Acid Rain Research

REPORT 23/1990

Reversibility of acidification: soils and surface waters

Risdalsheia

alkalinity

observed

MAGIC

treatment

1984   1986   1988   1990   1992

ueq/l

0
-50
-100
-150

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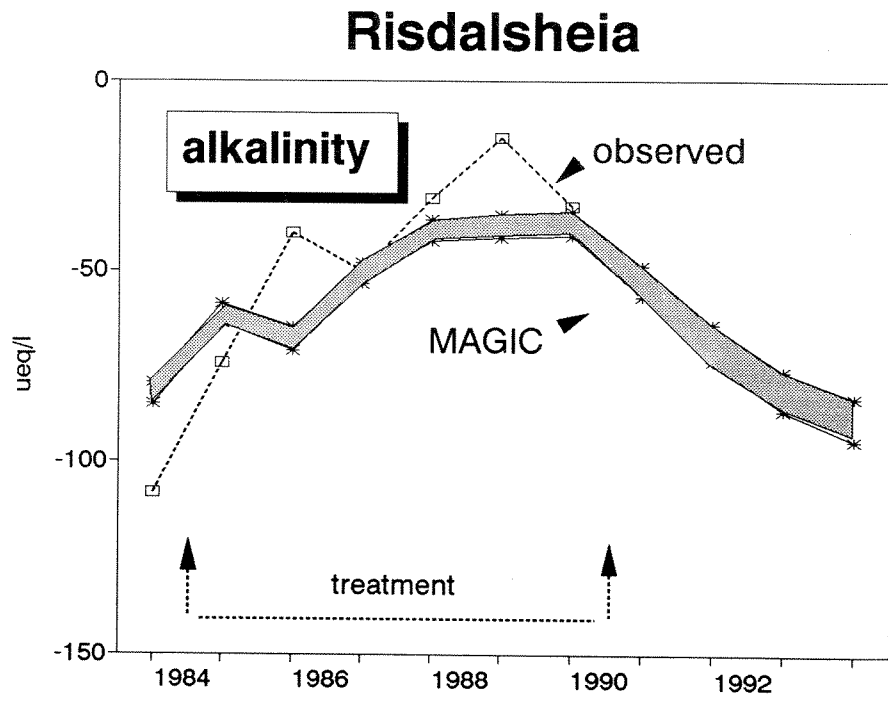
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Discrepancies between observed and predicted effects are due in part to the increasingly important role of nitrogen in soil and water acidification. "Nitrogen saturation" threatens to offset the incipient recovery due to reductions in SO_2 emissions. Possible ecosystem impacts of future climate change add further uncertainty to predictions of long-term acidification trends.

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Reversibility of acidification:
soils and surface waters

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Abstract

As the empirical relationships between acid deposition and the effects on soils and surface waters has become increasingly well understood, the focus has shifted to the reversibility of acidification in response to decreases in acid deposition.

Recent declining levels of acid deposition in both North America and Europe have initiated reversal of acidification and recovery of impacted ecosystems. The reversibility of acidification has also been investigated by means of large-scale experiments with whole ecosystems. Predictive models can account for much of this empirical and experimental data, at least at coarse spatial and temporal scales.

Discrepancies between observed and predicted effects are due in part to the increasingly important role of nitrogen in soil and water acidification. "Nitrogen saturation" threatens to offset the incipient recovery due to reductions in SO_2 emissions. Possible ecosystem impacts of future climate change add further uncertainty to predictions of long-term acidification trends.

1. Introduction

The past 20 years of research on acid precipitation and its effects on terrestrial and aquatic ecosystems has provided copious documentation of soil acidification, freshwater acidification, and the impacts on terrestrial and freshwater organisms. The progress of understanding the cause-effect and dose-response relationships is documented in the proceedings of the major international conferences on acid deposition and its ecological impacts held in 1975 at Columbus, USA (Dochinger & Seliga 1976), in 1980 at Sandefjord, Norway, in 1985 (Drabløs & Tollan 1980), at Muskoka, Ontario, Canada (Martin 1986), and in 1990 at Glascow, UK. The focus of this research has moved from the documentation of the nature and extent of the effects, to elucidation of the mechanisms, and finally today to the dynamics of the historical development and prediction of future effects.

Much of the research has taken a "black-box" approach to whole ecosystems (Figure 1). Two of the three of inputs, processes and outputs must be known to infer the third. Usually the inputs are measured, the outputs are measured and the processes occurring within the ecosystem are inferred (Case A). Alternatively the inputs are measured, the processes within the ecosystem are known and the outputs are predicted (Case B). Or finally the processes are known, the outputs are measured, and the inputs inferred (Case C). This whole-ecosystem approach was pioneered at the Hubbard Brook Experimental Forest, New Hampshire (Likens et al. 1977).

Acid deposition research to a large extent has followed the "black-box" approach (Case A). Two types of input-output relationships are used: (1) empirical **spatial** relationships between acid deposition, sensitivity maps and the regional patterns of surface water acidification and damage to fish populations, and (2) empirical **temporal** relationships between emissions history, acid deposition trends and historical response of fish populations and paleolimnological indicators such as diatoms.

Together these empirical relationships have led to the identification of a few key soil processes active within the "black-box" that largely regulate the response of terrestrial

ecosystems to inputs of acid deposition (Reuss & Johnson 1986). These key processes are linked mathematically in intermediate-complexity process-oriented models such as MAGIC (Model for Acidification of Groundwater In Catchments) (Cosby et al. 1985a, 1985b). Such models work well at coarse spatial and temporal scales (whole-catchments or regions and years-to-decades), but less well at fine scales (soil profiles and days-to-months).

Any decision on mitigation strategies for affected ecosystems requires knowledge about the expected response to reduced deposition of acidifying substances from the atmosphere. Although the response to increased deposition over the last 150 years has been extensively documented, there are few cases of response to decreased deposition. Most acidification studies have been of insufficient duration to evaluate the rate of response and the time lags; typically field data are extensive in space but sparse through time.

Reversal of acidification thus is a Case B phenomenon. Can the response to changed acid inputs be predicted by process-oriented models? Whereas the formulation of models from the empirical input-output data entails advances in the scientific understanding of the dynamics of acidification, the use of such models for prediction of future effects is a technological application (Hauhs in press). Here large-scale experiments with whole ecosystems play an important role by providing the means to directly study dose-response relationships and to quantify the dynamics of acidification. Furthermore results from such large-scale experiments provide critical tests of the predictive ability of models.

Here we examine the dynamics of acidification of soils and waters and the links between acid deposition and terrestrial and aquatic ecosystems (Figure 1). The analysis will consider historical trends, reversibility and recovery, and predictions for the future.
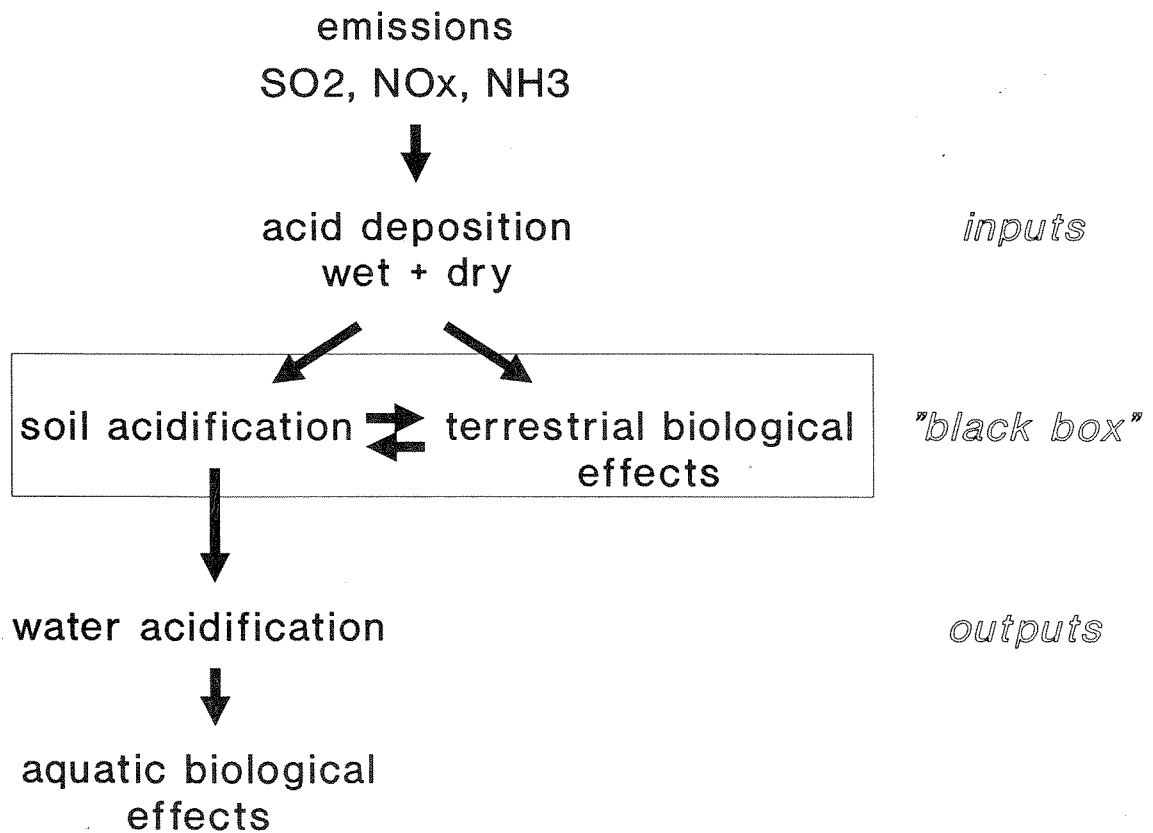
Figure 1. Causal chain linking emissions of SO_2 and NO_x to soil acidification, forest effects and aquatic effects. Reversibility of acidification may be delayed at one or more links. Some of the effects caused by long-term acid deposition may even be irreversible. A traditional approach to the study of cause-effect relationships has been to treat the terrestrial ecosystem as a "black box". Inputs and outputs are measured and used to deduce processes occurring within the box (from Hauhs & Wright, 1987).

2. Soil chemical processes -- keys to water acidification

The key to surface water acidification lies in soil chemical processes; most of the incoming strong acids from the atmosphere are deposited on terrestrial catchments. (This is not the case for perched seepage lakes). Soil processes are thus central to understanding the changes introduced by acid deposition (Reuss & Johnson 1986) and hence the reversibility of acidification.

