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International Cooperative Programme on Assessment and
Monitoring of Acidification of Rivers and Lakes



ICP Waters Report 89/2007

Review of the Gothenburg Protocol: Chemical and biological responses in surface waters and soils



Norwegian Institute for Water Research

Norwegian Institute for Water Research

– an institute in the Environmental Research Alliance of Norway

REPORT

Main Office

Gaustadalléen 21
N-0349 Oslo, Norway
Phone (47) 22 18 51 00
Telefax (47) 22 18 52 00
Internet: www.niva.no

Regional Office, Sørlandet

Televeien 3
N-4879 Grimstad, Norway
Phone (47) 22 18 51 00
Telefax (47) 37 04 45 13

Regional Office, Østlandet

Sandvikaveien 41
N-2312 Ottestad, Norway
Phone (47) 22 18 51 00
Telefax (47) 62 57 66 53

Regional Office, Vestlandet

P.O.Box 2026
N-5817 Bergen, Norway
Phone (47) 22 18 51 00
Telefax (47) 55 30 22 51

Akvaplan-NIVA A/S

N-9005 Tromsø, Norway
Phone (47) 77 68 52 80
Telefax (47) 77 68 05 09

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Author(s) Richard F. Wright, NIVA <i>Maximilian Posch, MNP/CCE, the Netherlands</i> <i>B. Jack Cosby, University of Virginia, USA</i> <i>Martin Forsius, SYKE, Finland</i> Brit Lisa Skjelkvåle, NIVA	Topic group Acid rain	Distribution
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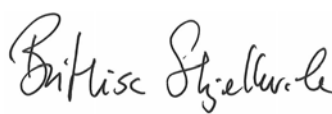
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Abstract Monitoring data from ICP Waters and ICP Integrated Monitoring as well as other sources show clear and large regional recovery in surface water chemistry in response to the large decreases in deposition of sulphur and to a lesser extent nitrogen since the mid-1980s. Waters have become less acidic and less toxic to biota. At many sites sulphate concentrations now approach the low levels expected following full implementation of the Gothenburg protocol. Nitrate, on the other hand, does not show consistent trends, and most sites are far from steady-state conditions. Results of multivariate statistical gradient analysis suggest that coniferous defoliation, discolouration and lifespan of needles in the diverse phenomena of forest damage are explained by the combined action of ozone and acidifying sulphur and nitrogen compounds in air. Dynamic models and critical load mapping indicate that a significant number of sites in several regions of Europe will continue to be acidified after 2010. Biological recovery has begun in many regions, but lags behind chemical recovery. Future climate change will affect acidification and recovery.
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Richard F. Wright
Project manager



Brit Lisa Skjelkvåle
Research manager



Jarle Nygard
Strategy Director

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CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION

Working Group on Effects

Prepared by
the ICP Waters Programme Centre at NIVA, Norway, and
the ICP Integrated Monitoring Programme Centre at SYKE, Finland

Review of the Gothenburg Protocol: Chemical and biological responses in surface waters and soils

Preface

In conjunction with the ongoing review of the Gothenburg protocol to the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP), the Working Group on Effects (WGE) requested each of the International Cooperative Programmes (ICPs) to provide short statements on three topics: (1) recent information on effects-based approaches for the protocol reviews, (2) dose-response functions and stock at risk, and (3) links between observations and critical thresholds. ICP Waters and ICP Integrated Monitoring (IM) prepared these statements jointly. This report is an expansion of these inputs to the WGE. This report has also been prepared jointly by ICP Waters and ICP IM.

Richard Wright (NIVA) was responsible for preparing the report, with inputs from the other authors. The work was financed in part by the WGE, ICP IM and ICP Waters (through funding provided by the Norwegian Pollution Control Authority SFT).

