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Norwegian lakes show widespread recovery from acidification; results from national surveys of lakewater chemistry 1986–1997

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Abstract

Surveys of 485 lakes in Norway conducted in 1986 and again in 1995 reveal widespread chemical recovery from acidification. Sulphate concentrations in lakes have decreased by 40% in acidified areas in southern Norway. This decrease has been compensated about 25% by decreases in concentrations of base cations and about 75% by increased Acid Neutralising Capacity (ANC). The increased ANC in turn reflects lower concentrations of acidic cations Aluminium (Al^{n+}) and Hydrogen (H^+). A sub-set of 78 of the 485 lakes sampled yearly between 1986 and 1997 shows that, at first most of the decrease in non-marine sulphate (SO_4^*) was compensated by a decrease in base cations, such that ANC remained unchanged. Then as SO_4^* continued to decrease, the concentrations of non-marine calcium and magnesium ($(Ca+Mg)^*$) levelled out. Consequently, ANC increased, and H^+ and Al^{n+} started to decrease. In eastern Norway, this shift occurred in 1989–90, and came slightly later in southern and western Norway. Similar shifts in trends in about 1991–92 can also be seen in the non-acidified areas in central and northern Norway. This shift in trends is not as pronounced in western Norway, perhaps because of the confounding influence of sea-salt episodes on water chemistry. This is the first documented national-scale recovery from acidification due to reduced acid deposition. Future climate warming and potentially increased N-leaching can counteract the positive trends in recovery from acidification.

Introduction

Atmospheric deposition of sulphur in Norway decreased by about 40% from 1980 to 1995 as a result of major reductions in emissions of SO_2 in Europe (Tørseth and Semb, 1995). These reductions come in part from international agreements obtained through the UN/ECE protocols of 1986 (30% club) (UN/ECE, 1985) and 1994 (Oslo protocol) (UN/ECE, 1994). Deposition of acidifying compounds in Norway reached a peak in the late 1970s following decades of increasing emissions of sulphur (S) and nitrogen (N) gases in Europe. Acidification of freshwaters with widespread damage to fish populations occurred over large acid-sensitive regions of southern Scandinavia (Dickson, 1978; Overrein *et al.*, 1980). Beginning in the early 1980s, however, deposition of S decreased in response to substantial reductions in S emissions in Europe (Fig. 1). The largest decreases occurred in southernmost Norway, which receives the highest absolute levels. The annual mean concentrations of SO_4^{2-} in precipitation have generally decreased from 1980 to 1995 by 30–45% in southern and 45–55% in northern Norway (Tørseth and Semb, 1995). The annual mean concentrations of particulate SO_4 and gaseous SO_2 (and hence dry deposition of S) have

by 50–65% in southern and 65–90% in central and northern Norway. This pattern fits well with the reported reductions in emissions of SO_2 in Europe. No significant trends can be seen for acidifying N components in air and precipitation (Tørseth and Semb, 1995) (Fig. 1).

Sulphate is the 'driving force' in the acidification process and it is therefore of major interest to study the effect of the reduced deposition of S on water quality. Here, the changes in water chemistry from 1986 to 1995 are reported for 485 Norwegian lakes, in relation to changes in acidic deposition loading and the timing of the changes within this period. The work is based on data from national lake surveys in 1986 (Henriksen *et al.*, 1988) and 1995 (Skjelkvåle *et al.*, 1997) and yearly measurements from 78 lakes in the period 1986–1997 as part of the Norwegian acid deposition monitoring programme (Johannessen, 1995; Skjelkvåle and Henriksen, 1995).

