is included in MAGIC and described by the traditional Langmuir isotherm, in which

$$E_s = \frac{E_{\max} * (SO_4)}{[C + (SO_4)]}$$

where E_s is the adsorbed sulphate on the soil (meq/kg), (SO_4) is the concentration of dissolved sulphate in soil solution (meq/m³), E_{\max} is the maximum adsorption capacity of the soil (meq/kg) and C is the half-saturation constant (meq/m³) (Cosby et al. 1986). The half saturation constant is defined as the concentration at which the amount of sulphate adsorbed on the soil is half the maximum capacity (Figure 2).

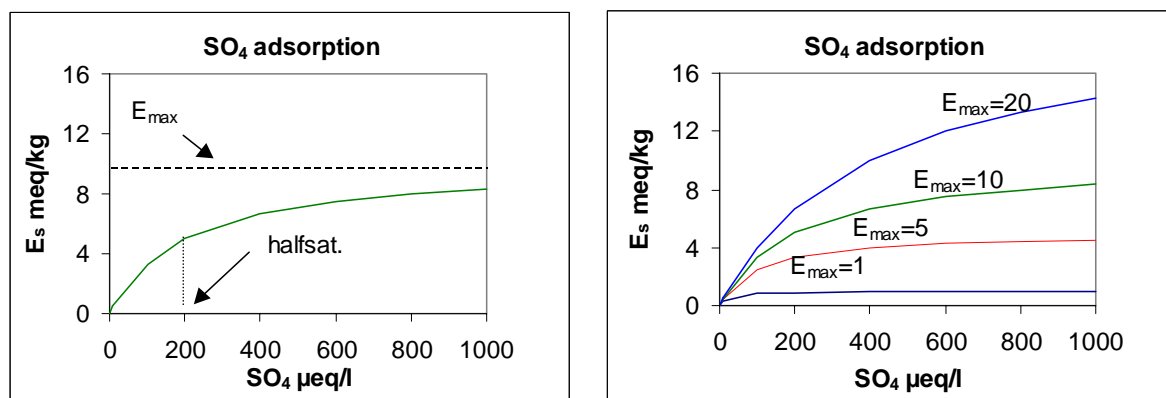


Figure 2. Sulphate adsorption in soil as described by Langmuir isotherm (left panel). Right panel shows curves for different values of E_{\max} (from Cosby et al. (1986)).

The effect of sulphate adsorption is to damp changes in sulphate inputs. Thus soils with high sulphate adsorption respond slowly to changes in sulphate deposition. The key here is the adsorption capacity relative to the rate of atmospheric deposition of sulphate. For a soil characteristic of say Birkenes of 0.5 m depth, bulk density of 1000 kg/m³, and sulphate adsorption capacity of 1 meq/kg will be able to store maximum 500 meq/m² SO₄. This is equivalent to approximately 5 years of sulphate deposition at 1990 levels. Sites with high sulphate adsorption capacities (5-20 meq/kg) can thus retain many decades of sulphate deposition, and result large lag times between onset of acid deposition and

response in surface water. Such lag times can be as much as several decades, as is the case for sites in, for example, White Oak Run, Virginia, in the southeastern United States (Cosby et al. 1985b) and Lange Bramke (Lükewille et al. 1995) and Solling (Alewell et al. 1995) in central Germany.

Sulphate adsorption is large in soils with high amounts of iron and aluminium sesquioxides. Such soils are typically old, highly weathered yellow and red clay-rich soils found in tropical and temperate regions. Areas with geologically young soils, such as those formed since the last ice age usually have low amounts of iron and aluminium sesquioxides and thus low sulphate adsorption capacities. Norway and indeed most of Scandinavia has this type of young soil.

Nevertheless sulphate adsorption can be significant even in young soils in Norway. At the acid-addition experiment conducted at Sogndal as part of the RAIN project, for example, sulphate adsorption measured in the soil was sufficient to retain nearly half the added sulphate during the 11 years of the experiment (Wright et al. 1994).

Retention of sulphate can also be caused by sulphate reduction and storage in the soil in the form of various forms of reduced sulphur, such as iron sulphide. This requires anaerobic environment, such as waterlogged soils or peaty soils. Reduced sulphur can be released following oxidation to sulphate, which occurs when the soil dries out or when oxygen-rich water flows into the stagnant anoxic zones.

Redox processes clearly are in action in Norwegian catchments and act to retain and release sulphate over time scales of weeks to months. Large peaks in sulphate concentrations are typically seen in the first runoff following long dry periods in the summer. This pattern is regularly seen, for example, at Birkenes (Christophersen and Wright 1981). It has also been hypothesised that redox processes can cause retention or release of sulphate over the long-term in response to changes in temperature and precipitation (Dillon and LaZerte 1992).

Cation exchange is the second major process by which recovery following reduction in sulphate deposition can be delayed. The cation exchange complex on soils is associated with organic matter and clays. Major cations involved include the base cations (Ca, Mg, Na, K), ammonium (NH₄) and the acid cations (H⁺ and Alⁿ⁺). There are several conceptual models of cation exchange in soils (Reuss 1983; Reuss and Johnson 1986). The most widely used in acidification models is the Gaines-Thomas expression, which for the case of exchange between calcium and aluminium is written as:

$$K_s * \frac{(Al^{3+})^2}{(Ca^{2+})^3} = \frac{E_{Al}^2}{E_{Ca}^3}$$

where K_s is the selectivity coefficient, the parentheses denote the molar activities in the solution phase, and E_{Al} and E_{Ca} are the fractions of calcium and aluminium on the exchange complex (equivalents per equivalent exchange capacity charge). Usually the amount of cations stored on the exchange complex is much larger than the amount in soil solution such that the right-hand side of the equation is constant, at least over periods of a few years or less, and thus cation exchange will tend to hold the ratio of Al to Ca in soil solution constant. Similar equations can be written for exchange between all the other cations.

The base cation storage on the exchange complex is depleted by acid deposition. Hydrogen ion will exchange with base cations either directly or indirectly by first bringing aluminium into solution which in turn can exchange with base cations. The base cation pool is replenished by atmospheric deposition of base cations of, for example, seasalt origin, and by inputs from weathering of soil minerals.

Soils in Norway have generally developed on moraine left from the last ice age about 10 000 years ago. Parent material of granitic or gneissic mineralogy is resistant to chemical weathering. Such

conditions produce soils with low base saturation, that is the fraction of base cations on the soil cation exchange complex is low. Typically base saturation in the mineral horizons of such soils is less than 20%, and most of the exchange complex is occupied by aluminium.

Deposition of acid from the atmosphere strips base cations from the soil, but since most of the exchanger is occupied by aluminium, acid inputs can also bring aluminium into soil solution, provided that the pH of soil solution is sufficiently low. This is the reason that surface waters in areas with soils of low base saturation are sensitive to acid deposition and become acidified, have low pH elevated concentrations of inorganic Al and negative ANC. All this is of course conditional on sulphate is mobile and not retained in the soil.

The fraction of the mobile sulphate accompanied by base cations (rather than acid cations) is in fact the famous Henriksen F-factor (Henriksen 1984; Brakke et al. 1990). At localities with low F-factors very little stripping of base cations occurs, whereas at sites with high F-factor large amounts of base cations come from the exchange complex.

This cation exchange process can produce delays in the response of ANC, pH and inorganic aluminium in surface waters to reductions in acid deposition. Sites with low F-factor and very little base cation exchange should show greater response in ANC relative to sites with higher amounts of base cation exchange. On the other hand, it is the sites with low F-factor that are generally the most acidic and have the lowest ANC. ANC may change more in absolute terms, but still not enough to reach values above the critical limit.

Cation exchange and aluminium dissolution are fully included in MAGIC. The model is thus well suited to predict the response of concentrations of base and acid cations in surface waters to changes in acid deposition.

There are of course other processes that can delay the response of freshwaters to reductions in acid deposition. Nitrogen retention in the catchment and water can play an important role at sites where nitrate comprises a significant fraction of the strong acid anion sum, or where there is a risk of increasing nitrate leakage from the catchment in the future (due to nitrogen saturation). In Norway nitrate must be considered especially in southwestern parts of the country, where N leakage in the form of nitrate may be as large as 30-40 % of N deposition. Nitrogen processes are included in an empirical manner in MAGIC and more thoroughly in a recently developed new version, MAGIC7 (Wright et al. 1998).

Finally land-use changes may affect recovery of freshwaters. In particular forest management practices such as clear-cutting and fertilisation are of importance. These can also be accommodated by MAGIC, *albeit* in an empirical manner.

3. Vikedal and Tovdal rivers

3.1 Site description

3.1.1 Vikedal

The Vikedal River is a small salmon river located on the southwestern coast of Norway (Figure 1). The catchment area (above the gauging station at Holmen) is 115 km², and stretches from high mountain terrain over 1000 m above sealevel to the low-lying salmon reaches in the lower 10 km. The bedrock geology is a mixture of Precambrian granitic and gneissic rock with Cambro-Silurian phyllites. Overburden consists of glacial moraine and glacio-fluvial lake and river sediments. The

catchment is sparsely inhabited, and apart from grazing by sheep, there is no major farming or industry. In 1973 the Vikedal River was protected by law from all future hydropower development. Additional details are found in Henriksen et al. (1983).

Investigations of water chemistry in Vikedal began in 1973 when the river was included as one of the rivers in DVF's (Directorate for Fish and Game, now Directorate for Protection of Nature) monthly sampling program. In 1980 the river was included as one of about 20 rivers in the newly established Norwegian Monitoring Programme for Long-Range Transported Air Pollutants under the auspices of the Norwegian State Pollution Control Authority (Johannessen 1995). This entailed monthly sampling for water chemistry at a station downstream Låkafooss (station 32.1) (NIVA), weekly samples for chemical analysis of precipitation (NILU), and studies of soil chemistry in 1982 and 1990 (NISK). In 1982 an intensive study of the water chemistry and fish status of the entire Vikedal water course was carried out (Henriksen et al. 1983).

Liming of the lower reaches of the river (from Låkafooss) began in 1987 as part of the Norwegian national Liming Programme ((Hindar and Lien 1999)). Powdered limestone is dosed continuously to the river at Låkafooss with the objective to maintain pH 6.3 during the period of salmon smoltification (15 February to 1 June) and pH 5.7 the rest of the year. The monitoring station for water chemistry was moved to above Låkafooss (station 32.9) in 1987.