Brit Lisa Skjelkvåle
ICP Waters Programme Centre
Oslo, August 2007

Martin Forsius
ICP Integrated Monitoring Programme Centre
Helsinki, August 2007

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Summary

Monitoring data from Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) and national sources show clear reductions (60-80%) in sulphur (S) deposition in Europe since the peak years in the early 1980s. Nitrogen (N) deposition has also declined, but to a much lesser extent. Monitoring data from ICP Waters (W) and ICP Integrated Monitoring (IM) as well as other sources show clear and large regional trends in surface water chemistry in response to the large decreases in deposition of S and N. At many sites sulphate concentrations now approach new levels expected following full implementation of the Gothenburg protocol. Nitrate, on the other hand, does not show consistent trends, and most sites are far from steady-state conditions. Waters have become less acidic and less toxic to biota. Acid neutralising capacity (ANC) has increased. Biological recovery of fish and invertebrates provide new support for empirical relationships between dose (ANC) and biological response in surface waters.

In forests, gradient analysis suggests that the combined action of O₃, acidifying S and N explains 18% of conifer defoliation, 42% of discolouration, and 55% of lifespan for needles. Deposition of nitrogen is important in determining the occurrence of acidophilic lichen species.

There is generally good agreement between exceedence of critical load for acidity and ANC in surface waters. Time delays are well explained by known processes acting in catchments and waters, and can be modelled by dynamic models. A critical deposition threshold of about 8-10 kg N ha⁻¹ yr⁻¹, indicated by several previous assessments, was confirmed by the N input-output calculations with the ICP IM data.

Dynamic models indicate that a significant number of sites in several regions of Europe will continue to be acidified after 2010. Biological recovery has begun in many regions, but lags behind chemical recovery. Future climate change will affect acidification and recovery. Uncertainties remain as to how N dynamics should be treated by dynamic models and as to the role of changing climate.

1. Introduction

The 1999 Protocol to Abate Acidification, Eutrophication and Ground-level Ozone (the Gothenburg protocol) to the United Nations Economic Commission for Europe's (UNECE) Convention on Long-range Transboundary Air Pollution (LRTAP) (UNECE 1999) entered into force in May 2005 and is currently under review. As part of the review process the Working Group on Effects (WGE) through the various International Cooperative Programmes (ICPs) provide scientific assessments of the current status of effects of long-range transboundary air pollutants in Europe and North America. As part of this effort ICP Integrated Monitoring (IM) and ICP Waters (W) have joined forces to assess the current state of recovery of ecosystems from damage caused by deposition of acidifying pollutants and to evaluate the efficacy of the critical loads and target loads methods used as a basis for the Gothenburg protocol.

2. Trends in deposition of S and N in Europe

Deposition of sulphur and to a lesser extent nitrogen compounds has decreased substantially in Europe during the past 20 years, in part due to implementation of protocols of the LRTAP Convention. Results from the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) clearly show that sulphur deposition has decreased by nearly 80% from peak levels in the early 1980s (Figure 1).

Deposition of inorganic nitrogen compounds has also decreased, but to a much lesser extent than sulphur (Figure 2). While long-term measurements of S deposition show strong decreasing trends at most sites in Europe, N deposition at some sites has decreased, while at others no long-term trend is apparent (Figure 3). The sites shown here were chosen for the length of the data series to illustrate trends and year-to-year variability and are not necessarily representative of a specific country or area.