Regional lake surveys

In the autumn of 1986, 1000 headwater lakes were sampled in the '1000 Lake Survey' (Henriksen *et al.*, 1988). The lakes were selected from areas underlain by bedrock

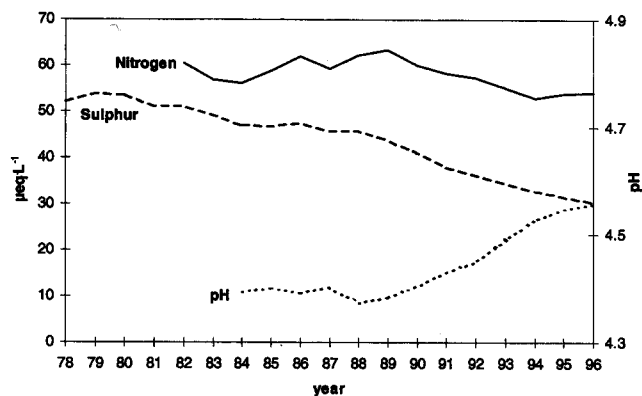


Fig. 1. 5-year moving average of S (as SO_4^{2-}) and N (sum of NH_4^+ and NO_3^-) concentrations and pH in deposition at 7 stations in southern Norway from 1974–1996 (data from Tørseth and Mano, 1996). Data are annual volume-weighted means of precipitation samples collected daily or weekly at 7 stations of the Norwegian monitoring network (Johannessen, 1985). Year denotes the last year of the 5-year period.

expected to yield runoff waters with low buffer capacities, with special emphasis on areas such as southernmost Norway known to be affected by acidification. The lakes are located in headwater areas with no direct human influence. 85% of the lakes sampled are between 0.2–2 km² in area.

In 1995, Norway conducted a new regional lake survey with two sets of lake populations: one was a resurvey of 485 lakes from the '1000 Lake Survey' and a second was based on a stratified random selection of all lakes in Norway ('statistical selection') (Skjelkvåle *et al.*, 1997) (Fig. 2). The surveys of both lake groups was a part of the Northern European Lake Survey 1995, in which the selection of lakes, sampling and analytical methods were harmonised across 6 countries (Henriksen *et al.*, 1998). In Norway, many lakes have been limed since 1986, and these were excluded. Also excluded were hydroelectric power reservoirs with water-level regulation >5 m, lakes with maximum depth <1 m, and lakes with catchment-to-lake area ratio >100:1.

The statistically-selected lakes give information on lake-water chemistry in lake populations in Norway differing in lake size and geographical region (Skjelkvåle *et al.*, 1997; Henriksen *et al.*, 1998), while the 485 lakes with data from 1986 and 1995 are used to document changes in lake acidification at the national scale. 78 of these 485 lakes from the lake survey have been sampled annually from 1986 to 1997 (Johannessen, 1995; Skjelkvåle and Henriksen, 1995).

Norway can be divided into 5 physiographic regions (Fig. 2, Table 1). These differ in continentality, predominant weather pattern, topography and dominant vegetation type. Eastern Norway is dominated by forests and receives moderate acid loading, southern Norway has high acid loading, western Norway receives high precipitation amounts, and central and northern Norway are little

affected by acid deposition (except for areas bordering Russia in easternmost parts of Finnmark county in northern Norway).

Methods

The sampling methodology and analytical methods were the same from 1986 to 1997. Water samples were collected at the outlet of the lakes after the onset of the autumn circulation period. Samples were analysed at the Norwegian Institute of Water Research (NIVA) within 2–3 days of collection. pH was analysed potentiometrically, base cations (Ca^{2+} , Mg^{2+} , Na^+ , K^+) by ICP, Cl and SO_4 by ion chromatography (IC), NO_3 by automated colorimetry (AA), Al species by automated colorimetry and Total Organic Carbon (TOC) by oxidation to CO_2 and detection by infrared. Alkalinity was measured by potentiometric titration to fixed endpoint pH 4.5.