3.1.2 Tovdal

The Tovdal River is a major river in southernmost Norway and runs north-to-south from headwaters in Telemark, through Aust-Agder to the coast at Krisitiansand (Figure 1). The catchment area is 1794 km² above the gauging station at Flaksvatn. The catchment has predominantly Precambrian granitic and gneissic bedrock, with thin and patchy moraine of the same lithology. The higher lying areas of the catchment are characterised by alpine, heathland and peaty soils. Much of the lower parts are forested with pine, spruce and birch. There is very little farming, industry or habitation in the catchment, and it has not been developed for hydropower significantly.

The Tovdal catchment contains nearly 300 lakes, the majority of which are highly acidified and lost their native fish populations (mainly brown trout) in the period 1940-1980. The river itself is acidified and the salmon population disappeared in the 1960's following decades of steady decline. Studies of water chemistry and fish populations in the Tovdal region began in earnest with the SNSF-project 1972-1980. The river was included in 1973 as one of the rivers in DVF's (Directorate for Fish and Game, now Directorate for Protection of Nature) monthly sampling program. In 1980 the river was included as one of about 20 rivers in the newly established Norwegian Monitoring Programme for Long-Range Transported Air Pollutants under the auspices of the Norwegian State Pollution Control Authority (Johannessen 1995). This entailed monthly sampling for water chemistry at Boen Bruk (station 7.1) (NIVA).

Beginning in late 1996 a major whole-river liming programme began, and was fully implemented during 1997. In 1997 10 lakes were limed with a total of 1879 tonne NK3 limestone (86% Ca CO₃), of which 1500 tonne went to the lake Ogge. In addition a total of 3742 tonnes of NK3 were added to the river at 6 dosing points. This is about 2/3 of the full amount of limestone from the dosers specified in the liming plan for Tovdal.

3.2 Soils data

3.2.1 Vikedal

Soils in the Vikedal catchment were sampled in 1985 by NISK (Norwegian Forest Research Institute) as part of SFT's Norwegian Monitoring Programme for Long-Range Transported Air Pollutants (SFT

1986). Four areas with homogeneous vegetation and overburden were selected within the entire catchment. At each area 50x4 soil cores were collected. Each core was split into 2-cm layers, and all 50 samples from each layer bulked to one overall sample representing a 2-cm layer. Samples were dried and sieved and sent to the laboratory at NISK for analysis by standard methods (Ogner et al. 1991). Soils at Vikedal were resampled in 1995 by the same procedures (SFT 1996). There were only minor differences in chemistry between 1985 and 1995 samplings. The 1985 data are used here.

For MAGIC it is necessary to aggregate the soils data into one set of numbers characteristic for the whole catchment. This has been done as part of previous application of MAGIC to Vikedal (Wright et al. 1990; Reuss 1990), and these data are used here in the MAGIC application (Table 1).

Table 1. Vikedal. Aggregated soils data used to calibrate the MAGIC model (from Wright et al. 1990; Reuss 1990).

Horizon means	Depth cm	Bulk density kg/m ³	CEC mmol/kg	Ca %	Mg %	Na %	K %	BS %
O	5	243	280	26.5	20.1	1.7	4.4	52.6
Ah	7	753	58	10.6	11.1	1.7	5.7	29.0
E	6	1025	24	4.8	5.0	1.7	2.6	14.1
B	39	1042	21	2.5	1.8	1.8	1.9	7.9
C	19	1326	8	3.4	1.0	2.5	1.9	8.8
Total	75	1037	23	3.8	2.6	2.0	2.2	10.7

3.2.2 Tovdal

Soils data are available for Storgama and Birkenes, calibrated catchments that lie within the Tovdal drainage basin and are monitored as part of SFT's Norwegian Monitoring Programme for Long-Range Transported Air Pollutants (SFT 1998). Storgama lies in high-elevation headwater area typical of much of the upland areas of the Tovdal catchment. Birkenes lies at lower elevation at the lower reach of the basin, and is characterised by podsollic soils and mature productive forest of pine, spruce and birch. Soils at Storgama were sampled in 1982 (SFT 1983) and again in 1990 (SFT 1991); Birkenes was sampled in 1984 (SFT 1985) and 1992 (SFT 1993). Sampling and analysis methods were the same as used for Vikedal, and the data were aggregated in a similar manner to obtain one set of data characteristic for each site (Reuss 1990) for use in MAGIC (Table 2) (Wright et al. 1990). In lieu of better estimate a simple arithmetic average of these 2 sets of data was used for Tovdal soils (Table 2).

Table 2. Storgama and Birkenes. Aggregated soils data used to calibrate the MAGIC model (from Wright et al. 1990; Reuss 1990).

Storgama								
Horizon means	Depth cm	Bulk density kg/m ³	CEC mmol/kg	Ca %	Mg %	Na %	K %	BS %
H1	15	235	259	7.5	4.6	1.0	1.0	14.1
H2	8	354	247	2.8	2.5	0.5	0.5	6.2
Ah,Bs	9	1083	35	2.0	1.2	0.6	0.6	4.5
Total	32	503	121	3.3	2.2	0.7	0.7	6.9

Birkenes								
Horizon means	Depth cm	Bulk density kg/m ³	CEC mmol/kg	Ca %	Mg %	Na %	K %	BS %
O	8	266	322	18.9	9.1	1.5	3.8	33.3
Ah	7	1063	36	4.1	2.0	1.8	2.7	10.6
E	8	963	53	2.3	1.2	1.3	1.4	6.2
B	12	1029	25	1.3	1.3	1.9	2.2	6.7
C	5	1560	7	2.2	0.7	3.0	1.6	7.5
Total	40	936	46	9.0	4.5	1.6	2.7	17.8
Tovdal (average Storgama and Birkenes)								
Total	36	717	84	6.2	3.4	1.2	1.7	12.3

3.3 Deposition data

3.3.1 Vikedal

Bulk deposition is collected in weekly samples at a station in the lower part of Vikedal catchment. Measurements began in 1984. Samples are sent to NILU (Norwegian Institute for Air Research) for chemical analysis. This activity is also part of the Norwegian monitoring programme, and results are reported annually (Tørseth and Manø 1997). Annual volume-weighted mean concentrations are given in Appendix B.

These deposition values, however, do not give representative deposition for the catchment as a whole. Dry deposition of both seasalts and acid pollutants are not included. Both precipitation volume and chemical concentrations vary with distance from the coast and elevation, due to among other things orographic effects. The measured annual bulk precipitation fluxes were thus adjusted in three ways to obtain estimates for catchment-representative deposition.

The measured runoff discharge and the measured arithmetic average chemical concentrations in the river were assumed to be exactly correct. First the annual flux of chloride (Cl) was calculated by multiplying the arithmetic average concentration by the annual discharge. Next the annual measured Cl fluxes in deposition were multiplied by a factor (1.20) such that the flux of Cl in deposition matched the flux of Cl in runoff for the entire period 1984-96. This estimate assumes that all Cl in runoff comes in atmospheric deposition, and that there are no sources or sinks for Cl in the catchment. Finally the inputs of the other seasalts (Na, K, Ca, Mg, SO₄) were increased by amounts equivalent to the ratios to Cl in seawater. This procedure adjusted for uncertainty in amount of deposition and seasalt inputs to the catchment.

Next the inputs of acid pollutants were adjusted such that the flux of sulphate in deposition matched the flux in runoff for the 3-year period 1984-86 (the first 3 years of deposition measurements). Annual fluxes in deposition of non-marine sulphate were multiplied by a factor (0.88) for all years. This procedure assumes that for the 3-year period 1984-86 that all SO₄ in runoff came in atmospheric deposition, that there are no sources or sinks for SO₄ in the catchment. In fact there are soil processes operating on time scales of years-to-decades which retain and release SO₄, but these are assumed to be negligible (or at steady-state) for the 3-year period 1984-86.

3.3.2 Tovdal

Bulk deposition is collected in daily samples at 2 stations within Tovdal catchment, Treungen (near Storgama) and Birkenes. Measurements began in 1974 and 1971, respectively. Samples are sent to NILU (Norwegian Institute for Air Research) for chemical analysis. This activity is also part of the Norwegian monitoring programme, and results are reported annually (Tørseth and Manø 1997). Annual volume-weighted mean concentrations are given in Appendix B.

For this application to Tovdal, the data from Treungen (Storgama) were used as starting point. Birkenes lies very near the coast, and thus has much higher concentrations of seasalts and also acid pollutants than Tovdal catchment as a whole. The Treungen deposition data were then treated in the same manner as the Vikedal data, to correct for precipitation volume, seasalt deposition and sulphate deposition. For chloride (and the seasalt contribution to Ca, Mg, Na, K, and SO₄) the measured bulk deposition was multiplied by the factor 3.80 (match average deposition and runoff for period 1980-96). For non-marine sulphate the measured deposition was multiplied by the factor 1.76 (match average deposition and runoff for 3-year period 1980-82).

3.3.3 Historical trends

Historical trends for deposition of sulphate were taken from (Mylona 1996) (Figure 2) using values for the Birkenes block in the EMEP grid system and scaled to measured (adjusted) deposition for the years 1994-96. For nitrate the trends given by (Simpson et al. 1997) were used. Historical trend for ammonium was assumed to be the same as that for sulphate. Deposition of other ions was assumed to be unchanged (Table 4).

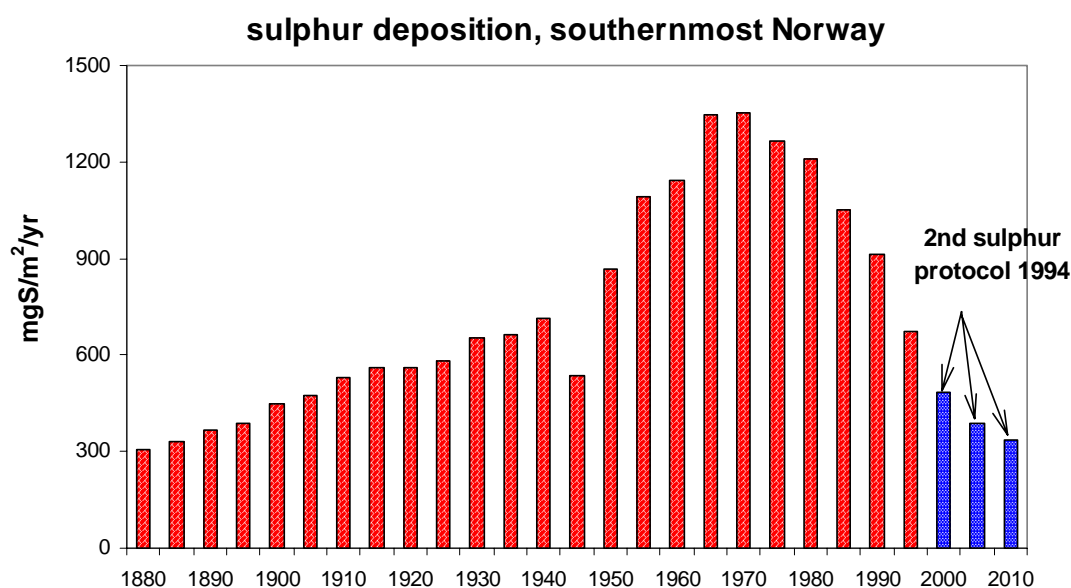


Figure 3. Historical deposition of sulphur in southernmost Norway (Birkenes block of the EMEP grid) and predicted deposition to the year 2010 assuming that the Second sulphur protocol of 1994 (Oslo convention) is fulfilled. From Mylona (1996).