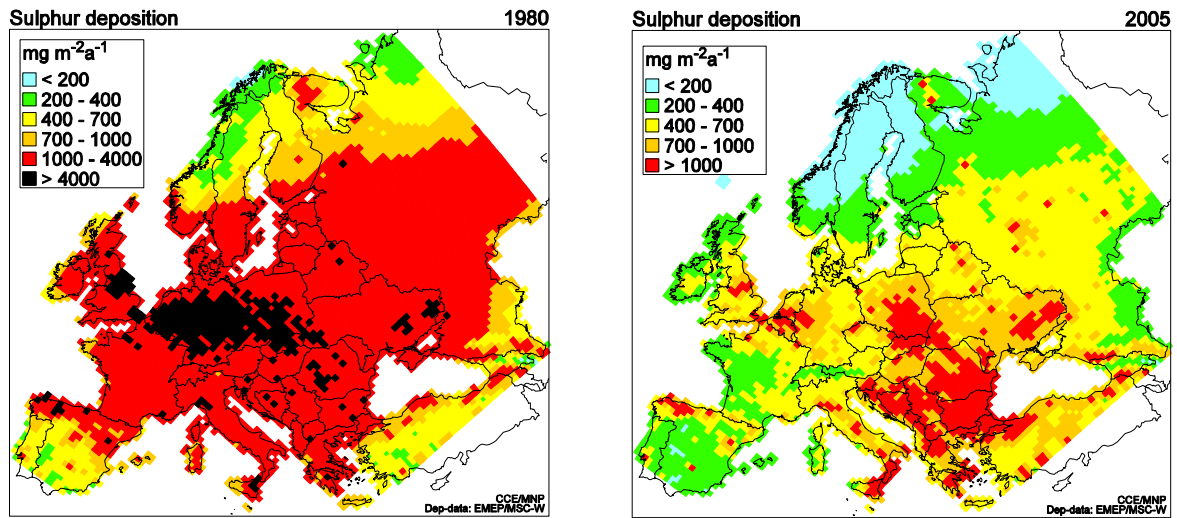


Figure 1. Modelled sulphur deposition in Europe in 1980 and 2005 as reported by EMEP.

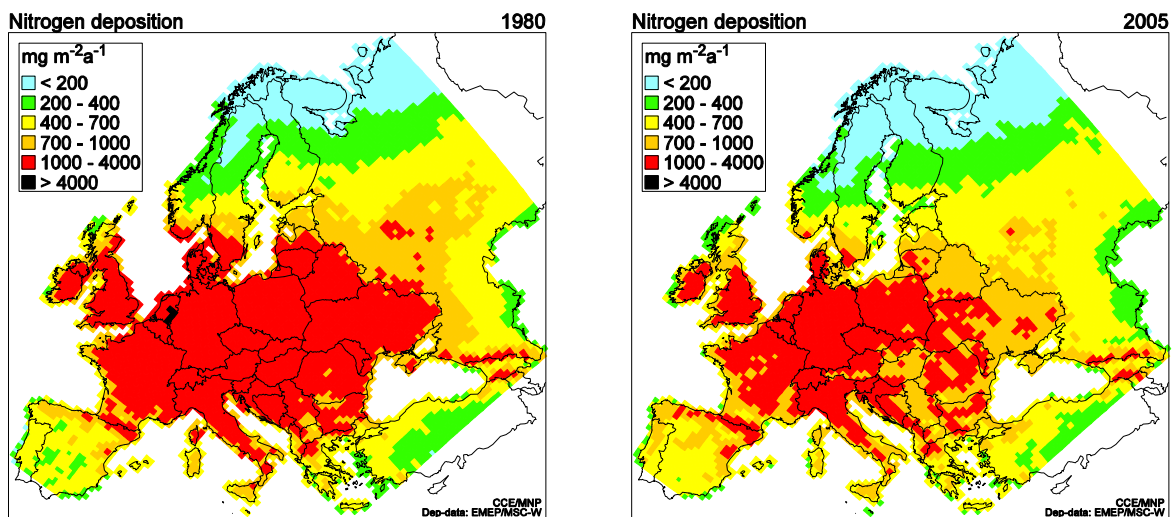


Figure 2. Modelled nitrogen deposition (sum of oxidised and reduced compounds) in Europe in 1980 and 2005 as reported by EMEP.