Non-marine fractions of SO_4 and base cations are calculated on the assumptions that all Cl is of marine origin (cyclic seasalts) and is accompanied by other ions in the

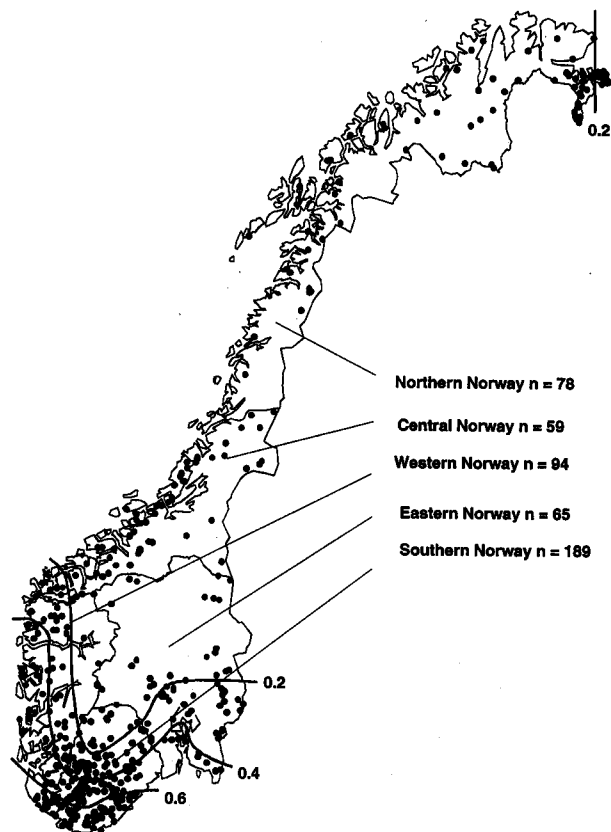


Fig. 2. Geographical distribution of the 485 lakes surveyed both in 1986 and in 1995. For treatment of the data, Norway is divided into 5 physiographic regions. The isolines show total wet S-deposition ($g S m^{-2}$) for the year 1996 (data from Tørseth and Mano, 1996).

Table 1. Number of lakes in each region of Norway in the two datasets used.

	Regional lake surveys 86-95	Yearly monitoring lakes 86-87
Eastern Norway	65	18
Southern Norway	189	22
Western Norway	94	14
Central Norway	59	8
Northern Norway	78	16
Total	485	78

same proportions as in seawater. The non-marine fractions are denoted by an asterisk. Concentrations of organic anions (org⁻) are calculated from TOC using an empirical relationship between TOC and the anion deficit (from the ionic balance) (Brakke *et al.*, 1987).

$$\text{Org}^- = \text{TOC} (4.7 - 6.87 \exp^{-0.322 \text{ TOC}})$$

where TOC is in mg C l⁻¹, and org⁻ is in µeq l⁻¹. This empirical relationship came from an analysis of the 1986 Norwegian lake survey data.

ANC (acid neutralising capacity) is defined as equivalent sum of base cations (Ca²⁺ + Mg²⁺ + Na⁺ + K⁺) minus equivalent sum of strong acid anions (SO₄²⁻ + Cl⁻ + NO₃⁻). From the ionic balance, and neglecting ions present in minor concentrations, ANC is also equal to: (HCO₃⁻ + org⁻) - (H⁺ + Alⁿ⁺), where Alⁿ⁺ is the sum of cationic Al species.

The differences in chemistry for lake populations between years were tested statistically with one-tailed t-test.

Results

CHEMISTRY OF THE LAKES

In the 1986 lake survey, the lakes selected were predominantly headwater lakes on granitic and gneissic bedrock, without significant local pollution. Bedrock, overburden and soils in such areas are dominated by minerals resistant to chemical weathering, and small amounts of ions are released to the water. The concentrations of base cations (Ca, Mg, Na, K) and alkalinity (HCO₃⁻) are therefore low. Ca concentrations are generally <100 µeq l⁻¹, and ANC <50 µeq l⁻¹. Such lakes are acid sensitive.

S and N deposition from long-range transported air pollution is highest in southern and western Norway and decrease northwards (Fig. 2). The eastern part of Finnmark in northern Norway is also affected by S pollution from metal smelters on the Kola peninsula in Russia. In general, low pH lakes are found in southern Norway where the acid lakes have high concentrations of SO₄^{*} relative to base cations and low and negative values of ANC.