A deposition file was prepared for MAGIC in which the adjusted deposition flux for the years prior to onset of measurements (1856-1983) the mean (adjusted) values for Ca, Mg, Na, K and Cl were used, and for SO₄, NO₃, and NH₄ the values were scaled according to the historical trends. The measured

(adjusted) yearly values were used for the years 1984-96 (Vikedal) and 1975-96 (Tovdal) (Appendix B).

Table 3. Vikedal and Tovdal. Mean deposition corrected for seasalts and non-marine sulphate for the period of measurements and the pre-industrial background (1856). Units: meq/m²/yr.

	Vikedal		Tovdal	
	Present 1984-96	Background year 1856	Present 1975-96	Background year 1856
Ca	21	21	8	8
Mg	69	69	16	16
Na	297	297	69	69
K	10	10	3	3
NH ₄	51	1	54	0
SO ₄	102 (1984-86)	34	88 (1980-82)	8
Cl	344	344	79	79
NO ₃	47	1	49	0

3.4 Runoff and water chemistry data

3.4.1 Vikedal

Discharge at Vikedal River is gauged continuously by NVE (Norwegian Electricity and Water Resources Board) at Holmen (Station number 38.1.0). Annual mean runoff for the years 1982-1997 is given in Appendix A. Mean runoff for the years 1984-96 was 2.946 m/yr. This value was used for the historical reconstruction for years 1856-1983.

Samples for chemical composition of river water are collected monthly at Låkafooss. Samples are sent to NIVA for chemical analysis, and the data are reported annually in the as part of the Norwegian monitoring programme (SFT 1998). The sampling point was below the falls (Låkafooss) (station 32.1) for the years 1980-85, and above the falls subsequently (above the point at which the river is limed). The annual arithmetic average concentrations are given in Appendix C. The average concentrations for the years 1984-86 used in calibration are given in Table 4.

3.4.2 Tovdal

Discharge at Tovdal River is gauged continuously by NVE (Norwegian Electricity and Water Resources Board) at Flaksvatn (Station number 20.3.0). Annual mean runoff for the years 1972-1997 (1996 missing) is given in Appendix A. Mean runoff for the years 1974-95 was 1.046 m/yr. This value was used for the historical reconstruction for years 1856-1974.

Samples for chemical composition of river water are collected monthly at Boen Bruk. Samples are sent to NIVA for chemical analysis, and the data are reported annually in the as part of the Norwegian monitoring programme (SFT 1998). The annual arithmetic average concentrations are given in Appendix C. The average concentrations for the years 1984-86 used in calibration are given in Table 4.

Table 4. Arithmetic average concentrations of major ions in river water at Vikedal for the calibration period 1984-96 and for Tovdal for the period 1980-82. Data from SFT (1998). SBC: sum of base cations. SSA: sum of strong acid anions. ANC: acid neutralising capacity. Al^{n+} : sum of positively-charged inorganic Al species. A^- : organic anions. Units: $\mu\text{eq/l}$.

Parameter	Vikedal 1984-86	Tovdal 1980-82
Ca	38	49
Mg	31	27
Na	98	64
K	6	8
NH_4	<1	<1
SO_4	51	91
Cl	112	70
NO_3	12	12
SBC	174	148
SSA	176	174
ANC	-3	-26
H^+	4	12
Al^{n+}	2	12
A^-	2	0

4. Calibration procedure

The calibration procedure first entailed compilation of data for soil, deposition and runoff that could be taken as characteristic for the entire catchment. Details of this step are described in the above sections. This resulted in the soil data given in Tables 1 and 2, the deposition data given in Table 3, and the runoff data given in Table 4.

MAGIC was then run for the 140-year hindcast period 1856-1996 and parameters adjusted such that the modelled concentrations in runoff and in soil matched those measured for the 3-year period 1984-86 (Vikedal) and 1980-82 (Tovdal). The parameters were adjusted in the following order.

1. The Cl concentrations were checked (adjustment came already in the estimation of the deposition).
2. The SO_4 adsorption capacity was adjusted such that the modelled SO_4 concentrations in runoff agreed with observed over the entire measurement period 1984-96 (Vikedal), 1980-1996 (Tovdal). For Vikedal this resulted in a relatively large value for maximum sulphate adsorption of 12 meq/kg, while for Tovdal the value was only 1 meq/kg (Table 5).
3. The values for % retention of nitrate and ammonium were adjusted such that the modelled NO_3 and NH_4 concentrations agreed with the observed. This resulted in values of 25% for NO_3 and 98% for NH_4 for Vikedal, and 75% and 98% for Tovdal (Table 5); these values were assumed to remain constant over time (i.e. no time trend in nitrogen saturation). At this point the sum of strong acid anions (SSA: $SO_4+Cl+NO_3$) should also be correct.
4. The base cations (Ca, Mg, Na, K) were optimised by a trial and error procedure in which the weathering rate and initial % cation on the soil were adjusted such that the modelled and observed concentration in runoff and % cation on the soil agreed with the measured for the 3-year period 1984-86 (Vikedal), 1980-82 (Tovdal). This resulted in cation weathering rates and selectivity coefficients that are held constant over time (Table 6). At this point the sum of base cations (SBC: $Ca+Mg+Na+K$) should be correct, and thus also acid neutralising capacity ($ANC = SBC - SSA$).

5. The next steps involved calibration of the weak acid components. These sum to give ANC ($\text{ANC} = \text{HCO}_3^- + \text{A}^- - \text{Al}^{3+} - \text{H}^+$). Concentrations of bicarbonate were assumed to be negligible. Concentration of organic anions was then fitted by adjusting the total organic acid concentration (triprotic) in streamwater (Table 5).
6. The partitioning of strong acid cations between H^+ and Al^{3+} was calibrated by adjusting the solubility constant (K_{Al}) for aluminium trihydroxide in the stream. A value of $\log K_{\text{Al}} = 10.1$ fit Vikedal (Table 5). This value is about that of amorphous aluminium hydroxide, indicating that inorganic Al originating from soil solution is probably precipitated out in the stream, presumably due to increase of pH following degassing of CO_2 as the water moves from the soil to stream environment. The value for Tovdal was somewhat lower 9.3.

Table 5. Fixed parameters (measured, estimated or calibrated) for Vikedal and Tovdal used in calibration of MAGIC.

	Vikedal	Tovdal	Units
<i>Runoff parameters</i>			
Discharge annual	2.946	1.046	m
Solubility $\text{Al}(\text{OH})_3$	10.1	9.3	log10
Temperature	6.6	5.0	$^{\circ}\text{C}$
CO_2 partial pressure	.0011	.0011	atm
Total organic acid	4	4	mmol m^{-3}
pK1	4.5	4.5	-log10
pK2	8.0	8.0	-log10
pK3	16.0	16.0	-log10
<i>Soil parameters</i>			
Soil depth	.75	.36	m
Porosity	50	50	%
Bulk density	1037	717	kg m^{-3}
Cation exchange capacity	23	84	meq kg^{-1}
Ca saturation	3.8	6.2	%
Mg saturation	2.6	3.4	%
Na saturation	2.0	1.2	%
K saturation	2.2	1.7	%
SO_4 ads. half-saturation	60	80	meq m^{-3}
SO_4 ads. max-capacity	12.0	1.0	meq kg^{-1}
Solubility $\text{Al}(\text{OH})_3$	8.8	8.1	log10
Temperature	6.6	5.0	$^{\circ}\text{C}$
CO_2 partial pressure	.006	.006	atm
Total organic acid	40	90	mmol m^{-3}
pK1	4.5	4.5	-log10
pK2	8.0	8.0	-log10
pK3	16.0	16.0	-log10

Table 6. Calibrated parameters for Vikedal and Tovdal obtained from MAGIC.

	Vikedal	Tovdal	Units
<i>Cation exchange selectivity coefficients</i>			
Al-Ca	-0.74	-0.08	log10
Al-Mg	-0.99	-0.08	log10
Al-Na	-1.60	-1.73	log10
Al-K	-5.30	-4.92	log10
<i>Weathering rates</i>			
Ca	57	28	meq m ⁻² yr ⁻¹
Mg	3	4	meq m ⁻² yr ⁻¹
Na	38	8	meq m ⁻² yr ⁻¹
K	0	0.5	meq m ⁻² yr ⁻¹
<i>Initial base saturation</i>			
Ca	12.9	20.1	%
Mg	9.9	11.3	%
Na	5.1	2.9	%
K	2.6	2.8	%
<i>Nitrogen retention</i>			
NH ₄	98	98	%
NO ₃	25	75	%

5. Results

5.1 Vikedal

Modelled concentrations of sulphate in river water at Vikedal well match the observed concentrations (Figure 4). Although deposition of sulphate decreased about 30% during the 1990's, concentrations of sulphate in the river decreased only about 10%. The pattern of sulphate concentrations in the river can be accounted for by a relatively large amount of sulphate adsorption in the catchment soils. Best fit was obtained with a value for maximum adsorption capacity of 12 meq/kg soil. With soil depth of 0.75 m and bulk density of 1037 kg/m³, this means that the total adsorption capacity of the soil is about 9000 meq/m². This amount is equivalent to 90 years of deposition at the average rate in the 1980's (100 meq/m²/yr).

Clearly the slow release of "old" sulphate acts to significantly delay recovery of river water at Vikedal in response to decreases in sulphur deposition. With no sulphate adsorption the SO₄ concentrations in river water would much more closely follow sulphate deposition. In 1995 and 1996 then, sulphate concentrations would have been only 20 µeq/l rather than 45 µeq/l observed (Figure 4).