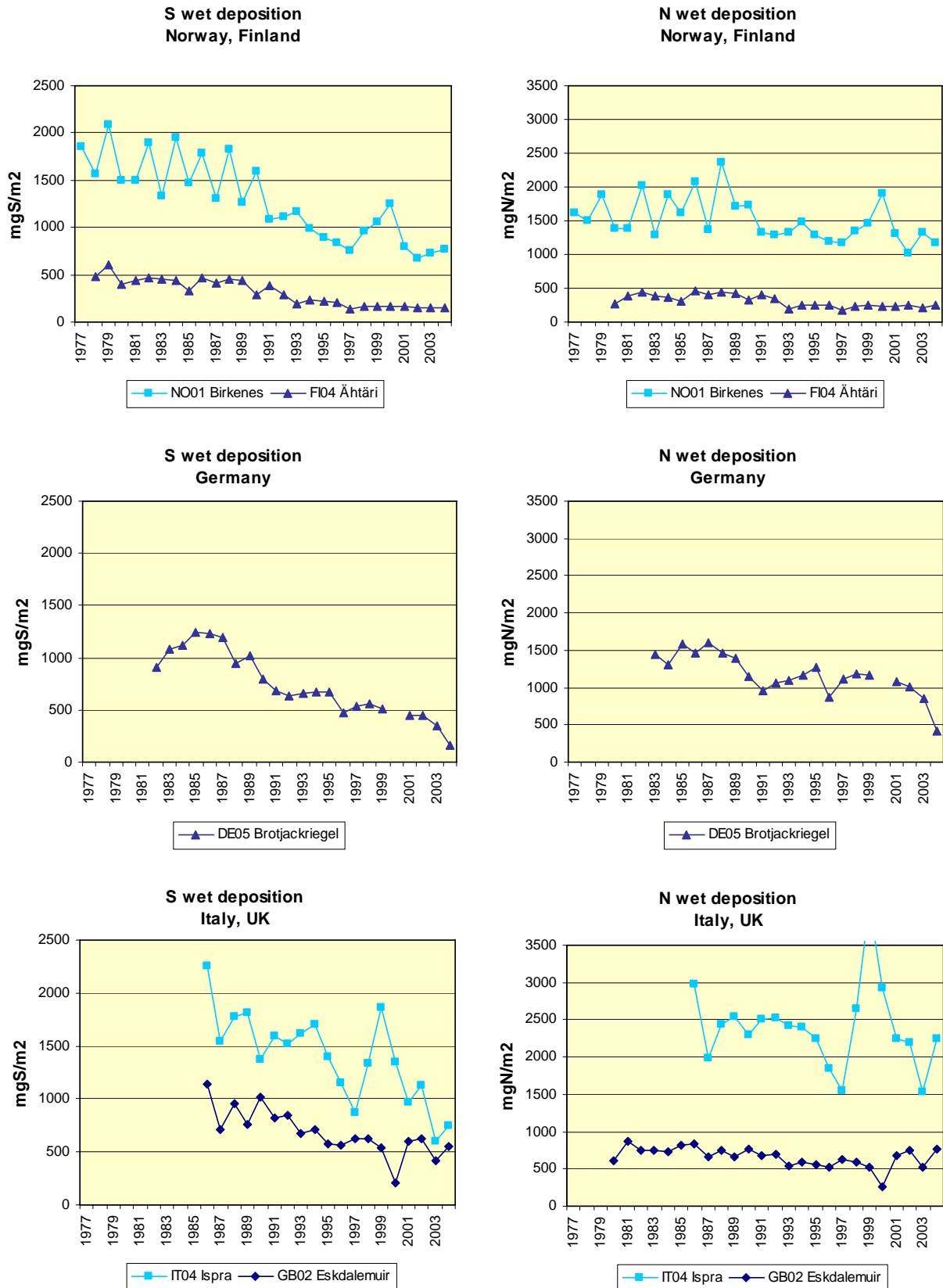


Figure 3. Measured wet deposition of sulphur (left-hand panels) and nitrogen (sum of $\text{NH}_4 + \text{NO}_3$) (right-hand panels) at several EMEP sites in northern, central, western and southern Europe (data from www.emep.int).

3. Response (recovery); dose-response relationships

3.1 Surface waters (acidification S and N)

For waters there are 2 steps in the dose-response chain: (1) deposition of acidifying S and N compounds leads to changes in water chemistry (acidification status, e.g. pH and ANC), and then (2) chemical changes lead to biological response (e.g. damage to fish populations).

3.1.1 Water chemistry

A major goal of ICP Waters is to evaluate the changes in surface water chemistry in relation to emission reductions. The strongest evidence that emissions control programs are having their intended effect comes from a consistent pattern of recovery (decreasing sulphate and increasing pH and alkalinity) across a large number of sites (Figure 4). ICP Waters has conducted statistical analyses of trends in surface water chemistry covering several time periods, including the period 1980-1995 (Stoddard et al. 1999), 1989-1998 (Skjelkvåle et al. 2000), and 1990-2001 (Skjelkvåle 2003). The most recent evaluation of trends in ICP Waters data consists of chemical records from the period 1994 to 2004 for 179 sites (73 from Europe, 106 from North America) grouped in twelve fairly homogeneous regions with regard to deposition level and acid-sensitivity (de Wit et al. 2007). The Mann-Kendall method was used for trend detection.