Much of the empirical evidence for soil and water acidification can be accounted for by only a few soil processes (Reuss & Johnson 1986, Reuss et al. 1987, Cosby et al. 1985a). These are cation exchange, chemical weathering, sulfate adsorption, dissolution and precipitation of aluminum, and dissolution of CO_2. In addition come flux parameters such as ionic inputs from the atmosphere, net uptake by terrestrial biomass, and loss to runoff. These processes can be coupled together in process-oriented models to provide quantitative explanation of observed spatial and temporal patterns of soil and water acidification (Cosby et al. 1985a, 1985b, Reuss et al. 1986).

Water acidification is loss of alkalinity. We follow the convention of Reuss & Johnson (1986) and define alkalinity of soil solution or surface water by either side of equation (1):

$$(Ca^{2+} + Mg^{2+} + Na^+ + K^+ + NH_4^+) - (SO_4^{2-} + NO_3^- + Cl^- + F^-) \quad (1)$$
$$= (HCO_3^- + A^-) - (H^+ + \Sigma Al^+)$$

where A^- denotes organic anions and ΣAl^+ denotes the sum of positively-charged aluminum species. Concentrations of OH^-, CO_3^{2-}, and other ions are generally low in soil- and surface-waters sensitive to acidification and thus can be neglected.

All the terms on the left side of equation (1) are independent of CO_2 partial pressure, whereas the right side includes all compounds that change when soil solution is degassed and becomes surface water. Alkalinity by this definition can

be used to estimate the transport of acidity from the soil to surface waters.

Acidification of soil and water requires the presence of mobile anions to mobilize and transport acid cations (Gorham 1958, Seip 1980). Only 6 anions need be considered: the strong acid anions Cl^-, F^-, NO_3^- and SO_4^{-2}, organic anions (lumped here as A^-), and bicarbonate HCO_3^-. Bicarbonate is the anion of a weak acid and thus not available for transport of acid cations at pH levels below about 5.

Of the strong acid anions Cl^- is the most highly mobile, usually following water through the terrestrial ecosystem to runoff. The major source of Cl^- is cyclic seasalts, a neutral salt mixture, and deposited in wet and dry precipitation.

Fluoride is also highly mobile but because F^- is generally present in low concentrations, it plays a minor role as a mobile anion.

Although NO_3^- is extremely mobile in soils, nitrogen is often a growth-limiting nutrient in temperate terrestrial ecosystems, and thus usually most of the incoming nitrogen is taken up by the terrestrial biomass. Nitrate concentrations in runoff are commonly low, and typically nitrate is not an important anion with respect to acidification. The exceptions, usually associated with some type of disturbance, will be discussed later.

Sulfate is, of course, the mobile anion of central interest in acidification. Acid deposition increases the external source of SO_4^{2-} and the accompanying cations are mainly acidic (H^+ and $NH4^+$). SO_4^{2-} in neutral cyclic seasalts is normally the only other source of importance. The non-marine fraction is denoted by asterisks. Atmospheric inputs generally far exceed the biological sulfur requirement.

The mobility of SO_4^{2-} in soils is strongly dependent on the characteristics of the soils. Adsorption may immobilize a significant fraction of the incoming SO_4^{2-} and delay acidification by decades to even centuries. For example, at White Oak Run, a small forested catchment in the Shenandoah National Park, Virginia, USA, for
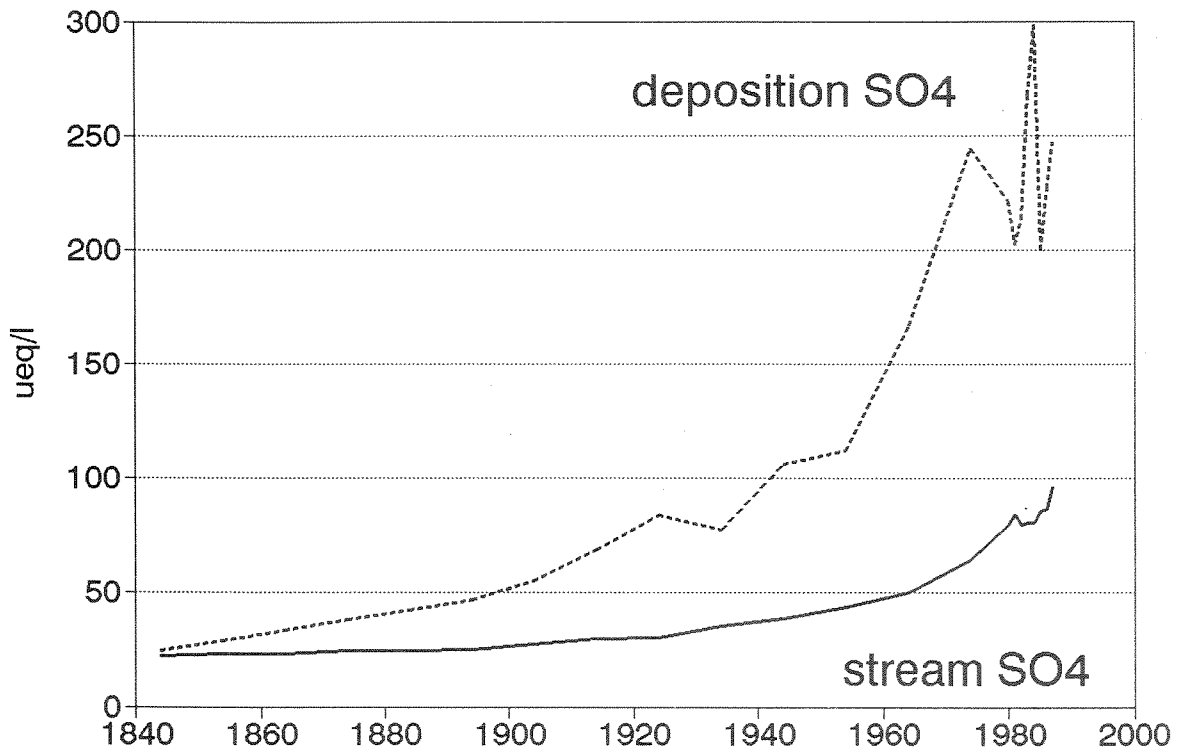
White Oak Run, Virginia, USA

Figure 2. Sulfate concentrations in total deposition and runoff at White Oak Run, Virginia, USA. Total deposition includes wet and dry and the total flux is divided by annual runoff volume to obtain concentration. Sea-salt contribution is low and has not been subtracted. The long-term historical trend of deposition is from estimated regional emissions for the southeastern US; the reconstructed SO_4^{2-} concentrations in streamwater are from MAGIC simulations (Cosby et al. 1986). Measured data for the period 1980-87 are from Ryan et al. (1989). The concentration of SO_4^{2-} in runoff is only about 40% that of deposition, due to substantial adsorption of sulfate in the catchment soils. Sulfate concentrations in runoff are increasing, however, at about 2 μeq/l per year as the SO_4^{2-} adsorption capacity becomes progressively saturated. Acidification increases at constant deposition.

example, SO_4^{2-} deposition (wet plus dry) is about 3 times that of SO_4^{2-} exported in runoff; the remaining 2/3 is retained in the catchment, most probably adsorbed on the soil (Figure 2) (Cosby et al. 1986). The SO_4^{2-} adsorption capacity is being used up, however, and the 8-year record from White Oak Run indicates that SO_4^{2-} concentrations in runoff are increasing at about 2 μeq/l annually (Ryan et al. 1989).

Old highly-weathered and leached soils rich in Al and Fe oxides typically have high SO_4^{2-} adsorption capacities (Johnson & Todd 1983). Such soils are widespread in regions that receive acid deposition such as the southeastern United States. Young soils in glaciated terrain, on the other hand, typically have low SO_4^{2-} adsorption capacity, and here the lag between increasing SO_4^{2-} deposition and response in leaching to runoff is much shorter (Rochelle et al. 1987). These soils are common in northeastern North America and much of northern Europe, regions that receive high levels of acid deposition (Johnson et al. 1980).

The mobility of organic anions is generally limited to the upper (O and A) horizons (Cronan et al. 1978). Organic anions play a major role in natural podolization processes in soils; they are present in high concentrations in organic-rich upper soil horizons, and precipitated out in lower mineral soil horizons. For the acid-base state of surface waters organic anions are important only when two chemical conditions are met simultaneously: their concentrations are high (i.e. colored waters) and the acid strength is sufficient (generally pK must be below 5) (Brakke et al. 1987, Reuss et al. 1987).

3. Reversibility -- the evidence

3.1 Sudbury, Ontario, Canada

That water acidification is reversible is indicated by both empirical and experimental data from North America and Europe. Lakes near the large copper-nickel smelter complex at Sudbury, Ontario, Canada, provide perhaps the clearest example.

Here local emissions of SO_2 have declined from about 1.5 millon tons in the early 1970's to 0.5-0.8 millon tons in the early 1980's (Figure 3a) (LaZerte & Dillon 1984, Dillon et al. 1986). In addition the installation of the nearly 400-m high stack in 1972 has substantially reduced local dry deposition of SO_2. In the early 1970's these local emissions accounted for about 50% of the acid deposition in the area; the remainder is long-range transport. The exact amount of SO_4^{2-} deposition and the magnitude of the change is difficult to assess because most deposition near Sudbury comes as dry deposition which is notoriously difficult to measure.

Water chemistry of several lakes near Sudbury has responded dramatically to the reduced local emissions of sulfur. Sulfate concentrations in lakewater have decreased by 20-60%. At Clearwater Lake and Swan Lake the change in SO_4^{2-} is accompanied by decreased base cation concentrations, increased alkalinity, and higher pH levels (Figure 3b) (Dillon et al. 1986, MacIsaac et al. 1986, Dillon pers. comm.). The decrease in SO_4^{2-} concentrations is compensated in part (about 60%) by a decrease in base cation levels; the soils of the catchment are responding to the decreased flux of mobile strong acid anions by retaining a larger fraction of the base cations released by weathering (Figure 3c).

The general trends shown by these lakes near Sudbury are also evident in lakes over the entire region (Keller & Pitblado 1986, Keller et al. 1986). A survey of 209 lakes conducted in 1974-76 and again in 1981-83 shows a regional decline in SO_4^{2-} concentrations in lakes with high SO_4^{2-} concentrations (within about 120 km of the SO_2 point source at Sudbury); again the change in SO_4^{2-} is accompanied by an increase in pH (Keller & Pitblado 1986). Base cation data for the resurvey were not reported. Reduction of SO_4^{2-} deposition in this region has thus clearly reversed the acidification of the lakes.