Concentrations of Cl in Vikedal river water vary by a factor of 2 from year-to-year (Figure 4). These variations are due to differences in seasalt deposition, which in turn are related to frequency and intensity of storms. The modelled Cl concentrations show generally the same pattern, although the observed levels do not fluctuate as widely as the modelled. This is particularly true for the year 1993, which was characterised by very high Cl deposition due to several extreme storms in the winter (Hindar et al. 1993; SFT 1994). There is apparently a minor amount of retention and release of Cl in the catchment, most probably due to adsorption of Cl. The degree of Cl absorption is much less than that of SO₄, however, because the affinity of Cl to soil is generally much lower than that of SO₄.

A good match between modelled and observed for SO_4 and Cl means that the sum of anions also matches, because nitrate concentrations are only 9-14 $\mu\text{eq/l}$ and comprise < 10% of the sum of anions, and bicarbonate and organic anions are present only at negligible concentrations.

Modelled concentrations of sum of base cations (SBC) agree well with observed. In particular the marked decline in SBC in the last several years of the record are also simulated by MAGIC (Figure 4). Year-to-year variations are generally followed, although the model indicates larger variations than observed, probably because of the modelled larger variations in Cl.

The picture for ANC (acid neutralising capacity) is not as clear (Figure 4), although here the difference in scale distorts the picture somewhat. The modelled ANC agrees with observed to within about 10 $\mu\text{eq/l}$ for most years, with 1993 being a notable exception. In 1993 the model predicts a significantly lower ANC due to the seasalt episode, whereas the observed value indicates no decline in ANC in 1993. Here the explanation may lie in the fact that while the MAGIC values are volume-weighted means, the observed are simply arithmetic means of the 12 monthly samples.

This calibrated model can now be used to predict future trends in water chemistry at Vikedal given specified future deposition. The lag in response can thus be evaluated. As a first scenario sulphate deposition was held constant at 1997 levels and the model run for 50 years into the future (Figure 5). Sulphate concentrations in river water are predicted to decrease steadily, but take more than 20 years to reach new steady-state level, at which output of sulphate equals input from the atmosphere. Base cation concentrations, on the other hand, very quickly reach a new steady-state. ANC is predicted to increase relatively rapidly during the next 10 years and reaches a new steady-state level by about the year 2010.

If future sulphate deposition at Vikedal follows the trend predicted by the Oslo protocol, then sulphate deposition will continue to decrease to the year 2010. Sulphate concentrations in river water are then predicted to decrease gradually but not approach the new steady-state concentration until the year 2050, nearly 40 years later (Figure 6). Again little of this decline in SO_4 is accompanied by change in SBC, and most of the change is predicted to come as increase in ANC. ANC will increase from about -5 $\mu\text{eq/l}$ in 1997 to + 5 $\mu\text{eq/l}$ in 2010 and then gradually increase to about 12 $\mu\text{eq/l}$ by the year 2050 (Figure 6). Again this slow change in ANC is directly due to the slow response of SO_4 .

For Vikedal it is clear that most of the lag is due to slow response of sulphate to decreased sulphate deposition since 1980. Both SBC and ANC respond faster than SO_4 . The rapid response of SBC reflects the rather low F-factor for Vikedal, and most of the change in sulphate concentrations is accompanied by change in ANC.

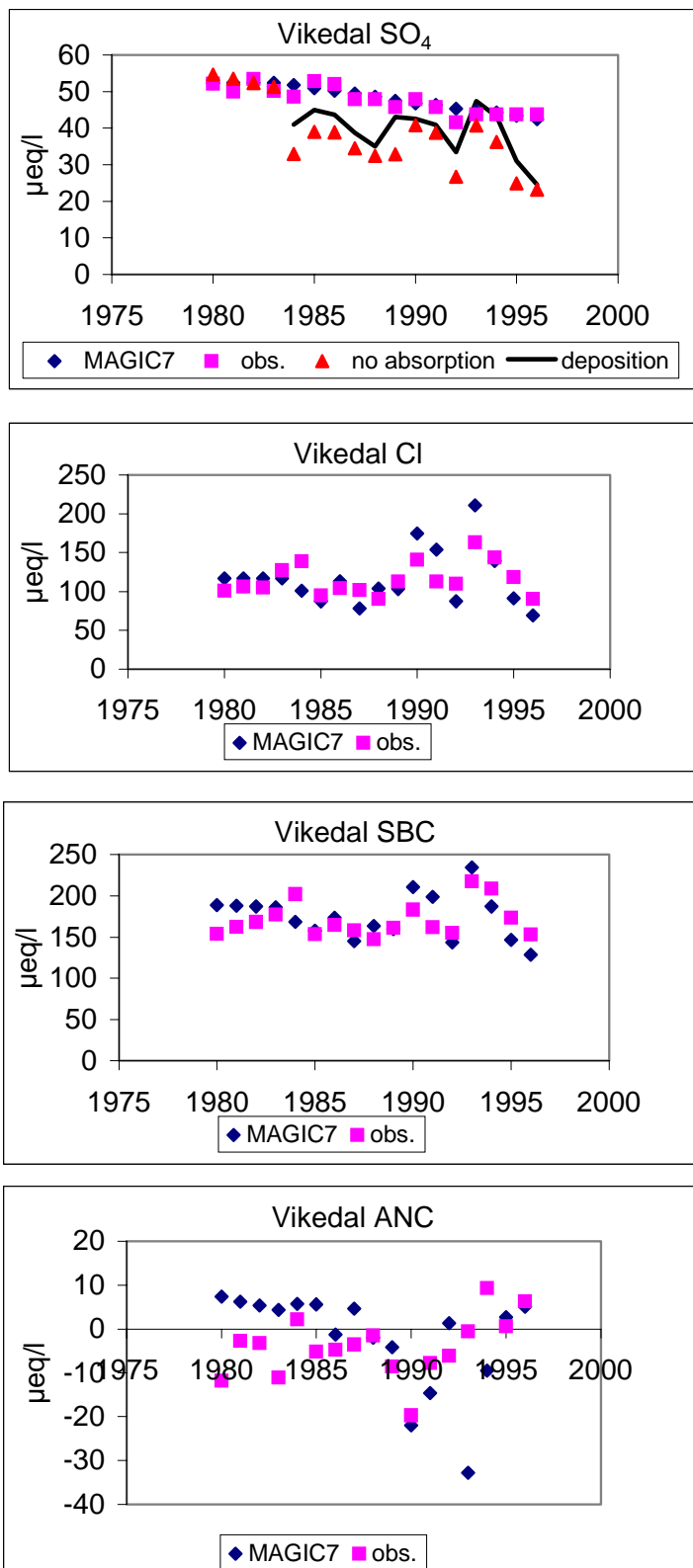


Figure 4. Vikedal. Comparison of calibrated and observed annual concentrations of SO₄, Cl, SBC (sum base cations) and ANC (acid neutralising capacity) in river water. For SO₄ (top panel) are also shown concentrations in precipitation (scaled to discharge in river) and expected concentrations in river if sulphate adsorption is zero in the catchment.

If SO₄ deposition is constant at 1997 level

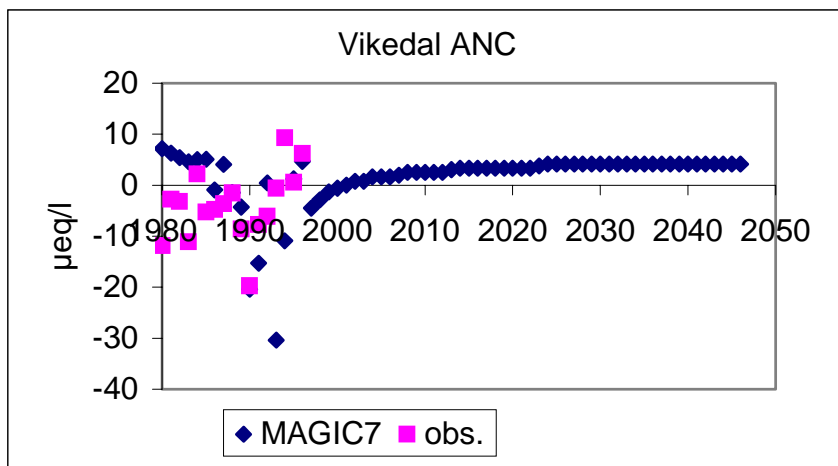
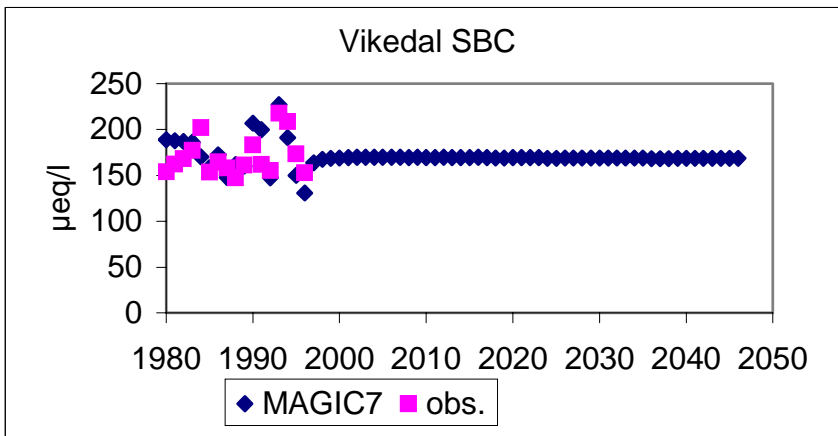
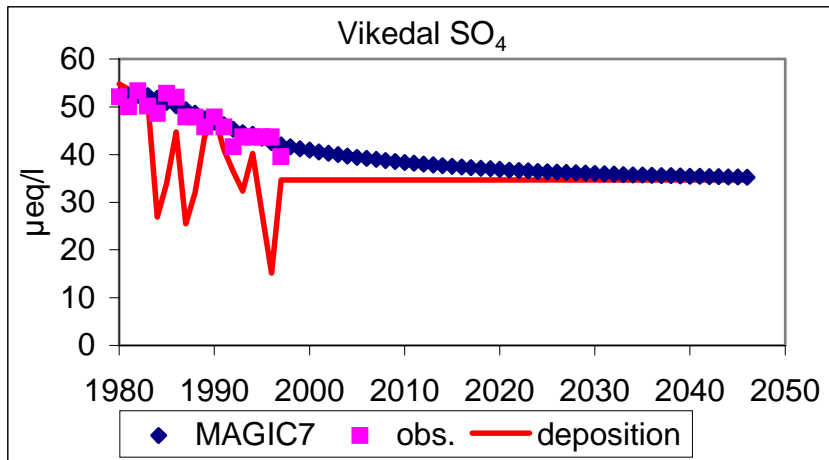


Figure 5. Vikedal. Predicted concentrations of SO₄, SBC (sum base cations) and ANC (acid neutralising capacity) with sulphate deposition held constant at 1997 level. Also shown are modelled and observed yearly values for the period of measurements 1980-1996.