The lakes in central and northern Norway show higher pH and lower concentrations of SO₄^{*} and high and positive ANC values (Table 3).

Precipitation and runoff amounts are particularly high along the western coast of Norway and decrease eastwards. High water flux dilutes surface waters, and lakes in areas with high precipitation generally have low concentrations of ions. The geographical pattern of ion concentrations in lakes, therefore, generally reflects the precipitation pattern in Norway (Table 3).

High TOC lakes (>5 mg Cl⁻¹) are found in forested areas in eastern Norway, while western Norway with sparse forest and high water fluxes shows very low TOC values (<1 mg Cl⁻¹).

The 1995 survey included an additional 1004 lakes selected by a stratified-random sampling of the entire lake population in Norway (Skjelkvåle *et al.*, 1997). The median values for the total lake population shows that the 485 resampled lakes including the 78 yearly monitoring lakes are in general more dilute (lower concentrations of non-marine base cations), have higher concentrations of SO₄^{*}, and are more acidified (lower pH and ANC) than the total lake population in the same area (Fig. 3). The levels of organic acids (TOC) are higher in eastern regions of Norway and lower in southern and central regions of Norway relative to the total lake population in the same area.

This comparison is based on the calculated median values for the total lake population. The median values for the statistically selected lakes were calculated from weighted samples. The sampling strategy favoured lakes in acidified regions and large lakes, therefore the lakes had to be weighted relative to the total number of lakes in each size class and region, in order to estimate unbiased descriptive statistics (Henriksen *et al.*, 1998).

TRENDS IN LAKEWATER CHEMISTRY FROM 1986-1997

The lake water chemistry has changed significantly from 1986 to 1995. SO₄^{*} concentrations in lakes show a decrease ranging from 40% in southern, 25% in central, to 11% in northern Norway (Table 2). Due to the decrease in SO₄^{*}

Table 2. Median and percentage change in non-marine sulphate for the period 1986-1995. All changes are significant at the 0.005 level (one-tailed t-test).

	n	ΔSO ₄ [*] µeq l ⁻¹	% change in SO ₄ [*]
Eastern Norway	65	-21	32
Southern Norway	189	-21	40
Western Norway	94	-10	25
Central Norway	59	-5	25
Northern Norway	78	-7	11