The most important finding in all these trend analyses is the widespread chemical recovery (increase in pH, alkalinity and/or acid neutralising capacity ANC) in streams and in lakes in most regions in Europe and North America (Table 1). ANC is defined as the equivalent sum of concentrations of base cations (Ca, Mg, Na, K) minus the equivalent sum of the strong acid anions (SO₄, Cl, NO₃). The recovery began in the 1980s, continued through the 1990s and has flattened out somewhat since 2000. The recovery is largely due to the decline in sulphate. In the most recent period (1994-2004) most sites (65%) showed a significant decreasing trend in non-marine sulphate, whereas there were no sites where non-marine sulphate had increased. All regions except two showed a significant increase in pH and/or alkalinity, and/or acid neutralizing capacity. The regions without signs of chemical recovery were Ontario and the Virginia Blue Ridge mountains in North America. Soil characteristics in the Blue Ridge Mountains make a sulphate decrease unlikely in the short term. The lack of coherent trends in Ontario is probably due to the large variability in the trends in individual sites in the region (de Wit et al. 2007). The ICP IM results are largely consistent with the ICP Waters data (Table 2).

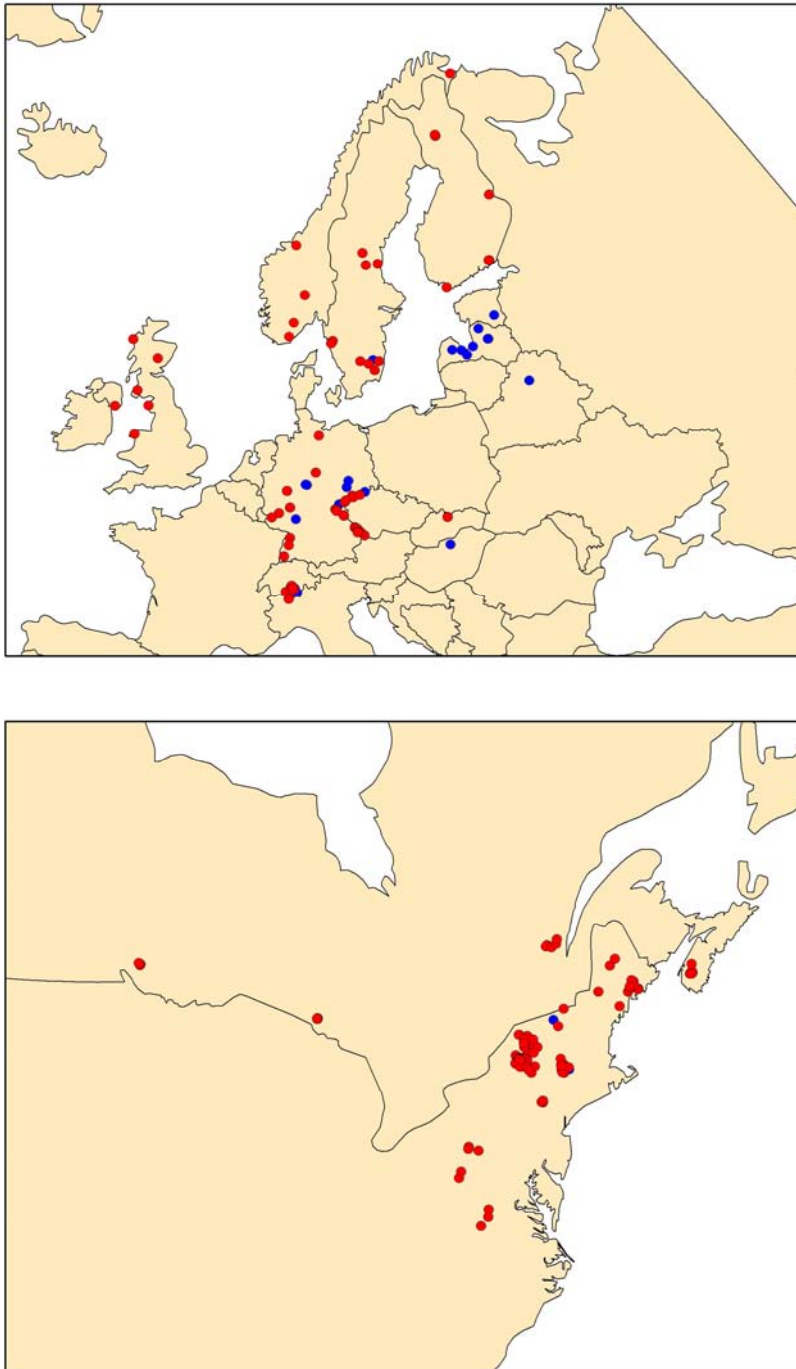


Figure 4. Map showing location of ICP Waters sites used for analysis of trends 1994-2004. Red dots are sites included in the trend analysis, while the blue dots are sites with trend analysis only for sulphate (de Wit et al. 2007).