If SO₄ deposition follows the Oslo protocol

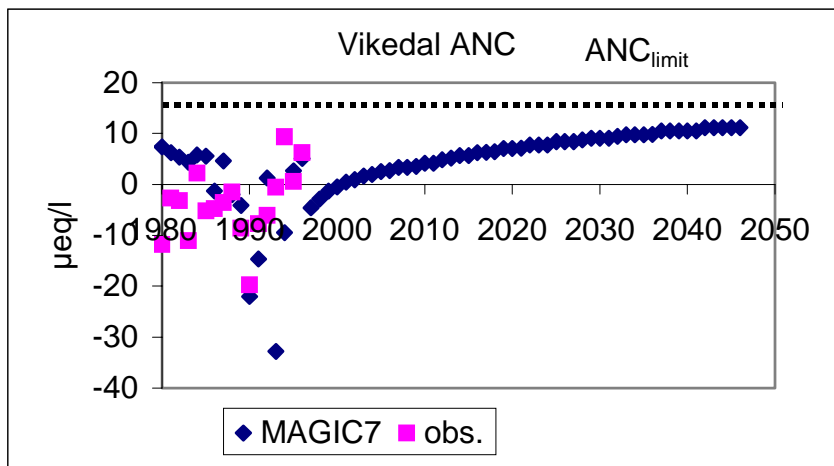
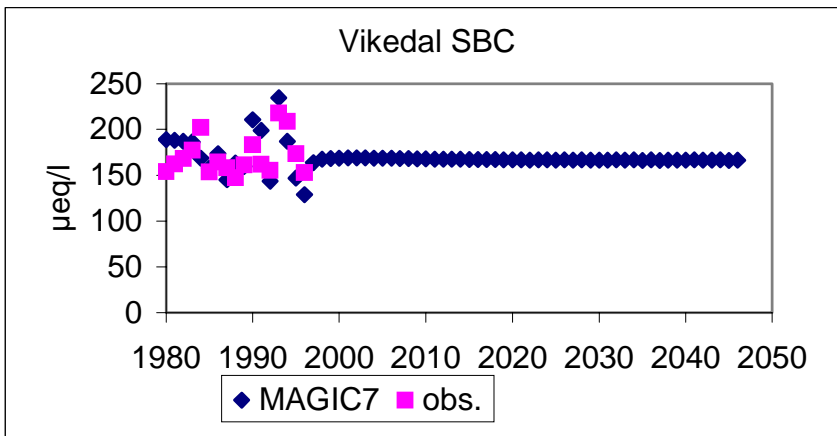
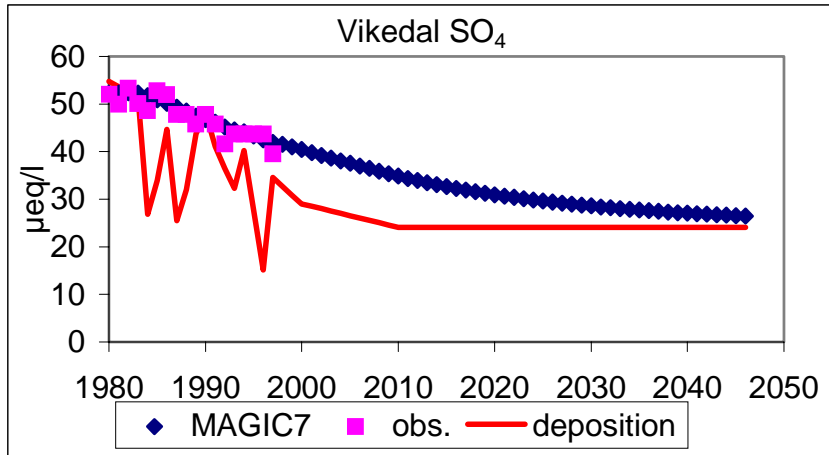


Figure 6. Vikedal. Predicted concentrations of SO₄, SBC (sum base cations) and ANC (acid neutralising capacity) with if sulphate deposition follows the Oslo protocol (Mylona 1996). Also shown are modelled and observed yearly values for the period of measurements 1980-1996.

5.2 Tovdal

For Tovdal the calibrated model indicates very little retention or release of sulphate in the catchment (Figure 7). Sulphate adsorption capacity was set to a very low value of 1 meq/kg, which for soil depth of 0.36 m and bulk density of 717 kg/m³, comes to a maximum capacity of only 260 meq/m². This is equivalent to only 5 years of sulphate deposition at the rate measured in the 1980's. Thus concentrations of sulphate in Tovdal river water closely track the deposition of sulphate as measured at Treungen and scaled to the calibration years 1980-82. There is essentially no lag time.

Variations in Cl concentrations are reasonably well-accounted for by MAGIC (Figure 7). Measured Cl levels do not show quite as much variation as the MAGIC simulated levels, however. In particular the seasalt year 1993 shows less response than predicted by MAGIC. This was also the case at Vikedal, and indicates that a small amount of retention/release of Cl does take place in the soils. MAGIC assumes that Cl is a completely mobile anion with no retention or release in the catchment.

At Tovdal the decreasing sulphate concentrations during the period 1985-1995 are accompanied by increase in base cations (Figure 7). Here the curves match quite well.

The measured trend in ANC in Tovdal has been steady increasing beginning in the mid-1980's, with the highest levels measured in 1994 and 1995, the last 2 years prior to onset of liming (Figure 7). MAGIC also indicates an increase in ANC, but not to such a large degree as the measured. For the last 2 years this discrepancy is apparently due to predicted SBC levels lower than measured.

For Tovdal there is little change in SO₄ concentrations predicted by MAGIC under the scenario of SO₄ deposition held constant at 1997 levels (Figure 8). The lag time is very short. Sum of base cations also is predicted to change very little. ANC, on the other hand, is predicted to increase somewhat to the year 2010, and then remain at about 0 µeq/l into the future (Figure 8). The lag time is about 10 years.

For the scenario of continued decrease in SO₄ deposition to the year 2010 (Oslo protocol), SO₄ concentrations in the river are predicted to decrease to less than ½ of present-day levels (Figure 9). SO₄ concentrations in the river closely track deposition. Base cations decrease slightly at first. This is due in part to the fact that seasalt deposition for future years in the simulation were set to the mean for the entire period of measurement (1980-95) whereas levels of seasalts in deposition (and the river) were about 10% higher than the average during the last few years of the measurements (1993-95). Base cations are then predicted to increase somewhat over the next 20 years due to replenishment of base cations in the soil cation exchange complex. ANC increases and approaches a new steady-state level of about 10 µeq/l already by the year 2020, only 10 years following the full reduction in SO₄ deposition called for by the Oslo protocol.

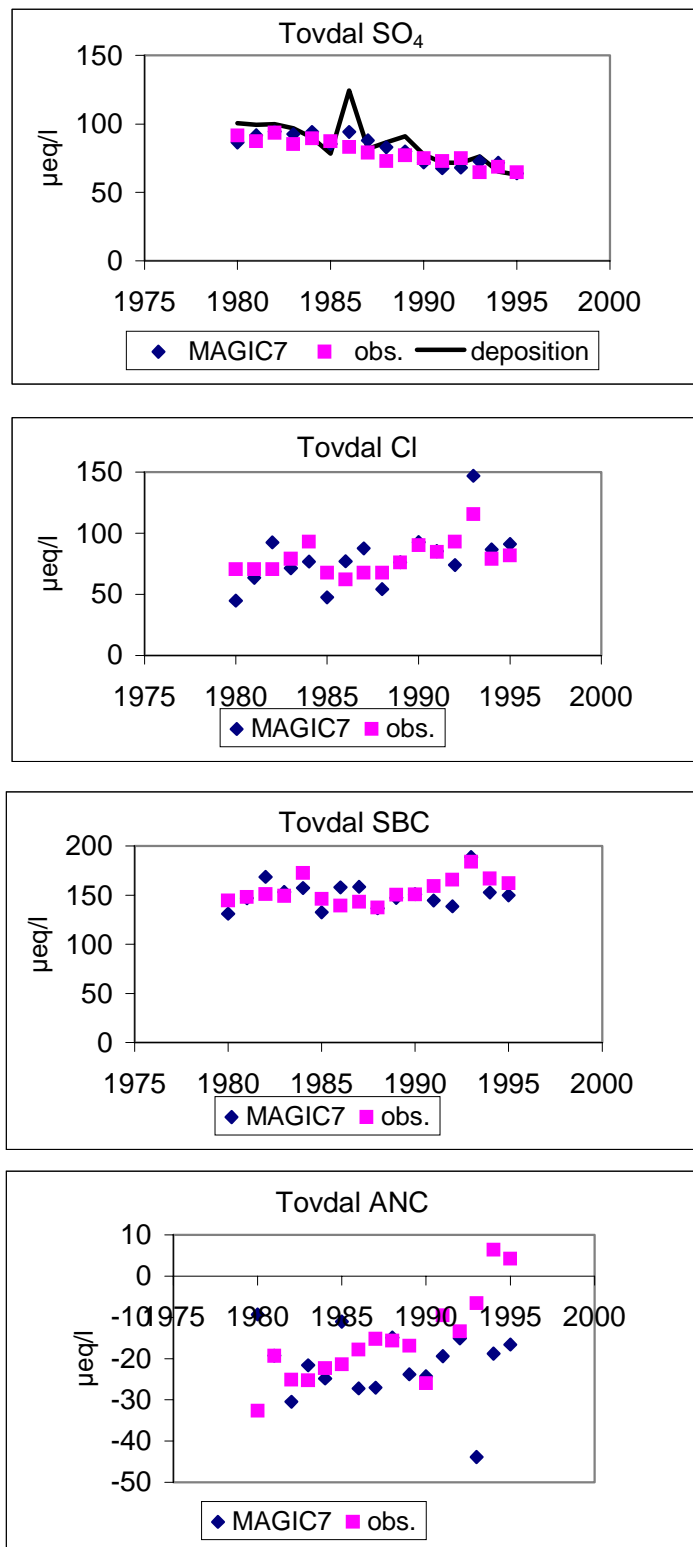


Figure 7. Tovdal. Comparison of calibrated and observed annual concentrations of SO_4 , Cl, SBC (sum base cations) and ANC (acid neutralising capacity) in river water. For SO_4 (top panel) are also shown concentrations in precipitation (scaled to discharge in river).