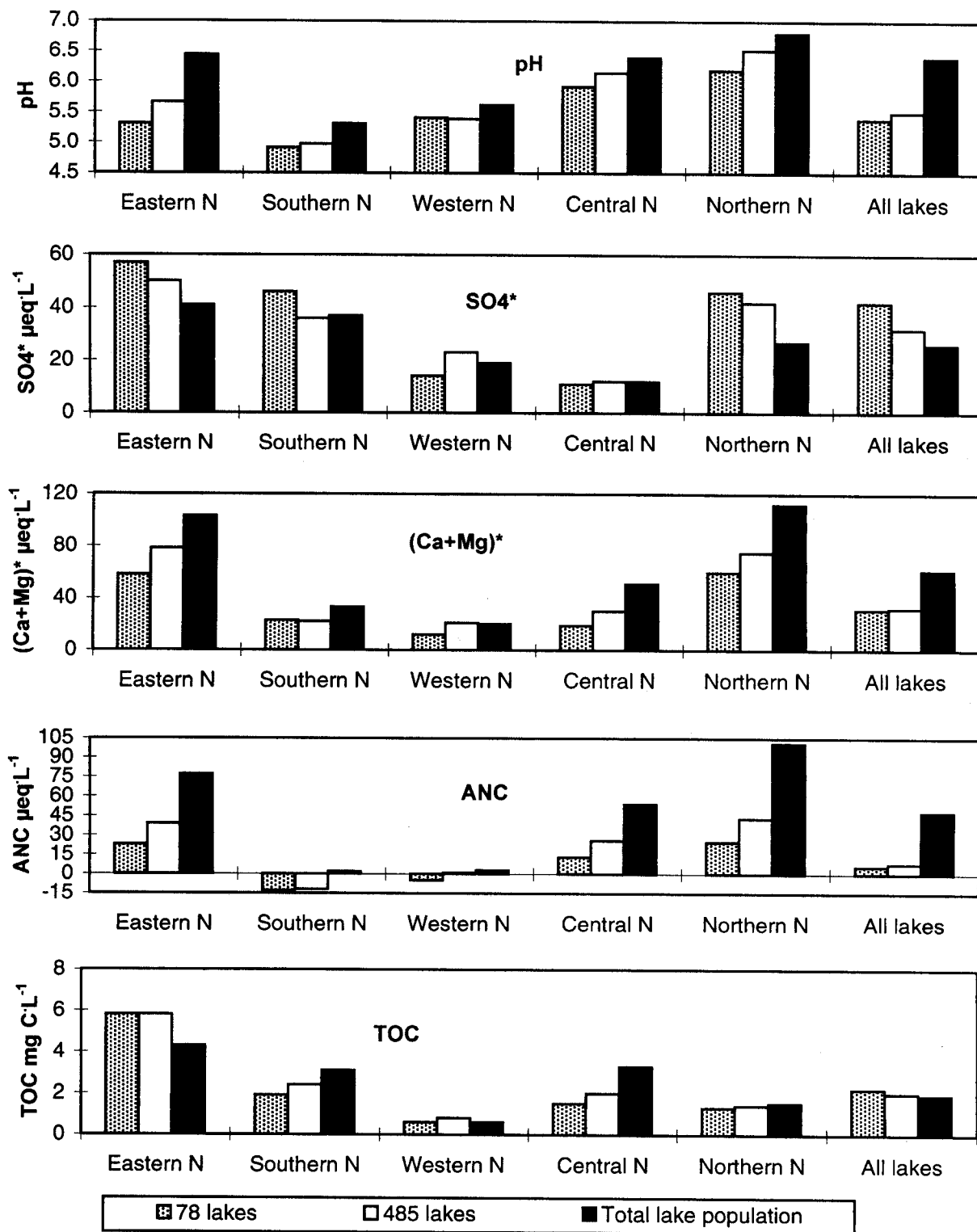


Fig. 3. Median value for 1995 for key chemical parameters for the 2 datasets used in this work (the 485 lakes sampled in both 1986 and 1995 and the 78 lakes with data from each year from 1986–1997) and the estimated median value for the total lake population from the statistically-sampled lakes (data from Skjelkvåle et al., 1997).