3.2 Muskoka, Ontario, Canada

Concurrent with the major decrease in SO_2 emissions from the Sudbury smelters, a general regional decline in SO_2 emissions over the past 20 years in eastern North

Sudbury, Ontario
local SO2 emissions

Clearwater Lake, Ontario
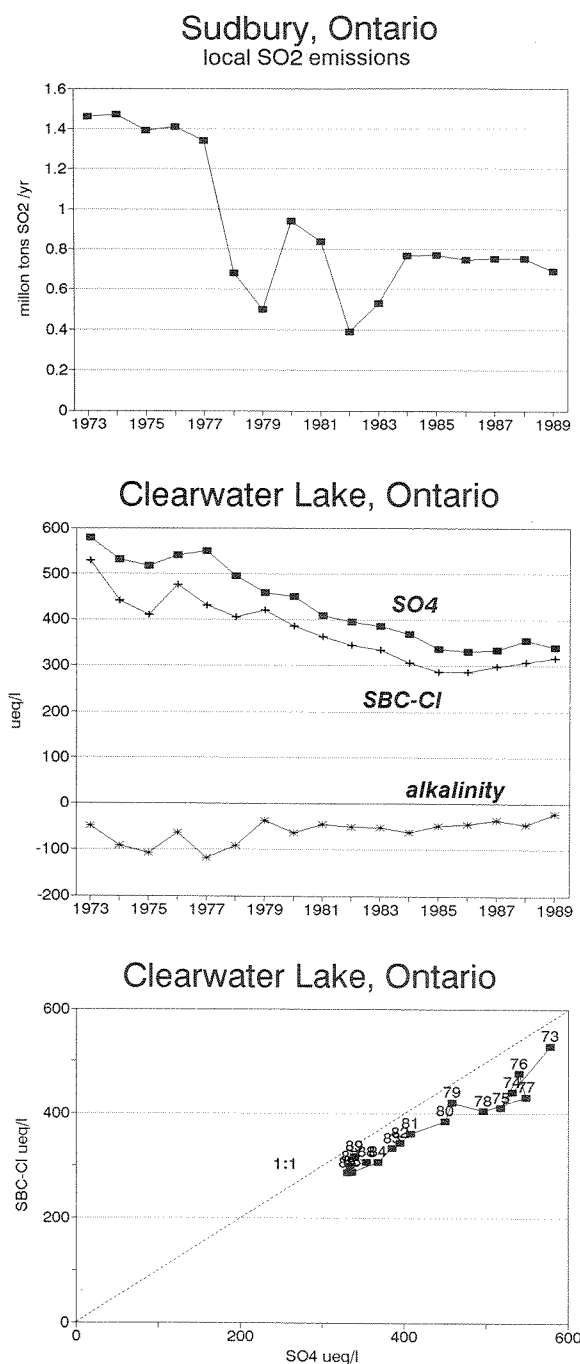

Clearwater Lake, Ontario

Figure 3. Emissions and lakewater chemistry at Clearwater Lake, near Sudbury, Ontario, Canada. a. Local SO_2 emissions from the metal smelters have decreased markedly during the period 1973-1989. Local deposition has decreased in parallel. b. SO_4^{2-} base cations, and alkalinity in nearby Clearwater Lake show substantial changes due to declining deposition. Sea-salt contribution is low and has not been subtracted. Cl is subtracted to account for the influence of road de-icing salts. c. Base cations vs. SO_4^{2-} follow a path with slope less than 1 suggesting that also the soils are recovering due to reduced deposition (modified from LaZerte & Dillon 1984, Dillon et al. 1986, and Dillon, pers. comm.).
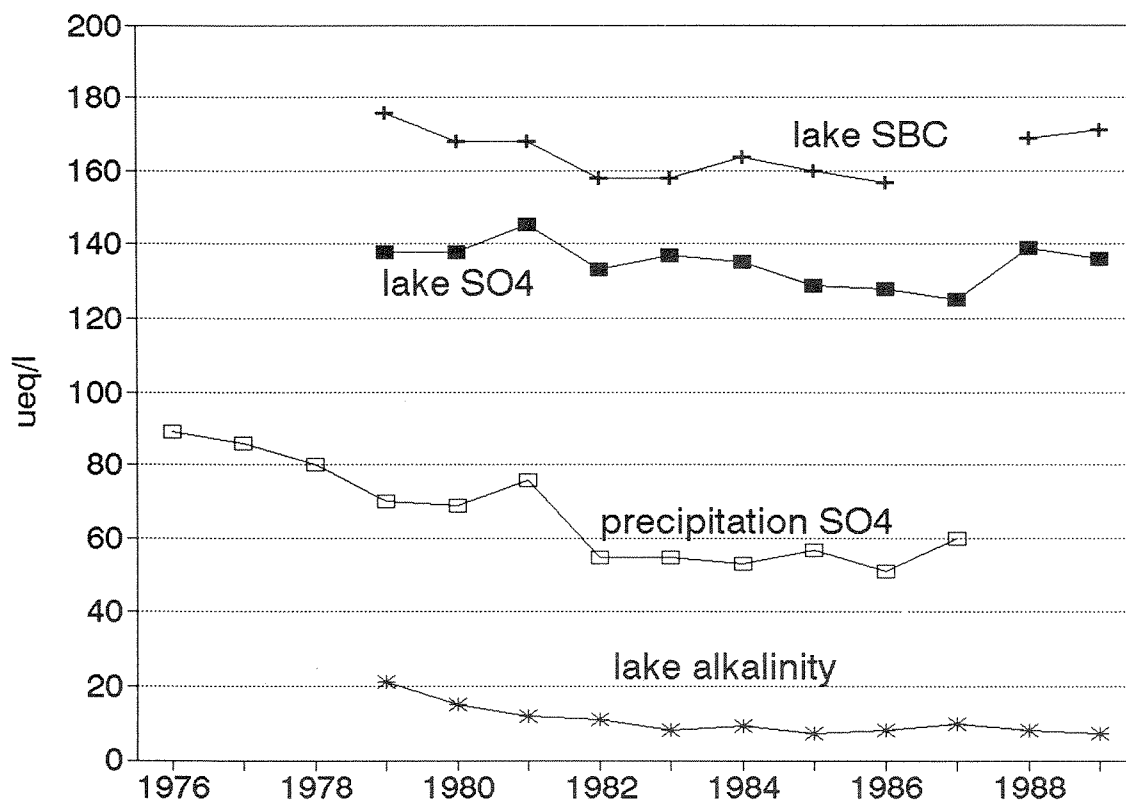
Plastic Lake, Ontario

Figure 4. Emissions, SO_4^{2-} deposition and lakewater chemistry at Plastic Lake in the Muskoka area, southern Ontario, Canada. a. Regional emissions of SO_2 in eastern North America have declined since the mid-1970's. b. SO_4^{2-} concentrations in precipitation show a parallel decline. Sea-salt contribution is low and has not been subtracted. During the 11-year period 1979-89 Plastic Lake has lost alkalinity, and base cation concentrations have decreased. Together these data point to continued acidification despite moderate decrease in acid deposition (from Dillon et al. 1987, Dillon pers. comm.).

America has occurred (Figure 4a). In the Muskoka-Haliburton region of southern Ontario this is reflected in a 30% decrease in SO_4^{2-} concentrations in precipitation over the period 1976-1987 (Figure 4b) (Dillon et al. 1987, Dillon pers. comm.).

The decrease in SO_4^{2-} deposition, however, has not yet resulted in a decline in SO_4^{2-} concentrations in Plastic Lake, a small headwater lake in the Muskoka-Haliburton region (Dillon et al. 1987, Dillon pers. comm.) (Figure 4c). The data record for Plastic Lake begins only in 1979, and the year-to-year variations apparently mask any long-term trend.

Concentrations of base cations, alkalinity and pH have declined significantly over this 11-year period (Figure 4c) (Dillon et al. 1987, Dillon pers. comm.). Together the data suggest that continued acid deposition is acidifying the soils by depleting the base cation pool, with the result that the SO_4^{2-} in runoff is to an increasing extent accompanied by acid cations. Here a moderate reduction in deposition is apparently insufficient to reverse the ongoing acidification of both soils and surface waters.

3.3 Hubbard Brook Experimental Forest, New Hampshire, USA

Sulfate deposition has also been declining at the Hubbard Brook Experimental Forest (HBEF), New Hampshire, USA (Figure 5a) (Likens et al. 1984, Hedin et al 1987, Driscoll et al. 1989). At Hubbard Brook an exceptionally long and continuous record of precipitation and streamwater chemistry is available. Annual volume-weighted, mean SO_4^{2-} concentrations in the stream draining Watershed 6 (unaffected by recent silvicultural activities) have declined about 30 % during the period 1970-87 (Figure 5b). The decline in sulfate is balanced principally by a decline in base cations, and the long-term data show no change in pH (Figure 5b). The data also show a period of elevated concentrations of NO_3^- in streamwater during the 1970's (Figure 5b).

The decline in acid deposition over the past 20 years has apparently been too little

Hubbard Brook
bulk precipitation
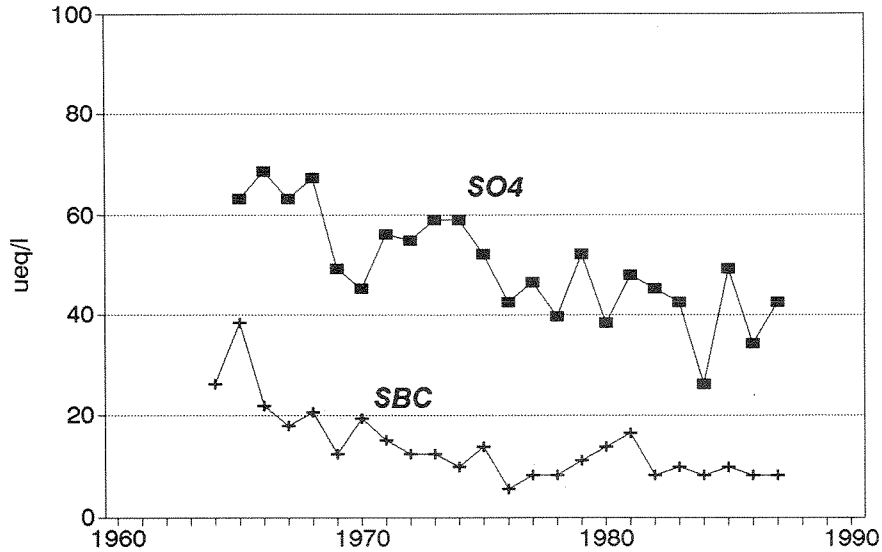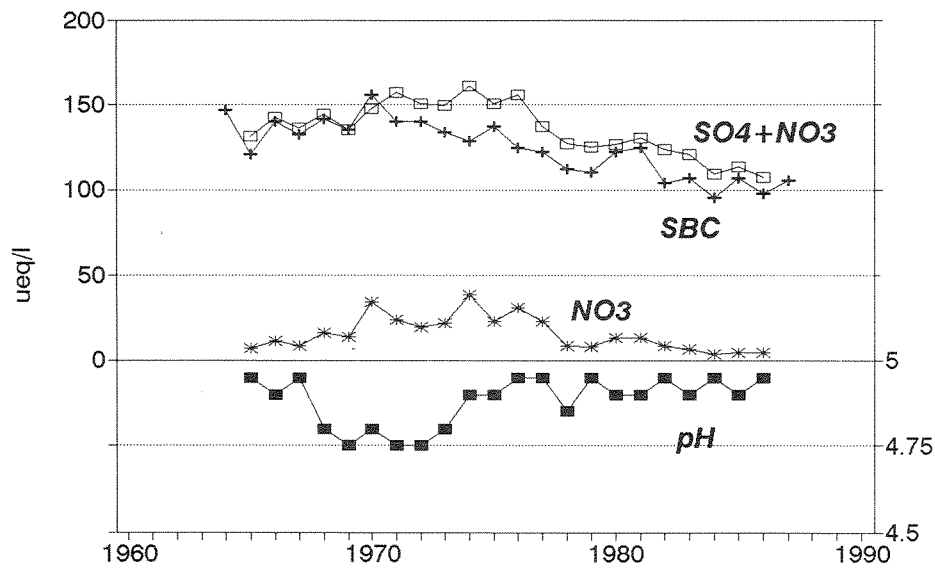