If SO₄ deposition is constant at 1997 level

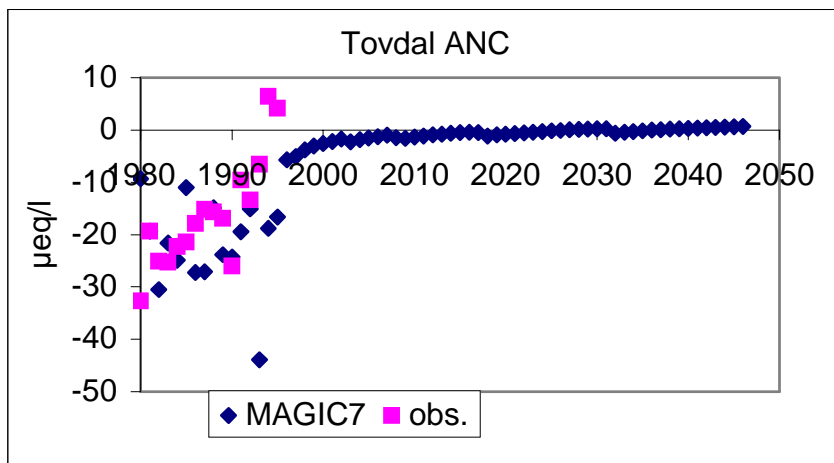
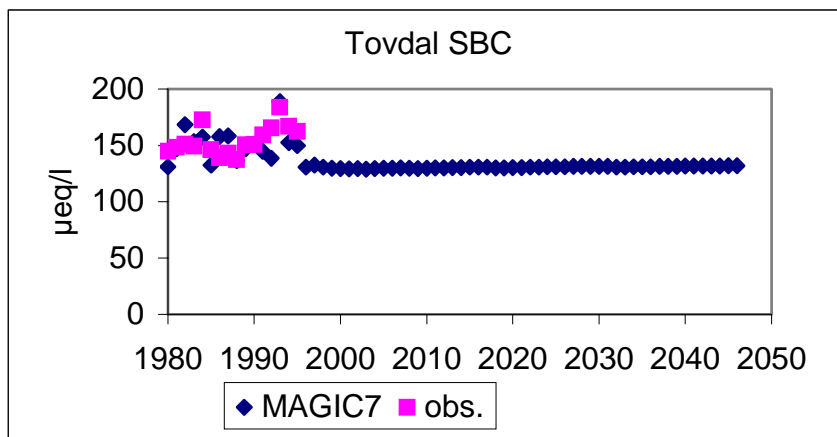
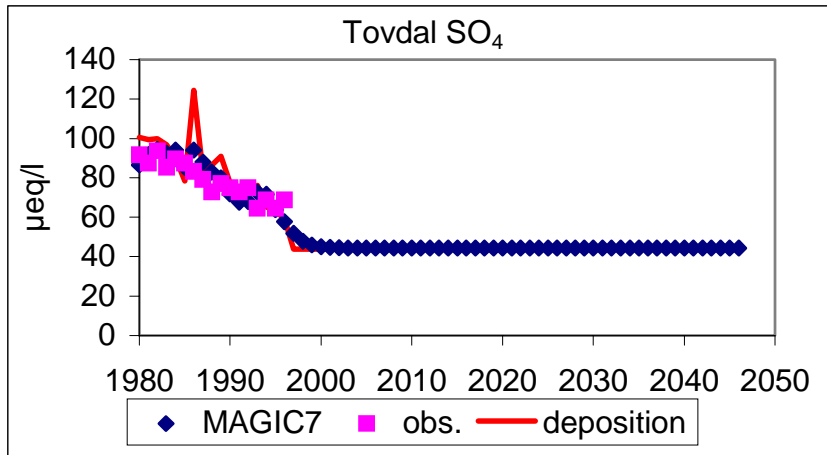


Figure 8. Tovdal. Predicted concentrations of SO₄, SBC (sum base cations) and ANC (acid neutralising capacity) with sulphate deposition held constant at 1997 level. Also shown are modelled and observed yearly values for the period of measurements 1980-1995.

If SO₄ deposition follows the Oslo protocol

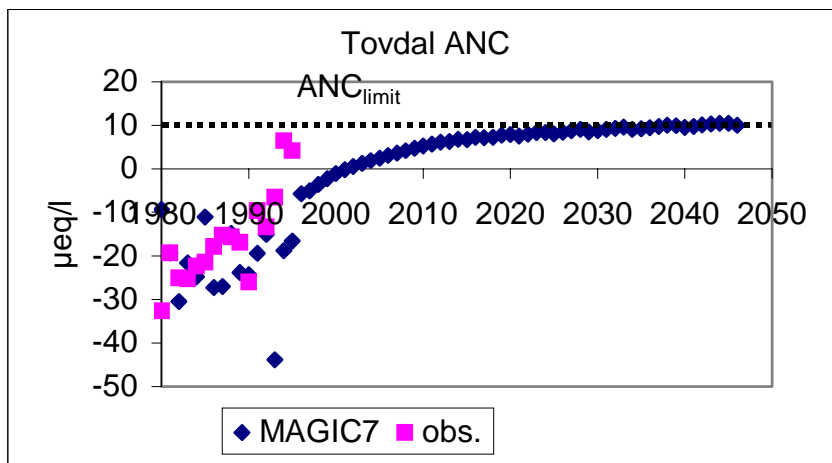
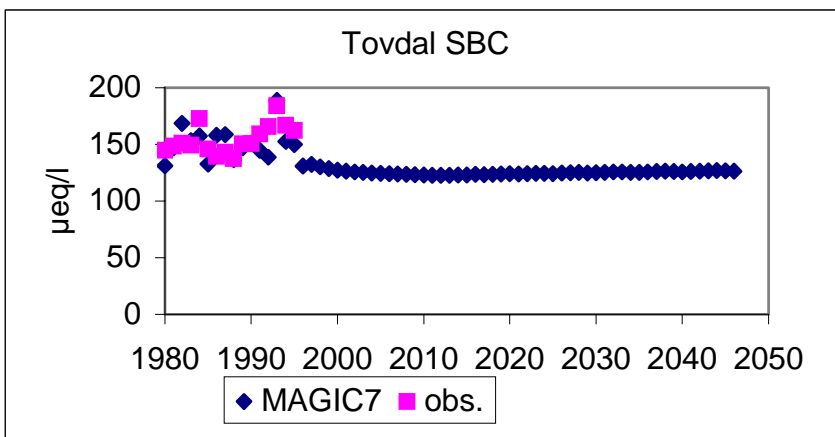
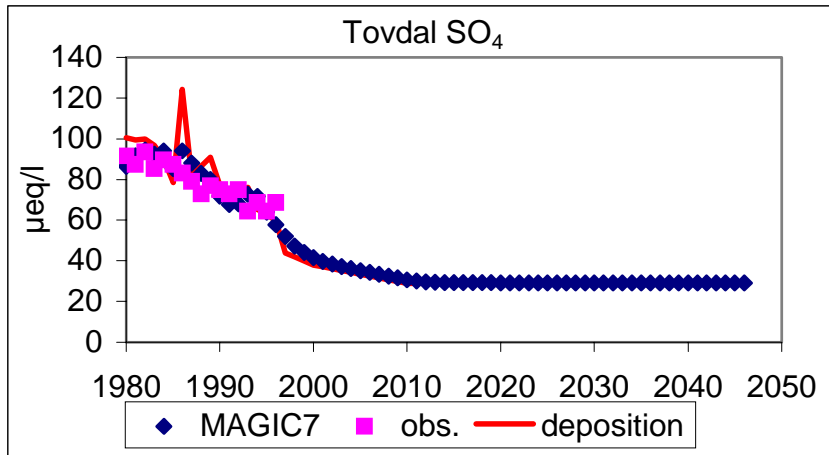


Figure 9. Tovdal. Predicted concentrations of SO₄, SBC (sum base cations) and ANC (acid neutralising capacity) with if sulphate deposition follows the Oslo protocol (Mylona 1996). Also shown are modelled and observed yearly values for the period of measurements 1980-1995.

6. Discussion

6.1 Lag times

The lag in response of sulphate concentrations in river water was quite different between the 2 rivers. Vikedal had a slow response with lag time on the order of 10-20 years, whereas Tovdal has a very rapid response and lag time of < 3 years. These 2 cases probably span nearly the entire range of response to be expected in Norwegian surface waters. Most of the acidified waters in southern Norway will probably have quick response to changes in sulphate deposition. Vikedal is probably the exception and this is likely due to soil characteristics at Vikedal. Perhaps the presence of soils developed from moraine and other overburden of Cambro-Silurian metamorphic rocks may be part of the explanation. In any case it is clear that evaluation of the lag time for sulphate response must involve consideration of soil type in the catchment.

The lag in base cations is also different between the 2 sites, although the difference is relatively smaller than that for SO₄. Tovdal shows a small long-term increase of base cation concentrations following the full implementation of the Oslo protocol, whereas at Vikedal very little change in the future is predicted (Table 7).

Table 7. Concentrations of sulphate, nitrate, base cations and ANC in river water at Vikedal and Tovdal predicted by MAGIC given the scenario of full implementation of the Oslo protocol by the year 2010. Nitrate concentrations were assumed to remain unchanged over the entire period. Units: µeq/l.

	Vikedal				Tovdal			
	1997	2010	2046	Δ 2046-1997	1997	2010	2046	Δ 2046-1997
SO ₄	42	35	26	-16	52	31	29	-23
SBC	164	168	167	+3	132	123	126	-6
ANC	-5	+4	+11	+16	-5	+5	+10	+15
NO ₃	12	12	12	0	12	12	12	0

Thus for the 2 sites the lag time for ANC response is about the same, 10 years or so. At Vikedal this is primarily due to slow response of SO₄, while at Tovdal it is due to long-term response of base cations. Again these are probably the 2 extremes in response expected in most Norwegian freshwater localities.

6.2 Strengths and weaknesses of the data

The availability of long data series from these 2 sites greatly helps the modelling calibration. The response of the water chemistry to the changes in deposition over the past 15-20 years gives a very good indication of how the sites will respond to further changes in the future. Confidence in the modelling results will be largest for sites at which such long-term data exist. One of the main purposes of the Norwegian national monitoring programme is to provide documentation of changes in environmental quality due to long-range transported air pollutants. Modelling applications such as this clearly show that the data obtained by the monitoring programme fill this purpose.