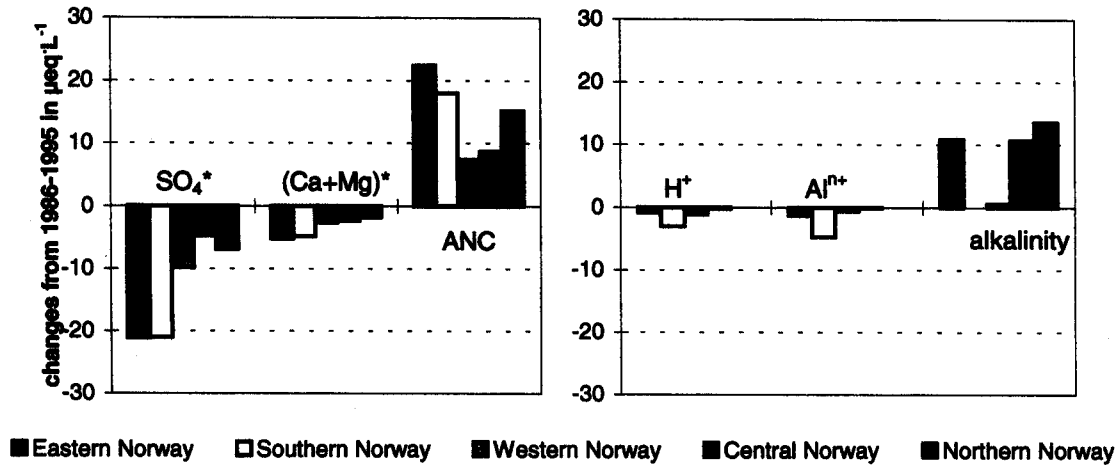


Fig. 4. Median changes in concentration from 1986 to 1995 for SO_4^* , $(Ca+Mg)^*$ and ANC (left panel) and H^+ , Al^{n+} and alkalinity (right panel) for lakes in 5 regions of Norway (Fig. 2). Median alkalinity in the southern Norway group was 0 in both 1986 and 1995.

concentrations, there has also been a significant change ($p < 0.005$) in concentrations of many of the other constituents (Table 3, Fig. 4).

These decreases in SO_4^* concentrations have been compensated about 25% by a decrease in base cations (largely Ca^{2+} and Mg^{2+}), with the remainder coming as increased ANC. This, in turn, is due to decreased concentrations of acid cations (H^+ and Al^{n+}) for lakes with $ANC < 0$, and a significant increase in alkalinity for lakes with $ANC > 0$.

Concentrations of organic anions (org^-) in lakes in eastern and southern Norway show first a decline then an increase (Fig. 4). Organic anion concentrations are not measured directly, but calculated from TOC concentrations. Levels of TOC are very low in western Norway and, thus, calculated values of org^- in this region are negligible. The variations in TOC (and thus org^-) are probably related to year-to-year climatic variations and might be due to changes in delivery of TOC from the terrestrial

Table 3. Median value ($\mu eq\ l^{-1}$ except for Labile Al, $\mu g\ l^{-1}$) for all measured and calculated variables for the 5 different regions of Norway in 1986 and 1995, and the median value of change between these two years. Statistically significant changes (one-tailed t-test) for $p < 0.005$ are denoted by *.

	Eastern Norway			Southern Norway			Western Norway			Central Norway			Northern Norway		
	86	95	Δ	86	95	Δ	86	95	Δ	86	95	Δ	86	95	Δ
H^+	4	2	-2*	15	11	-4*	6	4	-2*	1	1	0.5*	0	0	0
pH	5.44	5.66		4.82	4.97		5.24	5.39		6.10	6.15		6.46	6.52	
Ca^{2+}	60	60	-2	25	21	-3	23	22	-1	28	28	-2	57	58	0
Mg^{2+}	26	23	-2	14	14	0	20	22	1	25	25	-1	41	38	-1
Na^+	34	37	3*	36	44	8*	67	79	11*	87	92	3	80	84	5
K^+	5	5	0	3	2	-1	3	4	0	5	4	0	7	6	0
Labile Al ($mg\ l^{-1}$)	42	28	-11*	113	64	-42*	22	15	-5*	4	1	-2*	0	0	0
SO_4^{2-}	75	54	-21*	65	42	-19*	40	30	-8*	31	25	-6*	63	52	-8*
Cl^-	25	25	0	40	45	8*	73	90	11*	102	93	0	86	88	0
Alkalinity	11	16	11*	0	0	0	0	2	1	9	21	11*	28	41	14*
NO_3^-	3	2	0	8	8	0	7	7	1	1	1	0	0	0	0
$A^-(org)$	17	21	0	2	4	1	0	0	0	3	2	0	0	0	0
$(Ca+Mg)^*$	79	78	-5*	29	22	-5*	25	21	-3*	32	30	-2*	73	75	-2
Na^*	13	16	1*	2	5	2*	1	2	2*	3	10	7*	3	13	6*
SO_4^*	73	50	-21*	60	36	-21*	30	23	-10*	16	12	-5*	47	42	-7*
SSA	100	83	-19*	112	93	-13*	114	127	3	121	119	-6*	155	139	-9*
SBC	130	134	0	78	84	4*	109	125	13*	144	151	1	205	194	3
ANC	19	39	22*	-28	-12	18*	-8	0	7*	17	26	9*	25	43	15*

catchments and changes in amount of oxidation of TOC in the lakes themselves. There is no evidence that TOC has changed in response to changes in loading of strong acids.