streamwater

Figure 5. Deposition and streamwater chemistry at Hubbard Brook Experimental Forest, New Hampshire, USA. Sea-salt contribution is low and has not been subtracted. a. SO_4^{2-} and base cation concentrations (SBC) in bulk deposition show significant decline over the 24-year record. b. SO_4^{2-} and base cations have also declined in streamwater at watershed #6, undisturbed by silvicultural activities. pH levels have not changed. Acidification has not been reversed; the decline in acid deposition has apparently been offset by continued soil acidification (data from Driscoll et al. 1989, Driscoll et al. 1990).

and too slow to reverse the acid-deposition induced soil acidification. The pool of exchangeable base cations in the soil is replenished by inputs from the atmosphere and from chemical weathering of soil minerals, and depleted by biological uptake in the aggrading forest and by leaching to runoff. When loss to biomass and runoff exceeds the inputs from the atmosphere and weathering, soil acidification occurs. At Hubbard Brook the flux of the mobile acid anions (mostly SO_4^{2-}) through the soils removes base cations at rates apparently greater than the replenishment. Although SO_4^{2-} flux has declined over the past 20 years, it is still greater than or perhaps equal to than the rate of replenishment. The fact that base cation inputs from the atmosphere have also declined over this same period exacerbates the situation; a more substantial reduction in SO_4^{2-} deposition is required to reverse soil acidification.

3.4 Other examples from North America

The decreased SO_4^{2-} deposition in northeastern North America has resulted in commensurate declines in SO_4^{2-} concentrations in many lakes and streams located north of the limit of Pleistocene glaciation. Several rivers in the United states also show close correlation between regional emissions of SO_2 and SO_4^{2-} concentrations in riverwater for the period 1967-1980 (Smith & Alexander 1986). For two rivers the declining SO_4^{2-} concentrations are accompanied by increased alkalinity and decreased base cation concentrations (National Research Council 1986). Similarly 12 rivers in Nova Scotia and 8 rivers in Newfoundland show a 45 % decrease in SO_4^{2-} transport over the period 1971-73 to 1982-83; this change is attributed to the 25 % decrease in SO_2 emission in the eastern United States and Canada over the same period (Thompson 1987). Together these data indicate that the decrease in SO_2 emissions has resulted in a regional decline in SO_4^{2-} concentrations in surface waters in glaciated areas of eastern North America.

3.5 Galloway area, southwestern Scotland

European emissions of SO_2 have declined slightly since the late 1970's, in part due

to national commitments by the "30% club", an international agreement by most European nations to reduce emissions of SO_2 by 30% over the period 1980 to 1993. The effects of these reductions are beginning to be picked up by various environmental monitoring networks.

In the Galloway area of southwestern Scotland the recent decline in regional emissions of SO_2 has resulted in declines in SO_4^{2-} deposition and SO_4^{2-} concentrations in lakes and has reversed the long-term acidification trends (Battarbee et al. 1988, Harriman 1989, Harriman pers. comm.).

The Galloway region is a classic acidified lake area; bedrock is granitic, soils are thin and organic-rich with low base saturation, and freshwaters are low-ionic strength and sensitive to inputs of strong acids from the atmosphere (Wright et al. 1980). The region has received high loadings of acid deposition for many decades; pioneering studies by Gorham (1958) in the nearby English Lake District indicate substantial acidification already in the 1950's.

Concentrations of non-marine sulfate in precipitation in the region have declined by about 35% over the period 1979-1989. This decrease is due to a large extent to the 30% decline in emissions of SO_2 in the UK over the same period (Figure 6) (Barrett et al. 1987, Harriman pers. comm.).

At Galloway there are three independent sets of data that all point to reversibility of acidification of lakes on a regional scale. First a 10-year record of lake water chemistry from 7 headwater lakes indicates declining SO_4^{2-} concentrations accompanied by increases in alkalinity and pH (Figure 7) (Battarbee et al. 1988, Harriman 1989, Harriman pers. comm.). Detailed paleolimnological investigations of diatom remains in lake sediments also point to increasing pH levels in several lakes during the 1980's (Battarbee et al. 1988). Finally a regional survey of 52 lakes conducted in 1979 and repeated in 1988 reveals consistent regional changes in lake chemistry that can be ascribed to the declining SO_4^{2-} concentrations in precipitation (Figure 8).
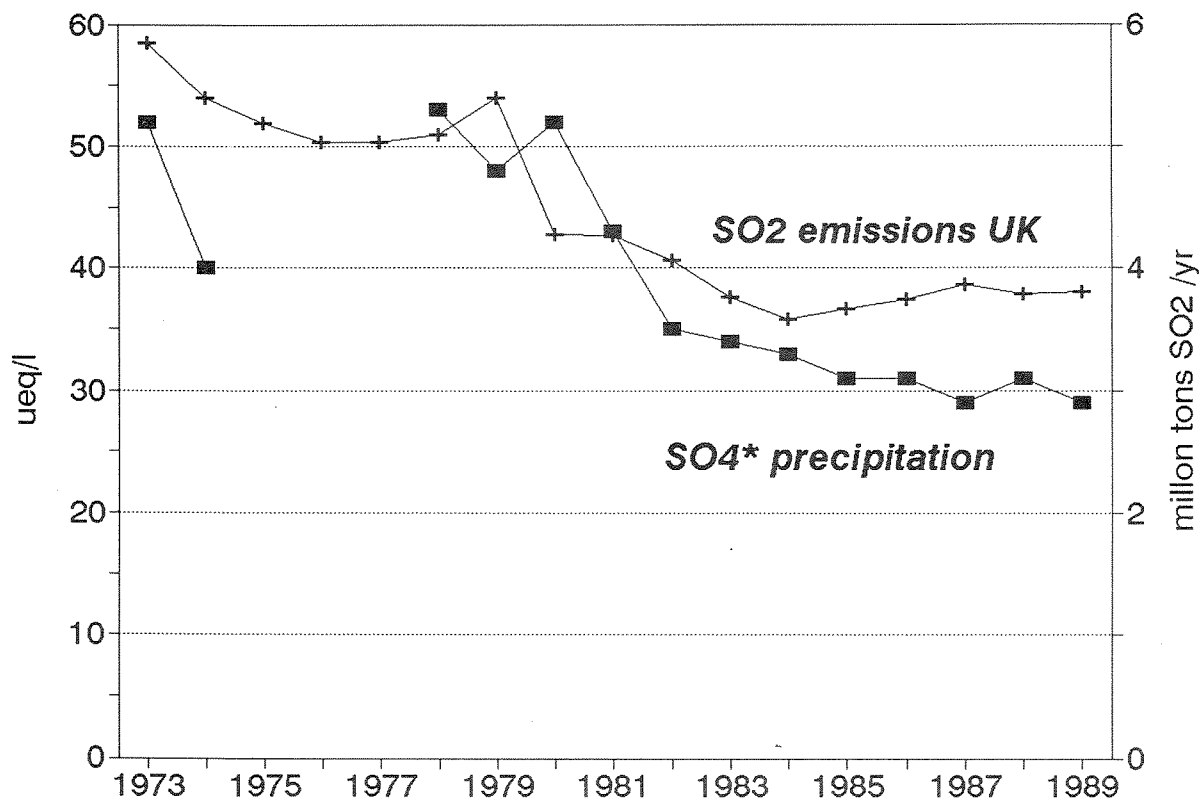
Galloway, Scotland

Figure 6. Emissions of SO_2 in the United Kingdom and SO_4^{2-} concentrations in wet precipitation at Eskdalemuir, southern Scotland over the period 1973-1989. The major decrease in emissions is reflected in deposition (from Barrett et al. 1987, Harriman pers. comm.).
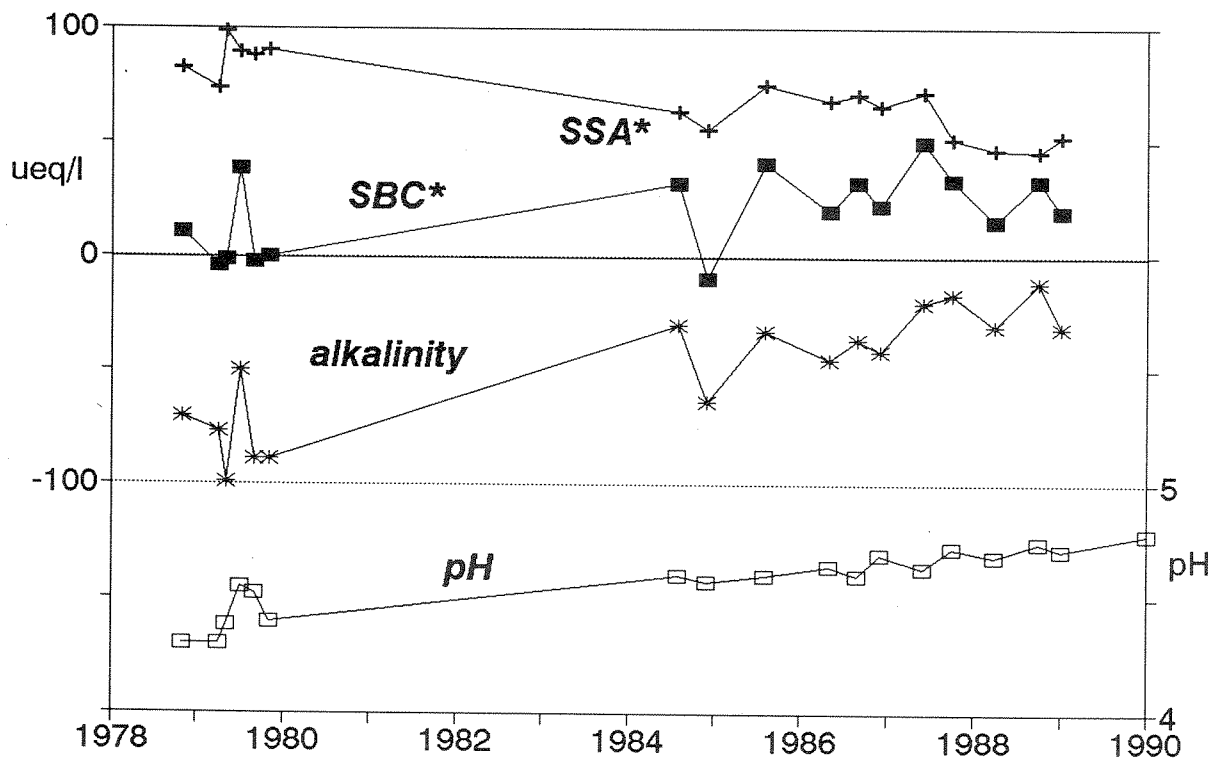
Loch Enoch, Scotland