For application of dynamic acidification models such as MAGIC, the weakest part of the calibration is usually the sparse soil data. This is also the case for both Vikedal and Tovdal. For both sites soils data from only a few points in the catchment must be used to estimate soil properties characteristic of the entire catchment. Since soil is notoriously heterogeneous, the paucity of soils data will be the greatest source of uncertainty in the input data.

6.3 Comparison of static and dynamic models: critical loads

Critical loads for acidity for surface waters in Norway are calculated using one of several static models (UN/ECE 1996). The steady-state water chemistry model (SSWC) is based on a large amount of empirical data relating deposition and water chemistry and fish status in Norwegian lakes and rivers (Henriksen et al. 1992; Henriksen et al. 1995). Recently this model has been replaced by the first-order acidity balance model (FAB) (Posch et al. 1997; Henriksen 1998). The major difference between these two models is the treatment of nitrogen. In the SSWC model nitrogen retention is assumed to remain at present-day level, while the FAB model assumes maximum potential nitrogen leaching.

The critical load for acidity (sulphur + nitrogen) is exceeded for both Vikedal and Tovdal rivers at present (1993-95) (Table 8). The FAB model calculates larger exceedance than the SSWC model, because FAB assumes larger NO_3 leakage and thus greater flux of acidity. ANC falls below the critical limit ($\text{ANC}_{\text{limit}}$). This of course is the reason that both rivers are limed.

For the year 2010 under the assumption that the Oslo protocol is fully implemented, exceedance using the SSWC model at both rivers will decline to about zero (Table 8). This means that the river water chemistry will have ANC levels about equal to the $\text{ANC}_{\text{limit}}$. Of course the static model does not specify how many years it will take to reach this ANC level.

The MAGIC predictions provide this information. For Vikedal ANC is predicted to increase from present-day level of about $-5 \mu\text{eq/l}$ to $+4 \mu\text{eq/l}$ in 2010 and then further by about $2 \mu\text{eq/l}$ per decade to $+11 \mu\text{eq/l}$ by 2046 (Table 7, Figure 6). At this time changes are small and a new steady-state is reached. ANC is slightly below the $\text{ANC}_{\text{limit}}$, but then the critical load is also slightly exceeded (SSWC model; Table 8). Of the total improvement of ANC, about 60% is reached during the first 13 years by the year 2010 and it takes another 36 years to reach the remaining 40%. The first period is also a time of declining sulphur deposition, however, so the response should be more rapid in any case.

For Tovdal a similar picture emerges. Here MAGIC predicts that ANC will increase from $-5 \mu\text{eq/l}$ at present to $+5 \mu\text{eq/l}$ by 2010 and to $+10 \mu\text{eq/l}$ by the year 2046 (Table 7, Figure 9). Again about 60% of the increase comes during the first 13 years.

All of these calculations assume that nitrogen deposition and leaching does not change from the present-day situation. Nitrate concentration in both rivers is about $12 \mu\text{eq/l}$, and shows no long-term change over the period 1980-95 (Table 7, Appendix C). The FAB model assumes that nitrate leaching will increase substantially in the future. Thus according to the FAB model both rivers will still be greatly exceeded in the year 2010, despite full implementation of the Oslo protocol (Table 8). More research is needed on nitrogen retention and release in areas subject to high nitrogen deposition, especially regions without forest.

Table 8. Critical loads and exceedance for Vikedal and Tovdal rivers calculated for present-day and the year 2010 assuming full implementation of the Oslo protocol using static models. SSWC: steady-state water chemistry model; FAB: first-order acidity balance model. Units: meq/m²/yr; Runoff: m; ANC_{limit}: µeq/l.

	Vikedal River	Tovdal River
Critical load	65	39
runoff	2.94	0.93
ANC _{limit}	16	10
<hr/>		
1993-95		
S* deposition	66	51
N deposition	96	90
SSWC Exceedance: S+N	+37	+23
SSWC exceedance: S only	0	+12
FAB exceedance	+96	+101
<hr/>		
2010		
S* deposition	36	22
N deposition	96	90
SSWC Exceedance: S+N	+7	-6
SSWC exceedance: S only	-36	-17
FAB exceedance	+60	+72

6.4 Liming needs in the future

Both Vikedal and Tovdal rivers are objects of major liming programmes, designed to provide water quality sufficient for reestablishment of salmon populations. At Vikedal whole-river liming began in 1987 ((Hindar and Lien 1999)) and has continued uninterrupted since. During the past few years about 500-600 tonnes limestone have been added annually (Table 9). The target at Vikedal for the years 1995-97 was pH 6.3 during the period of salmon smoltification (15 February – 1 June) and pH 5.7 the rest of the year. In 1994 the target was pH 6.5 in the smoltification period.

Large-scale liming in Tovdal began in late 1996. In 1997 10 lakes were limed with a total of 1879 tonnes powdered limestone (86% CaCO₃), and a further 3742 tonnes added at 6 dosing points to the river itself (Table 9). The amount added through the dosers was only 68% of the calculated demand. The pH target for 1997 was 5.8 – 6.0. There is no smolt in the river as yet.

Table 9. Annual addition of powdered limestone (about 85% Ca CO₃) to the Vikedal and Tovdal rivers. At Vikedal all the limestone is added by limestone dosing apparatus directly to the river at Låkafoss. At Tovdal limestone is added both to lakes and directly to the river at 6 dosing points. Units: tonnes/yr.

Year	Vikedal	Tovdal
1994	1007	
1995	599	
1996	512	
1997	661	5621

The increase in ANC due to reductions in sulphur deposition will mean a lower demand for liming in the future. For Vikedal the target for liming in the years 1995-97 of pH 6.3 and 5.7 translates to target ANC of about 40-50 and 10-15 $\mu\text{eq/l}$, respectively. A rough average for the year is about 30-35 $\mu\text{eq/l}$. The dose in 1995-97 of 500-660 tonnes/yr was sufficient to raise the ANC from about +3 $\mu\text{eq/l}$ to this target of 30-35 $\mu\text{eq/l}$. In the year 2010 MAGIC predicts that the river will have ANC +4, and thus the requirement for limestone will be about the same as at present. By the year 2046 the predicted ANC will increase to +11 $\mu\text{eq/l}$, and the requirement for limestone will be reduced to about 2/3 of the present, or 300-400 tonnes/yr.

For Tovdal the calculation is more uncertain, because full liming programme has not yet been implemented. Under the assumption that the total requirement for limestone from the dosers is 5480 tonne/yr, and that this quantity is sufficient to raise the pH from the level in 1993-95 to pH 6.3 during smoltification and 5.7 otherwise, then this full dose is required to raise ANC from +1 $\mu\text{eq/l}$ (1993-95) to about +30-35 $\mu\text{eq/l}$. In the year 2010 MAGIC predicts that ANC will have increased to +5 $\mu\text{eq/l}$, and the required limestone dose will be reduced by about 15% to about 4650 tonne/yr, and by the year 2048 the ANC will have increased to +10 $\mu\text{eq/l}$, and the required dose will be about 3800 tonne/yr, about 70% of present-day requirement (Table 10).

Table 10. Calculation of future requirement for limestone (tonne/yr) at Vikedal and Tovdal rivers. See text for explanation of assumptions.

	Vikedal	Tovdal
ANC target	+33 $\mu\text{eq/l}$	+33 $\mu\text{eq/l}$
<i>1995-97</i>		
ANC obs. 1995-97	+3	+1
Δ ANC required	+30	+32
Limestone dose 1995-97	510-660	5480
<i>2010</i>		
ANC pred. 2010	+4	+5
Δ ANC required	+29	+28
Limestone dose 2010	500-650	4650
Dose % of 1997	95%	85%
<i>2046</i>		
ANC pred. 2046	+11	+10
Δ ANC required	+22	+23
Limestone dose 2046	350-460	3800
Dose % of 1997	70%	70%

Target pH may be set somewhat lower in the future, however, and this will mean lower requirement for limestone. The pH targets entail levels that are probably higher than those naturally found in the river prior to acidification. As the natural water quality of the rivers recovers following full implementation of the Oslo protocol, it will probably not be necessary to raise the pH to such high levels. Today these targets are set such that acid episodes are dealt with, and so that a sufficient fraction of the toxic inorganic aluminium species are removed from the water. As recovery proceeds in the future, the amount of Al leached from the soils will decrease as will the intensity, frequency and duration of acid episodes. Thus it may be possible to have lower pH target in the future.

7. References

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Appendix A. Annual discharge Vikedal and Tovdal

Annual discharge for Tovdal River (Flaksvatn, Station number 20.3.0, catchment area 1794 km²) and Vikedal River (Holmen, Station number 38.1.0, catchment area 115 km²). Data provided by NVE Norwegian Water Resources and Electricity Board.

	Tovdal		Vikedal	
	m ³ /s	m/yr	m ³ /s	m/yr
1972	55.54	0.976		
1973	32.72	0.575		
1974	60.90	1.071		
1975	48.26	0.848		
1976	46.71	0.821		
1977	59.12	1.039		
1978	58.24	1.024		
1979	68.80	1.209		
1980	51.62	0.907		
1981	53.36	0.938		
1982	68.54	1.205		
1983	65.94	1.159	15.76	4.322
1984	63.19	1.111	8.79	2.410
1985	55.29	0.972	9.34	2.561
1986	66.45	1.168	12.33	3.381
1987	70.58	1.241	7.98	2.188
1988	78.09	1.373	10.56	2.896
1989	52.74	0.927	14.29	3.919
1990	78.72	1.384	12.71	3.485
1991	51.01	0.897	11.33	3.107
1992	53.32	0.937	14.31	3.924
1993	42.25	0.743	8.40	2.303
1994	57.29	1.007	11.86	3.252
1995	59.02	1.037	11.76	3.225
1996	a	0.960	7.05	1.933
1997	49.33	0.867	10.92	2.995
	mean 72-95	1.024		
	mean 74-95	1.046	mean 84-96	2.968
	mean 80-82	1.017	mean 84-86	2.784
	mean 93-95	0.929		

a. Data not available from Tovdal. Estimated from Birkenes discharge.

Appendix B. Deposition data for Vikedal and Tovdal

Total deposition (wet plus dry) used for Vikedal River. Data are derived from Vikedal weekly bulk deposition data (NILU as reported in SFT's annual reports) and scaled to match Cl and SO₄ in river water 1984-86. Units: meq/m²/yr.