There have been no significant changes in concentrations of NO_3^- from 1986 to 1995 in any part of the country. Chloride was significantly higher in 1995 relative to 1986 in southern and western Norway probably due to storms containing high seasalt concentrations in the period prior to sampling.

The timing of the responses differed between regions in Norway over the 10-year period is shown by results from the sub-set of 78 of the 485 lakes sampled yearly between 1986 and 1997. The dynamics in the response was the same between regions (Fig. 5). At first, the decrease in SO_4^* was accompanied by an equivalent decrease in base cations such that ANC remained essentially unchanged. Then, as SO_4^* continued to decrease, the concentrations of $(\text{Ca}+\text{Mg})^*$ levelled out and, consequently, ANC

increased. The increase in ANC was due to decreased concentrations of H^+ and Al^{3+} and increased alkalinity (mostly bicarbonate). In eastern Norway, this shift occurred in 1989–90, and came slightly later in southern and western Norway. Similar shifts in trends at about 1991–92 can also be seen in the non-acidified areas in central and northern Norway. This shift in trends is not as pronounced in western Norway, perhaps because of the confounding influence of sea-salt episodes on water chemistry (Hindar *et al.*, 1995)

Recovery from acidification seen at the national scale in Norway is also evident at individual sites and groups of lakes elsewhere in Europe as well as in North America. The international UN/ECE program on acidification of rivers and lakes (ICP-Waters), for example, reports decreased SO_4 concentrations with increased ANC at many lakes and streams in North America and Europe (Newell and Skjelkvåle, 1997; Lükewille *et al.*, 1997). And