Figure 7. Loch Enoch, Galloway area, southwestern Scotland. a. Concentrations of strong acid anions (SSA*, corrected for seasalts), base cations (SBC*, corrected for seasalts) and alkalinity all show major change in response to decreasing acid deposition. b. pH levels have increased from about 4.3 to 4.7, and as a result the diatom flora has changed (from Battarbee et al. 1988, Harriman 1989, Harriman pers. comm.).
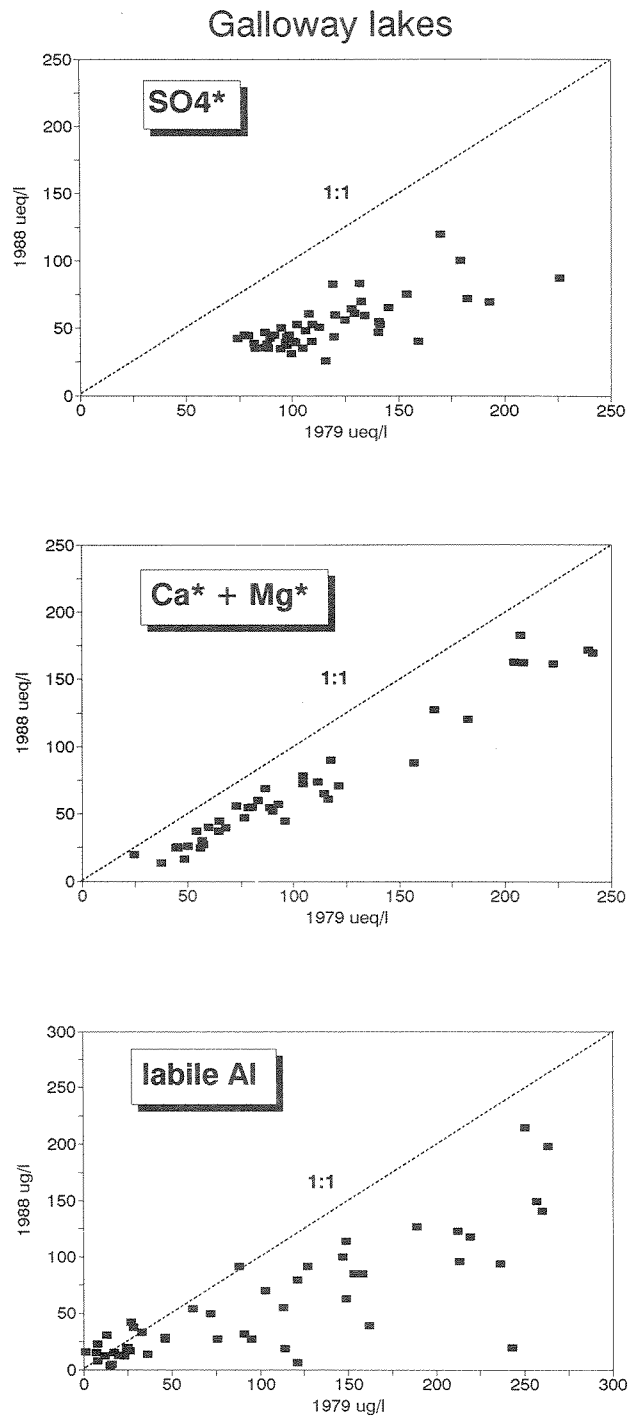
Galloway lakes

Figure 8. Regional surveys of 52 lakes in the Galloway area conducted in 1979 and again in 1988 reveal major changes in water chemistry that can be ascribed to declining acid deposition over this period. a. SO_4^{2-} concentrations (corrected for seasalts). b. Ca + Mg concentrations (corrected for seasalts). c. Labile Al concentrations.

3.6 Southern Norway

Regional surveys of lakes in southernmost Norway conducted in 1974 and repeated in 1986 indicate a modest decline in SO_4^{2-} concentrations (Henriksen et al. 1988). The decline in SO_4^{2-}, however, has been nearly exactly offset by an increase in NO_3^- concentrations. Thus the total concentration of strong acid anions (corrected for seasalts) has not changed significantly. This means that soil acidification continues, and indeed the concentrations of base cations and alkalinity in lakewater have decreased significantly.

3.7 Experimental evidence -- RAIN project

Large-scale experiments with whole ecosystems demonstrate the reversibility of soil and water acidification following drastic reduction in acid deposition. As part of the RAIN project (Reversing Acidification In Norway) a 860-m^2 catchment at Risdalsheia in southernmost Norway has been covered by a transparent roof to exclude ambient acid precipitation; clean rain with natural levels of seasalts is sprayed underneath the roof (Wright et al. 1988). Loading of acid, SO_4^{2-}, NO_3^- and NH_4^+ has thus been experimentally reduced by about 80%; the remaining 20% comes from dry deposition of gases and particles. An adjacent catchment also covered by roof receives recycled ambient acid precipitation and serves as control. Risdalsheia is located in Norway's zone of maximum acid deposition, and surface waters in the region are highly acidic; 90% of the lakes have lost their native fish populations (Sevaldrud et al. 1980).

After 6 years of acid exclusion concentrations of SO_4^{2-} in runoff have declined relative to the control catchments by about 70% from $\approx$ 105 μeq/l to $\approx$ 35 μeq/l (Figure 9). Both NO_3^- and NH_4^+ have also declined by >70%. The decline in strong acid anions of 95 μeq/l has been compensated by a decline in base cations of 36 μeq/l and an increase in alkalinity of 59 μeq/l. The F-factor as defined by Henriksen (1980) ((Ca^* + Mg^*)/ SO_4^*) is 0.2 (Wright et al. 1988).
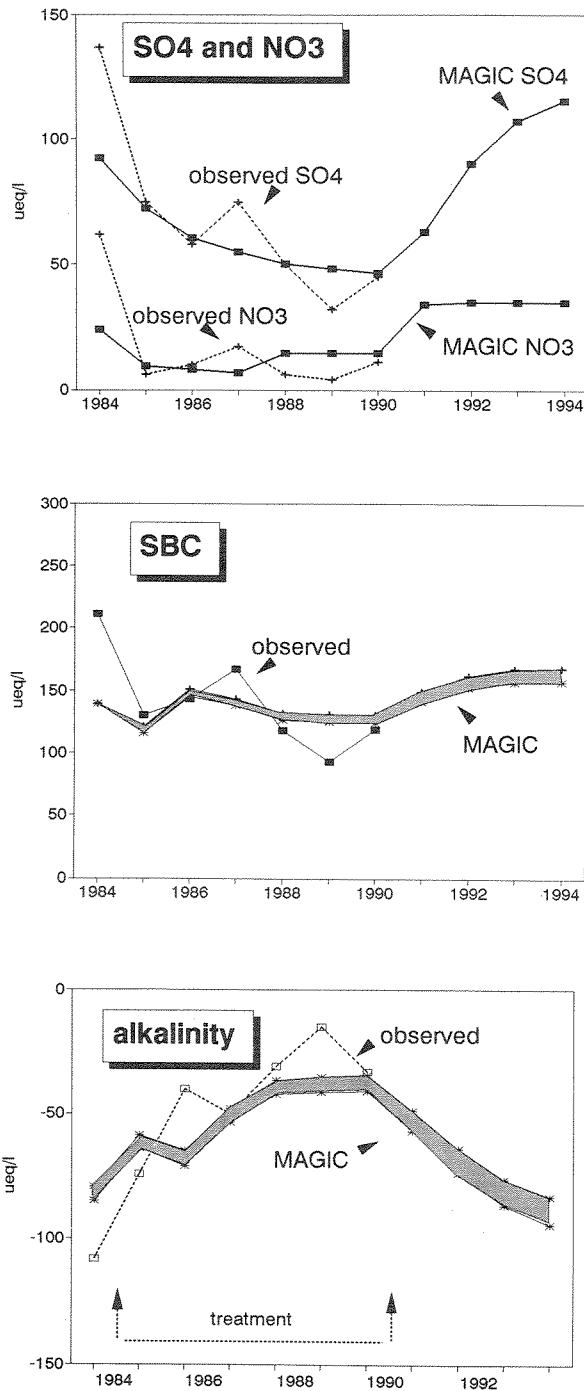
Risdalsheia

Figure 9. The acid-exclusion experiment of the RAIN project at Risdalsheia, southernmost Norway. SO_4^{2-} deposition is dramatically reduced by means of roof from about 60 meq/m²/yr to 16 meq/m²/yr. SO_4^{2-} concentrations in runoff have declined. Base cations concentrations have declined. Seasalt fractions are not subtracted. Alkalinity has shown major increase (from Wright et al. 1988). Also shown are the changes simulated by MAGIC, a process-oriented model of soil and water acidification. The bands indicate uncertainty in model parameters and measurements (from Wright et al. 1990).

Input-output budgets indicate that the catchment is now accumulating base cations; the long-term soil acidification has also been reversed (Wright et al. 1988).