Table 1. Results of trend analysis for 179 ICP Waters sites for the period 1994-2004. Number of sites with significantly increasing or decreasing trends for given variables. No trend at significance level $p > 0.05$ (de Wit et al. 2007).

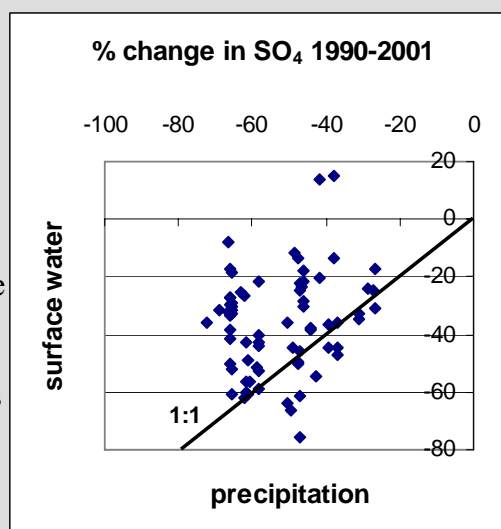
	SO ₄	NO ₃	Ca+Mg	Alkalinity	ANC	H ⁺	DOC/TOC
Europe							
Increasing	0	10	7	18	25	5	14
No trend	21	48	40	43	40	42	38
Decreasing	52	15	26	2	0	26	0
North America							
Increasing	0	3	1	32	31	3	14
No trend	41	72	59	71	44	86	92
Decreasing	61	29	19	1	1	17	0
Total no of sites	175	177	152	167	141	179	158
Total increasing	0	13	8	50	56	8	28
Total no trend	62	120	99	114	84	128	130
Total decreasing	113	44	45	3	1	43	0
% increasing trends	0	7	5	30	40	4	18
% no trends	35	68	65	68	59	72	82
% decreasing trends	65	25	30	2	1	24	0

Table 2. Number of sites showing significant ($p < 0.05$) trends for the period 1990-99 detected by the seasonal Mann-Kendall tau (SKT) in runoff water chemistry at 23 European (mostly Nordic) ICP IM sites. Asterisks denote the non-marine fraction. From Bull (2004).

	Trend	SO ₄ *	Ca*+Mg*	H+	NO ₃	ANC
Europe	Increasing	2	2	2	3	4
	No trend	10	10	11	9	6
	Decreasing	11	11	9	11	2
	Insufficient data	0	0	1	0	10

Text box 1.

Sulphate concentrations in surface waters respond rapidly to changes in S deposition. (This statement generally holds for waters in regions with young soils in areas glaciated during the Pleistocene.) As S deposition has decreased since the mid-1980s, waters have responded with lower concentrations of SO₄, and as a result acidification has decreased (increase in acid neutralising capacity ANC).