	Ca	Mg	Na	K	NH ₄ -N	SO ₄ -S	Cl	NO ₃ -N
1984	23.9	47.7	173.6	42.7	37.3	79.2	243.4	33.1
1985	24.0	43.9	159.8	40.0	52.4	100.0	224.0	47.6
1986	23.7	76.9	279.5	64.7	64.7	131.7	377.8	53.9
1987	13.1	34.3	124.2	30.3	47.2	75.2	170.3	37.5
1988	18.4	63.0	218.0	48.7	48.1	94.3	298.1	50.0
1989	22.2	77.4	272.6	64.7	49.3	128.9	404.8	68.5
1990	26.8	115.7	425.8	94.3	74.0	142.0	594.7	52.5
1991	22.1	96.1	345.3	78.4	57.1	121.0	480.1	55.0
1992	20.3	69.9	253.1	59.3	55.1	107.5	357.8	50.5
1993	23.5	95.2	342.6	78.5	38.7	95.2	485.4	34.4
1994	21.9	96.5	354.5	75.4	58.8	118.6	464.3	54.9
1995	18.0	61.9	221.2	49.1	43.3	81.7	301.4	43.3
1996	14.9	28.3	93.4	22.1	36.4	44.6	132.3	29.9

Total deposition (wet plus dry) used for Tovdal River. Data are derived from Treungen bulk deposition data (NILU as reported in SFT's annual reports) and scaled to match Cl and SO₄ in river water 1980-82. Units: meq/m²/yr.

	Ca	Mg	Na	K	NH ₄ -N	SO ₄ -S	Cl	NO ₃ -N
1975	9.1	16.8	73.3	3.1	38.2	95.8	85.6	41.6
1976	5.8	13.2	57.9	2.9	37.3	86.5	67.6	44.4
1977	9.0	18.2	79.6	3.2	57.1	110.7	92.9	64.4
1978	8.3	11.8	51.6	2.8	48.7	94.9	60.3	45.1
1979	8.3	14.6	60.1	2.9	54.4	94.5	71.4	48.6
1980	6.4	8.0	32.4	1.9	37.2	76.3	38.2	35.3
1981	7.4	12.9	53.2	3.0	54.9	94.3	62.0	46.5
1982	11.1	22.9	94.9	3.8	71.0	112.9	113.8	63.9
1983	12.1	16.1	70.5	3.1	59.0	105.6	80.7	54.9
1984	11.4	17.3	75.7	3.1	40.6	107.7	85.9	54.1
1985	7.0	9.1	40.0	2.6	41.5	69.9	43.0	43.7
1986	9.8	19.3	79.5	4.4	81.6	128.1	92.7	73.8
1987	10.4	22.3	93.1	4.3	52.7	92.9	109.3	52.7
1988	8.7	16.0	62.0	2.7	76.3	116.5	72.2	84.7
1989	5.8	14.3	61.1	1.9	41.7	68.5	73.5	57.8
1990	7.2	25.5	107.9	3.2	55.1	91.7	129.4	62.5
1991	7.4	14.9	63.2	2.5	34.7	58.3	75.6	42.8
1992	5.6	13.7	59.5	2.1	39.5	66.0	68.2	46.4
1993	7.8	23.2	101.3	3.0	32.3	61.1	119.6	41.4
1994	6.3	15.7	66.9	2.1	44.7	66.3	79.5	56.2
1995	6.7	19.8	82.4	2.9	45.4	56.9	95.8	49.9
1996	5.9	12.5	53.1	1.7	39.0	49.9	62.9	42.1
Mean 93-95	6.9	19.6	83.5	2.7	40.8	61.4	98.3	49.2

Appendix C. Water chemistry data for Vikedal and Tovdal

Vikedal. Arithmetic mean concentrations of major ions in river water collected monthly (NIVA) and reported in SFT's annual reports. Station 32.1 1980-85. Station 32.9 1986-1997. Units: µeq/l, TOC mmol/l. Org⁻: organic anions; SBC: sum of base cations; SSA: sum of acid anions; ANC: acid neutralising capacity; Ali: inorganic aluminium.

Vikedal	Ca µeq/l	Mg	Na	K	SO4	Cl	NO3	org-	SBC	SSA	ANC	H+	Ali	TOC mmol/l
1980	36.9	27.4	84.1	5.5	52.1	101.1	12.5	-8.1	153.9	165.7	-11.8	3.7		
1981	36.1	28.8	92.5	5	50	106.3	8.8	0.7	162.4	165.1	-2.7	3.4		
1982	40.3	31.3	91	5.7	53.4	104.9	13.2	0.1	168.3	171.5	-3.2	3.3		
1983	35.8	32	104.5	4.9	50.1	127.3	10.9	-6.6	177.2	188.3	-11.1	4.5		
1984	42.2	34.4	119.6	5.9	48.6	138.6	12.7	5.5	202.1	199.9	2.2	3.3	1.9	75
1985	36.8	28.1	83.8	5.1	52.8	94.7	11.5	-1.8	153.8	159.0	-5.2	3.4	1.5	108
1986	35.9	31.4	90.5	6.7	52.0	104.3	12.9	2.4	164.5	169.2	-4.7	4.1	3.1	108
1987	35.4	29.7	87.9	5.1	47.8	101.5	12.4	4.6	158.2	161.7	-3.6	4.3	3.9	100
1988	36.4	27.3	78.7	4.9	47.8	90.2	10.7	4.3	147.3	148.8	-1.5	2.9	2.9	92
1989	31.9	28.9	95.3	5.1	45.8	112.8	11.3	-0.3	161.2	169.8	-8.6	4.8	3.5	75
1990	31.4	33.0	112.7	6.1	47.8	141.0	14.1	-8.9	183.3	203.0	-19.7	5.8	5	75
1991	32.4	28.9	95.3	5.4	45.8	112.8	11.1	-0.7	162.0	169.7	-7.7	3.8	3.2	100
1992	31.9	26.4	90.9	5.9	41.6	110.0	9.7	-1.6	155.2	161.3	-6.1	2.8	1.8	83
1993	36.1	36.3	139.6	5.6	43.7	163.6	11.0	6.6	217.7	218.2	-0.5	3.8	3.3	75
1994	36.4	33.9	132.2	6.1	43.7	143.8	11.9	14.3	208.7	199.4	9.3	3.1	1.9	83
1995	35.9	31.4	101.4	4.9	43.7	118.4	10.9	4.6	173.5	173.0	0.6	2.5	1.6	92
1996	38.9	27.3	80.5	6.4	43.7	90.2	12.9	8.4	153.1	146.8	6.3	1.5	0.6	100
1997	35.9	29.7	94.4	4.9	39.5	112.8	11.1	5.1	164.9	163.4	1.5	2.3	1.3	92
avg 84-86	38.3	31.3	98.0	5.9	51.1	112.5	12.4	2.0	173.5	176.0	-2.6	3.6	2.2	97.2

Tovdal. Arithmetic mean concentrations of major ions in river water collected monthly (NIVA) and reported in SFT's annual reports. Station 7.1. Units: $\mu\text{eq/l}$, TOC mmol/l. Org⁻: organic anions; SBC: sum of base cations; SSA: sum of acid anions; ANC: acid neutralising capacity; Ali: inorganic aluminium.

Tovdal	Ca	Mg	Na	K	SO4	Cl	NO3	org-	SBC	SSA	ANC	H+	Ali	TOC
	$\mu\text{eq/l}$													
1980	47.9	26.4	62.6	7.7	91.5	70.5	15.3	-5.5	144.7	177.3	-32.6	14.1	13.0	
1981	48.9	26.4	63.9	9.0	87.4	70.5	9.7	2.6	148.2	167.6	-19.3	11.0	11.0	
1982	51.4	27.3	64.8	7.7	93.6	70.5	12.1	-1.9	151.2	176.2	-25.1	12.0	11.2	
1983	45.9	25.6	69.6	8.2	85.3	79.0	10.4	-0.4	149.3	174.6	-25.3	12.9	12.0	
1984	51.4	28.9	84.0	8.4	89.4	93.1	12.5	7.4	172.7	195.0	-22.3	13.5	16.2	342
1985	45.9	24.8	67.4	7.9	87.4	67.7	12.4	5.6	146.0	167.5	-21.4	13.2	13.8	308
1986	44.9	23.1	63.1	8.2	83.2	62.0	11.9	6.7	139.3	157.2	-17.9	11.5	13.1	317
1987	46.4	25.6	64.4	6.9	79.0	67.7	11.8	12.1	143.3	158.5	-15.2	12.6	14.7	308
1988	40.9	23.1	66.1	7.2	72.8	67.7	12.5	13.5	137.3	153.0	-15.6	13.5	15.7	308
1989	46.4	25.6	70.5	7.9	77.0	76.1	14.2	6.9	150.4	167.3	-16.9	11.2	12.6	217
1990	43.4	24.8	74.8	7.7	74.9	90.2	11.6	0.7	150.7	176.7	-26.0	12.9	13.8	258
1991	46.9	24.8	80.0	7.4	72.8	84.6	11.3	9.9	159.2	168.7	-9.5	8.9	10.5	242
1992	50.4	25.6	82.2	7.4	74.9	93.1	11.1	3.9	165.6	179.0	-13.4	8.5	8.8	317
1993	51.4	26.4	99.2	6.9	64.5	115.6	10.4	10.8	183.9	190.5	-6.5	7.6	9.8	217
1994	49.4	24.0	86.6	6.9	68.6	79.0	12.8	20.6	166.8	160.4	6.4	6.8	7.4	317
1995	48.9	24.0	82.2	7.2	64.5	81.8	11.8	18.2	162.2	158.0	4.2	6.3	7.7	308
1996	60.9	24.8	72.6	7.7	68.6	70.5	14.6	20.6	166.0	153.7	12.3	3.5	4.9	292
1997	96.8	24.0	70.5	6.9	58.2	76.1	9.4	55.8	198.1	143.8	54.3	0.6	0.9	333
avg 80-82	49.4	26.7	63.8	8.1	90.8	70.5	12.4	-1.6	148.0	173.7	-25.7	12.4	11.7	
avg 84-86	47.4	25.6	71.5	8.2	86.7	74.3	12.3	6.6	152.7	173.2	-20.5	12.7	14.4	322
avg 93-95	49.9	24.8	89.3	7.0	65.9	92.1	11.6	16.6	171.0	169.6	1.4	6.9	8.3	281

Appendix D. Naturens Tålegrenser - Oversikt over utgitte rapporter

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