4. Integration by MAGIC, an acidification model

The response of soils and surface waters to changes in acid deposition can be simulated with the help of acidification models. Several acidification models such as the Birkenes model (Christophersen et al. 1982), ILWAS (Gherini et al. 1985), and MAGIC (Cosby et al. 1985a, 1985b) have been used to simulate and predict acidification of soils and waters (Reuss et al. 1986).

MAGIC is an intermediate-complexity process-oriented model which lumps key soil processes at the catchment scale over monthly or yearly time-steps (Cosby et al. 1985a, 1985b). MAGIC is comprised of (1) a soil-soil solution equilibrium section in which the chemical composition of soil solution is assumed to be governed by SO_4^{2-} adsorption, CO_2 equilibria, cation exchange, and dissolution of aluminum hydroxide, and (2) a mass balance section in which the flux of major ions is assumed to be governed by atmospheric inputs, chemical weathering inputs, net uptake in biomass, and loss to runoff. Here we use two examples to illustrate the use of MAGIC for simulation of reversibility: the RAIN project and regional chemistry of lakes in southernmost Norway.

4.1 MAGIC applied to the RAIN project

The experimental data from the reversibility experiment of the RAIN project provide unique basis for the evaluation of predictive acidification models. Wright & Cosby (1987) conducted a preliminary application of MAGIC to data from the first 1 ½ years of acid-exclusion that was recently followed up by a more rigorous calibration and application to the 4-year record (Wright et al. 1990).

For the acid-exclusion experiment at Risdalsheia MAGIC satisfactorily predicts the observed trends in concentrations of the strong acid anions Cl^-, SO_4^{2-}, and NO_3^-

(Figure 9). Here the major changes have been declining levels of SO_4^{2-} and NO_3- in runoff. MAGIC also simulates well the observed increase in alkalinity over the 6-year experimental period (Figure 9). For both base cations and alkalinity the replications of the optimization procedure indicate relatively narrow bands of uncertainty related to estimation of weathering rates and initial base saturation (Wright et al. 1990).

Changes in both pH and Al^{3+} are also simulated well (Figure 9). Runoff at Risdalsheia contains high levels of dissolved organic matter which buffers pH. Organic acids are incorporated in MAGIC using the measured concentrations of total organic carbon with a charge density of 5 μeq/mgC and a pK of 4.5.

4.2 MAGIC applied to Norwegian lake surveys

The recovery of acidified lakes in entire regions following reduction in emissions of SO_2 can be also be simulated using conceptual models such as MAGIC. Wright et al. (in press) use a regional version of MAGIC that is first calibrated to data from the 1974-75 survey of lakes in southernmost Norway and then validated using data from the 1986 resurvey, to predict regional acidification in response to 30%, 50% and 70% reductions in acid deposition (Figure 10). The calibrated model qualitatively and quantitatively simulates the observed changes in water chemistry between the 1974-75 and 1986 surveys. The model indicates that a 30% deposition reduction is insufficient to reverse the ongoing regional acidification of lakes, whereas reductions of 50% and 70% result in recovery of progressively larger numbers of lakes (Figure 10b).

5. Discussion

Just as the rate of acidification of surface waters depends on the inherent sensitivity of the catchment soils, the rate of reversal following reductions in acid deposition also depends on the sensitivity and degree to which the system has been affected (Reuss et al. 1987). Prior to the onset of acid deposition most
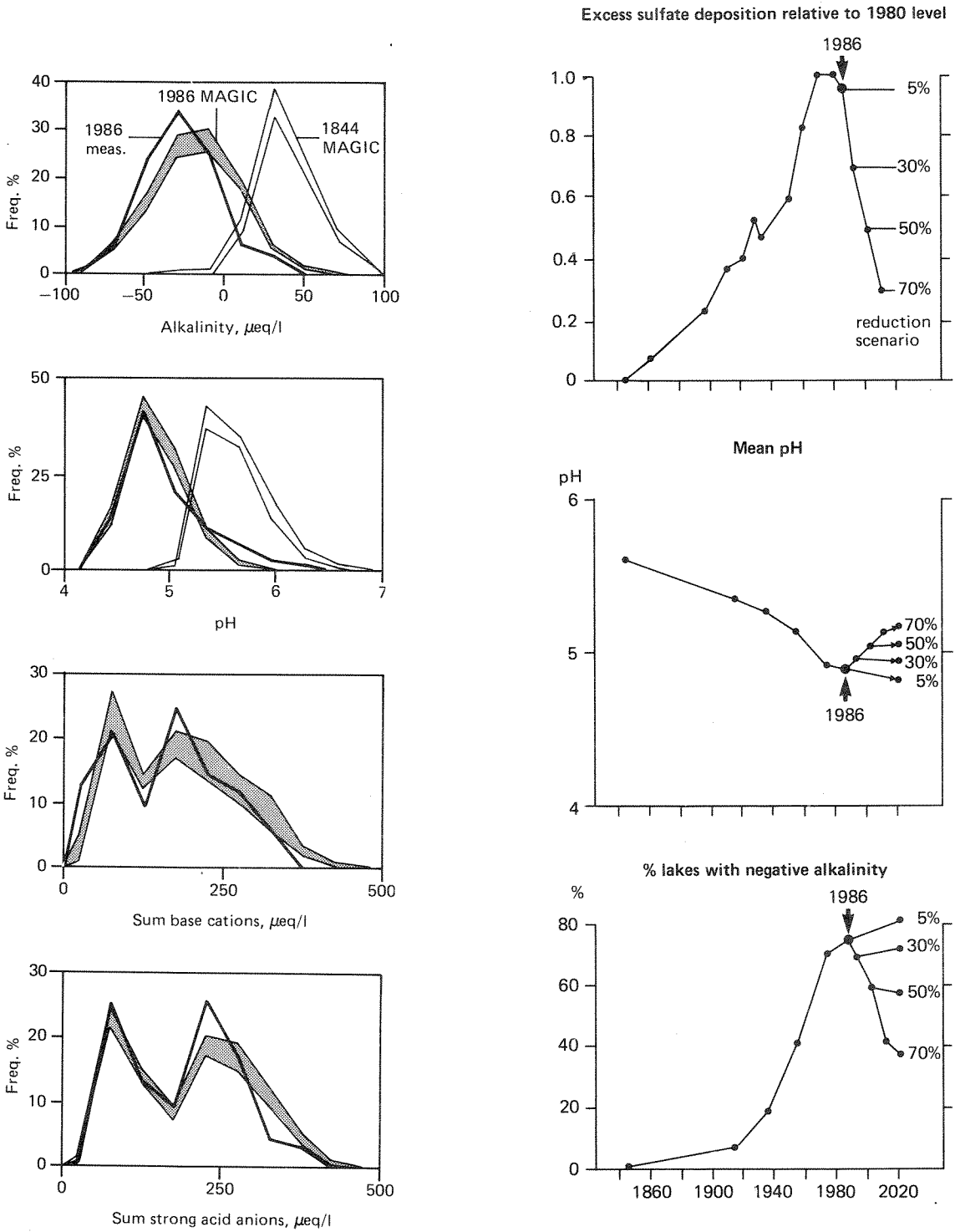
Figure 10. MAGIC applied to 180 lakes in southernmost Norway surveyed in 1974 and resurveyed in 1986. a. The model was calibrated to the frequency distributions measured in 1974, validated by comparing predicted with measured 1986 data. b. The calibrated model was then used to predict future acidification in this lake region given several alternative future deposition scenarios (from Wright et al. in press).

ecosystems were probably at steady-state with outputs of base cations and SO_4^{2-} in runoff balanced by inputs from the atmosphere and from chemical weathering in the soils. Acid deposition has increased the flux of SO_4^{2-} to these systems and increased the flux of SO_4^{2-} in runoff to a greater or lesser extent depending on the SO_4^{2-} retention characteristics of the catchment and surface water.

Catchments that retain significant amounts of incoming sulfur will continue to acidify even if acid deposition is reduced, unless such reductions reach levels at which SO_4^{2-} inputs are lower than outputs (Reuss et al. 1987). Here the example of White Oak Run in Virginia, USA, provides a good example.

The increased flux of SO_4^{2-} in runoff in turn increases the flux of cations from the soil; the degree to which these cations are base cations or acid cations depends on the base saturation of the soil. Soils with moderate to high base saturation will lose mostly base cations (Henriksen's F-factor will be high). The base saturation of such soils will thus decrease as acid deposition continues. Such soil acidification has been demonstrated by experiments in which soil plots or lysismeters are treated with strong acids (Abrahamsen & Stuanes 1980, Bjør & Teigen 1980, Lee & Weber 1982, Brown 1987, Abrahamsen et al. 1987, Stuanes et al. 1987) as well as from field studies in Sweden (Tamm & Hallbäcken 1986, Falkengren-Grerup et al. 1987) and Germany (Matzner & Ulrich 1984, Hauhs 1990).

A moderate decrease in acid deposition may not be sufficient to reverse soil acidification at such systems (Table 1). The remaining levels of mobile SO_4^{2-} will continue to acidify the soil, albeit at a lower rate. And as the base saturation declines, the runoff will also acidify (Reuss et al. 1987). This is apparently the case at Plastic Lake, Ontario, Canada; here base cations, alkalinity and pH have decreased over the period 1979-85, even though deposition has been reduced over the same period (Figure 4) (Dillon et al. 1987).

Soils with low base saturation will lose acid cations and give rise to acidic

| Deposition | Start year | End year | No. years | SO4* deposition meq/m2/yr | | | Surface water ueq/l | | | | | | F obs. | Acidification trend, water |
				start	end	change	start year Ca*+Mg*	SO4*	alk	end year Ca*+Mg*	SO4*	alk		
Constant														
White Oak Run	1980	1987	8	100	115	15%	63	77	24	67	94	12	0.24	acidifying
Slight decrease														
Plastic L.	1979	1989	11	77	62	-19%	146	137	21	141	135	7	2.50	acidifying
Large decrease														
Clearwater L.	1973	1989	17	140	100	-29%	460	575	-49	365	313	-22	0.36	recovering
Hubbard Brook	1965	1986	22	135	90	-33%	76	124	-25	63	102	-23	0.59	no change
Loch Enoch	1978	1988	11	75	50	-33%	19	62	-79	16	27	-25	0.09	recovering
RAIN KIM	1984	1989	6	58	16	-72%	22	125	-108	1	35	-33	0.23	recovering

Table 1. Summary of examples of acidification and reversibility of surface waters. The examples are grouped by acid deposition trend over the period of measurement from constant and slightly increasing deposition, to slight decrease, and to large decrease. Together the data point to continued acidification with constant or even slightly decreasing acid deposition, but reversal and recovery in response to rapid decrease in deposition. Data sources given in text.

aluminum-rich runoff. Waters draining such soils are thus inherently sensitive to acid deposition. Sensitive systems can move relatively rapidly to a new steady-state following change in acid deposition (Reuss et al. 1987). Here the reversal of water acidification in response to decreased acid deposition can be prompt. The RAIN project experiment in southern Norway illustrates this rapid reversal (Wright et al. 1988).