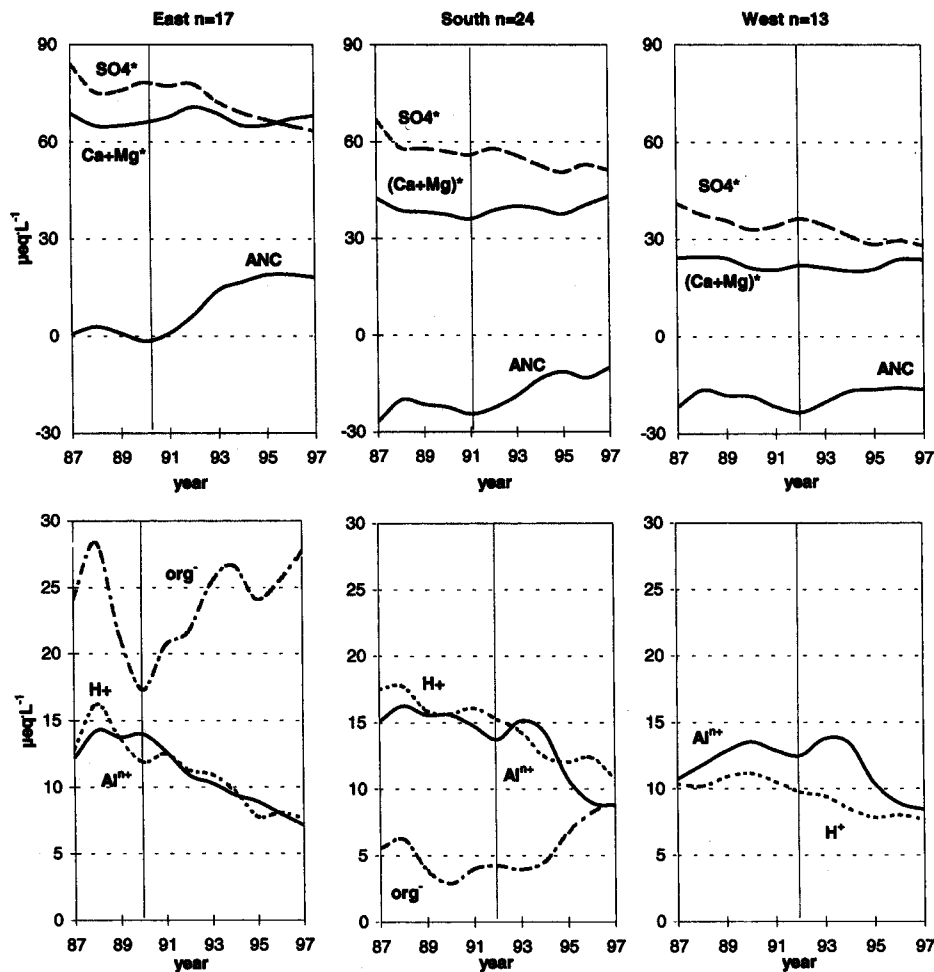


Fig. 5. 2-year moving average for SO_4^* , $(\text{Ca}+\text{Mg})^*$ and ANC (top panels) and Al^{3+} , H^+ and org^- (organic anions) (bottom panels) from 1986 to 1997 in the sub-set of 78 lakes sampled each autumn. Year denotes the second year of the 2-year period. Lakes in the three acidified regions of Norway are shown. Org^- concentrations in the lakes in western Norway are negligible. The dotted vertical line indicates the time of the major change in ANC.

recovery from acidification has been reported at lakes in the Czech Republic (Vesely *et al.*, 1998; Kopacek *et al.*, 1998) Galloway, Scotland (Wright *et al.*, 1994) and The Netherlands (van Dam, 1996).

In Norway the improved water quality with regard to higher ANC and pH together with decreased concentrations of Al^{n+} has resulted in improved conditions for fish and invertebrates in lakes and rivers (Hesthagen *et al.*, 1995; Raddum and Fjellheim, 1995).

Future perspectives

In 1985, the critical load of S and N acidity was exceeded in 37% of the area in Norway (Henriksen, 1998); by 1990 only 25% was exceeded. When the Second Sulphur Protocol (UN/ECE, 1994) is fully implemented in 2010, and S deposition in Norway reduced a further 20%, the critical load for S and N deposition will still be exceeded in 11% of Norway (Henriksen, 1998). N-deposition will then account for a relatively large part of the remaining exceedence and further improvements will necessitate reduction in N-emissions. The outcome of the ongoing negotiations for the second NO_x -protocol will be crucial for future reductions in acidification of lakes.

Global change may affect acidification of lakes in the future. There are clear interactions between climatic change and acid deposition (Wright and Schindler, 1995; Sommaruga-Wögrath *et al.*, 1997). Climatic warming and drought might result in oxidation, mineralisation and release of S and N stored in catchment soils with the subsequent acidification of runoff. The large-scale CLIMEX experiment in southern Norway in which whole catchments are subjected to increased CO_2 and temperature showed increased release of nitrate in runoff (Lükewille and Wright, 1997; Wright, 1998). There is also a potential danger of future increased N leaching due to N saturation at present-day N deposition (Stoddard, 1994; Dise and Wright, 1995; Hessen *et al.*, 1997). The magnitude, geographical extent and timing of the effects of climate change and potential N saturation are largely unknown. Future climate warming and increased N leaching can, therefore, potentially offset the positive trends in recovery from acidification obtained from international negotiations of S emission reductions.

Major and widespread improvement of water quality in acidified lakes in southern Norway is a success story, showing that international negotiations to reduce transboundary air pollution can work. Nevertheless, there is still a long way to go, and the threat of N leaching due to N saturation and climate change looms on the horizon. Although the situation has improved, the critical load is still exceeded in large areas and will be for many decades to come. Thus, liming as a mitigative measure is still needed to provide suitable conditions for salmonid fish in Norwegian lakes and rivers. Systematic long-term monitoring programmes provide the essential documentation to

show that costly and extensive emission reduction measures actually have the intended effect of protecting the environment.

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