In-lake processes may speed up the reversal of lake acidification following reductions in acid deposition. Sulfate reduction and retention in lake sediments is the process of potentially greatest importance. Schindler (1986) shows that SO_4^{2-} reduction can contribute significantly to alkalinity generation in lakes. Indeed Dillon et al. (1986) suggest that the rapid decrease in SO_4^{2-} concentration at Swan Lake near Sudbury, Ontario, Canada, is due in part to SO_4^{2-} reduction in the anoxic

hypolimnion. The relative importance of such in-lake processes is highly dependent on the flushing time of the lake; for lakes with rapid water renewal, in-lake processes are minor. Norton et al. (1988) show that for typical drainage lakes in the United States sulfur stored in lake sediments can account for only a few μeq/l decrease of SO_4^{2-} concentrations in lake water.

6. The future -- nitrate as a wildcard

The role of SO_4^{2-} in acidification of soils and waters and the reversibility of SO_4^{2-} - induced acidification is now well understood. The wealth of information regarding acid deposition inputs, the response of water chemistry and the key processes operating within the "black-box" are successfully modelled at the coarse scale. It is the lack of agreement between modelled and measured response, however, that results in the identification of additional processes of importance. Models advance understanding and point the way to future research. The increasingly central role of nitrogen in soil and water acidification is a case in point.

International negotiations to reduce the emissions of sulfur to the atmosphere have reached the stage at which the prognosis now is for continued declining deposition of sulfur in most acidified areas of Europe and North America in the near future. Because SO_4^{2-} is the mobile anion responsible for the bulk of acidification, this prognosis should mean reversal and recovery of acidified terrestrial and aquatic ecosystems. Increasing leaching of NO_3^- from terrestrial ecosystems, however, can offset the decreasing SO_4^{2-} levels.

6.1 Nitrate and forest decline

Nitrogen is commonly the growth-limiting nutrient in temperate coniferous forests. Nitrogen inputs in acid deposition should therefore be taken up in biomass and retained in the terrestrial ecosystem. There is a growing body of evidence, however, indicating that terrestrial ecosystems subjected to prolonged high loadings of acid sulfur and nitrogen compounds eventually reach "nitrogen

saturation" and an increasing fraction of the incoming nitrogen is leached to runoff instead of being retained. Furthermore "nitrogen saturation" is apparently a consequence of disturbance to the terrestrial ecosystem; the tight internal cycle of nitrogen is broken when the vegetation is disturbed by natural events such as fire and windthrow or by man-induced events such as clearcutting or forest decline (Vitousek et al. 1979, Ulrich 1990).

That forest decline is linked to increasing leaching of NO_3^- is well-illustrated by the 15-year record of forest damage symptoms, soil solution chemistry and runoff chemistry from Lange Bramke, a 0.76 km^2 coniferous-forested headwater catchment in the Harz Mountains, Federal Republic of Germany. This catchment receives high loadings of sulfur (45 kg S/ha/yr) and nitrogen (19 kg N/ha/yr) from the atmosphere. Nitrate concentrations in soil solution began to increase in the late 1970's, visible symptoms of forest damage were first recorded in 1983, and concentrations of NO_3^- in runoff have shown a steady increase during the 1980's. An adjacent catchment, Dicke Bramke, has progressed further; here outputs of NO_3^- now are about 50% of inputs (Hauhs 1990) (Figure 11).

Increasing concentrations of NO_3^- in surface waters have also been reported from other acidified areas in Europe. In Norway the 1000-lake survey revealed that during the period 1974-86 NO_3^- concentrations in acid lakes in southernmost Norway have approximately doubled (Figure 12), although nitrogen deposition has shown only modest increase over this time period (Henriksen et al. 1988). This NO_3^- increase almost exactly offsets the decrease in SO_4^{2-} concentrations.

Prediction of future nitrogen leaching from terrestrial ecosystems is difficult. Although most acidification models include the processes of nitrogen deposition, uptake and release cycle, none of existing models includes links between, for example, soil acidification and nitrogen uptake. There simply is not enough information available at present to formulate a predictive model for forest decline. Thus predictions of future acidification of soils and waters and the reversibility of acidification all are conditional on empirical assumptions of the future role of NO_3^-.
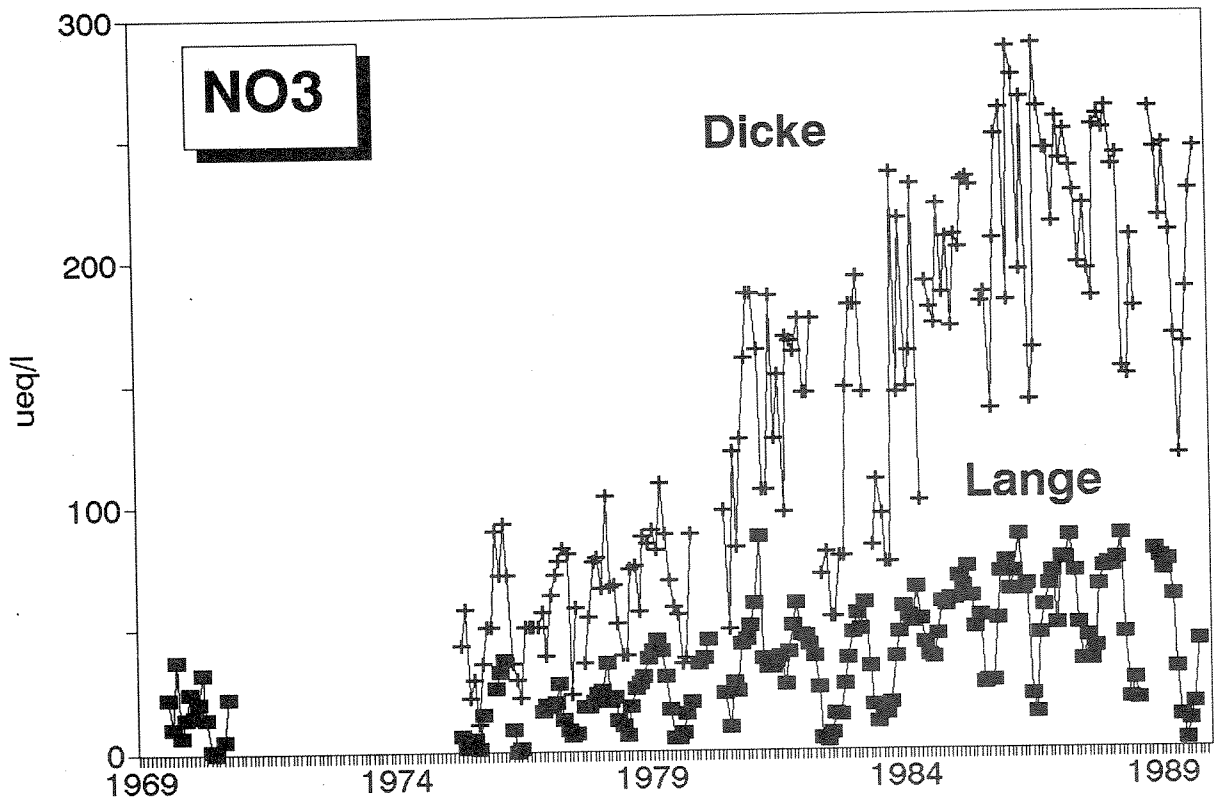
Lange and Dicke Bramke

Figure 11. Nitrate concentrations in runoff at 2 catchments in the Harz Mountains, Federal Republic of Germany. At Lange Bramke NO_3^- levels show steady increase beginning in the mid-1980's despite constant nitrogen deposition. At the adjacent catchment Dicke Bramke the increase began several years earlier and concentrations are now about 2x higher than Lange Bramke. Increased leaching of NO_3^- is apparently related to forest decline (from Hauhs 1990).
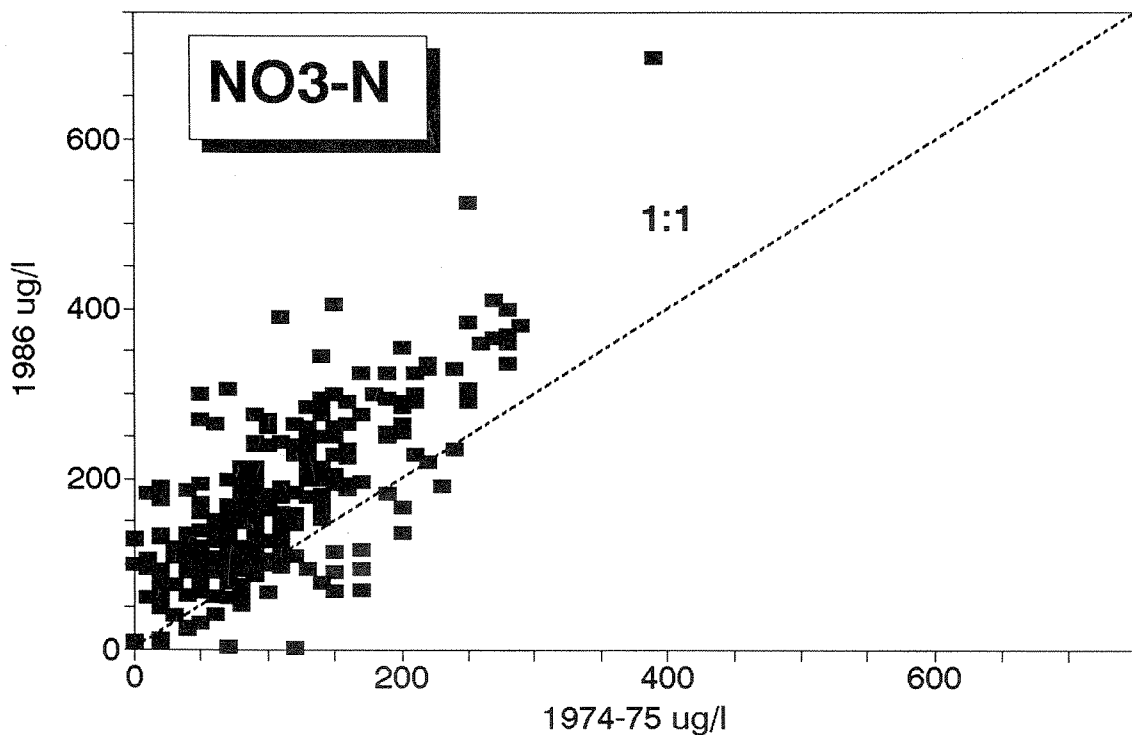
Norwegian lakes

Figure 12. Nitrate concentrations in lakes in southern Norway sampled in 1974 and resampled in 1986. Concentrations have approximately doubled over this 12-year period despite relatively constant nitrogen deposition; an increasing portion of incoming nitrogen is apparently being leached from the terrestrial catchments and the resultant acidification to a large extent offsets effects of reduced sulfate deposition (from Henriksen et al. 1988).

The entire problem complex of nitrogen saturation in coniferous forest ecosystems is the subject of the European NITREX project, a series of large-scale experiments with nitrogen addition or removal to precipitation in forested stands or headwater catchments located across the gradient of present-day nitrogen deposition from the Netherlands to Norway.

6.2 Nitrate and climate change

The process of acidification and recovery occurs at time scales of decades. Over this time scale in the future major changes in climate are expected. Climate change can be expected to have important interactions with acidification. Here again nitrogen is in focus.

Changes in global atmospheric composition and the subsequent changes to global climate will have profound effects on the natural environment. For the temperate and boreal forest regions the prognosis is substantial increase in temperature. One of the major effects of higher temperature may be an increased mineralization of organic material stored in soils. Mineralization releases nutrients such as nitrogen and phosphorus which can be leached to streams, lakes and the marine environment. If nitrogen is released as NO_3^- then soil and water acidification will result; both nitrogen and phosphorus can lead to increased eutrophication of the freshwater and marine environments (Figure 13).

A simple calculation suggests a large potential for increased nitrogen leaching due to increased temperature. Organic soils typical of high latitude regions contain 3-7 ton N/ha. Acid deposition in southern Scandinavia amounts to about 20 kg N/ha/yr of which at present generally less than 10-20% leaves in runoff. If climate warming leads to mineralization of the stored pool of nitrogen in soils, then NO_3^- release to surface waters can contribute substantially to nitrogen loadings. Two tons of N is equivalent to 100 years of present-day atmospheric deposition of N in these regions.
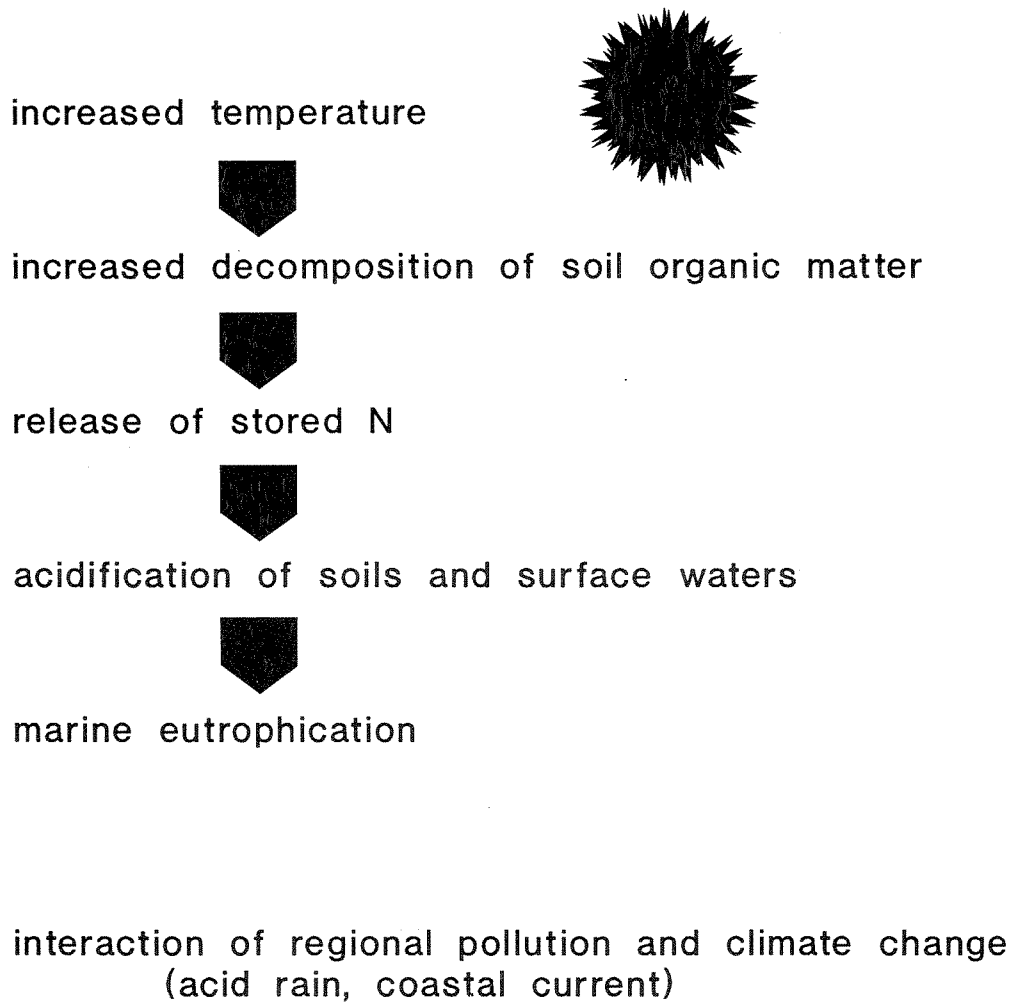
Possible effects of climate change

increased temperature

increased decomposition of soil organic matter

release of stored N

acidification of soils and surface waters

marine eutrophication

interaction of regional pollution and climate change
(acid rain, coastal current)

Figure 13. Possible effect of climate change on nitrogen flux from terrestrial ecosystems.

Kristiansandsfjord

annual N-loading

today 2100 ton
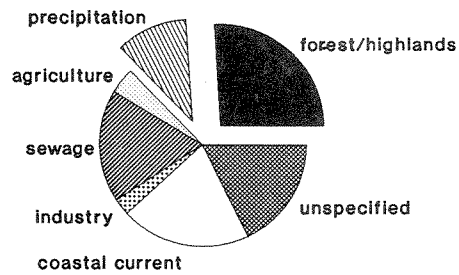
climate effect

Year 2030

N
-saturation

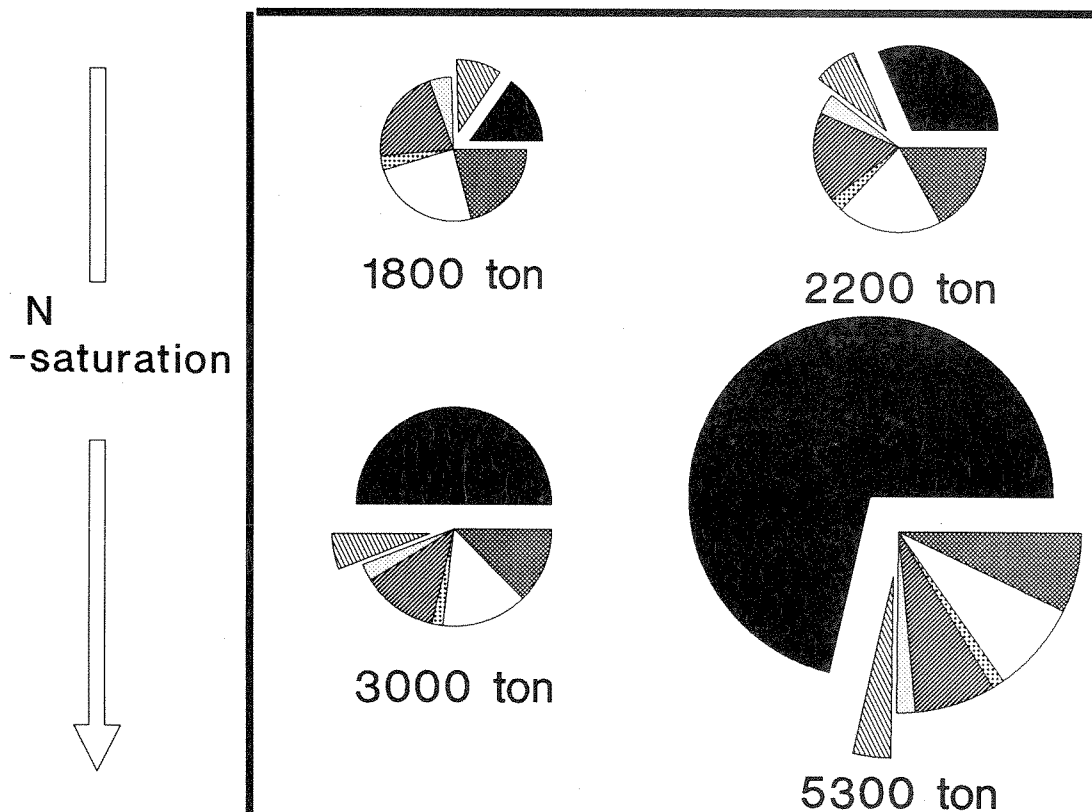1800 ton

2200 ton

3000 ton

5300 ton

Figure 14. Estimated annual loading of nitrogen to Kristiansandsfjord, southernmost Norway. About 40% of present-day loading comes from precipitation directly on lakes, rivers and the fjord itself, and from runoff from undisturbed highlands and forests in the terrestrial catchment. Future climate change and increased nitrogen saturation with subsequent loss of nitrogen to runoff might result in substantial increases in annual nitrogen loading (from Hindar et al. 1989 and Gulbrandsen et al. 1990).

Increased nitrogen flux from terrestrial ecosystems due to acid deposition and nitrogen saturation and climate change can dramatically alter the nitrogen loading to marine coastal ecosystems. In southernmost Norway nitrogen inputs from direct precipitation on open water surfaces and runoff from undisturbed forests and highlands already account for about 1/3 of the nitrogen loading to fjords (Hindar et al. 1989). The combined effects of nitrogen saturation and increased mineralization due to climate change could result in a 2-3 fold increase in nitrogen loading (Figure 14) (Gulbrandsen et al. 1990).

Large-scale whole-ecosystem experiments provide a powerful tool to study the potential effects of climate change. The whole-lake experiments in Canada and the RAIN-project in Norway demonstrate the utility of using ecosystem experiments to investigate ecosystem-scale response. The ecosystem response to the warmer, CO_2-rich environment of the future can be experimentally simulated over entire catchment ecosystems by means of large enclosures similar to those of the Norwegian RAIN-project; such an experiment has been proposed as the CLIMEX project.

7. Acknowledgements

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