Percent change 1990-2001 in sulphate concentrations in precipitation and surface water at ICP Waters sites (aggregated by region). In most regions the decline in SO₄ concentrations in surface water is smaller than the decline in precipitation. One major factor is the time lag in response of surface water chemistry to changes in precipitation chemistry. From Skjelkvåle (2003).

interest in analysis of surface water recovery, because it is an indicator of organic (natural) acidity which may counteract the positive effect of declining sulphate. For the period 1990-2004, an increase in DOC was found at some sites in formerly glaciated parts of North America and Europe (de Wit et al. 2007).

Nitrogen concentrations in surface waters are generally much lower than expected if all N deposition left the soil in runoff water. Typically 90% of incoming N deposition is retained by the soil. Nevertheless, moderate to high level of N deposition is a necessary factor for elevated NO_3^- concentration in runoff (Figure 5).

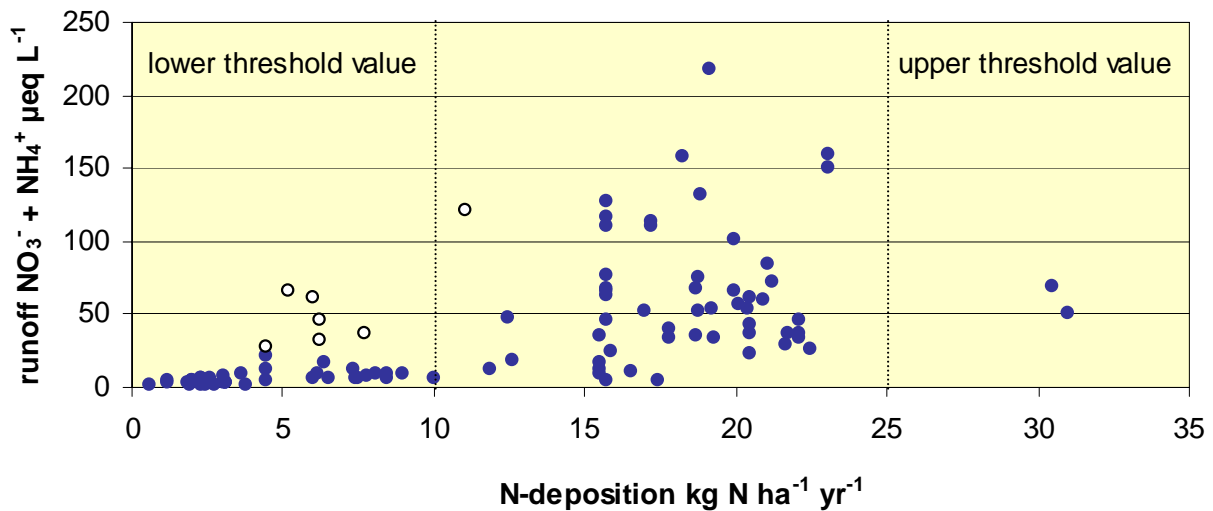


Figure 5. Nitrogen ($\text{NO}_3^- + \text{NH}_4^+$) concentration in runoff and total N deposition fluxes (wet + dry) for 99 European ICP Waters sites in 1999. Open circles: sites with possible inputs of N from agriculture. From Skjelkvåle et al. (2001)

The most recent data (1994-2004) show that the majority of the ICP Waters sites (68%) had no trend in nitrate concentrations while about (25%) showed a decreasing trend (Table 1). A few sites (7%) showed an increasing trend. The trends in nitrate have thus levelled off compared with the 1990s, when about $\frac{1}{2}$ of the sites showed negative trends in both N deposition and NO_3^- concentration in surface waters (Figure 6). The mixed tendencies in nitrate trends have been shown previously in reports of ICP Waters. The lack of a uniform trend in nitrate concentrations illustrates that nitrate leakage from catchment is affected by a variety of processes, in contrast to sulphate leakage from catchment which is largely controlled by sulphate deposition (de Wit et al. 2007). More research is needed on the processes affecting nitrogen retention and loss in catchments, especially in the light of potential future climate change.

A) 1990-2001 (N=54)

		Trend in N deposition		
		+	0	-
Trend in N runoff	+	0	7	2
	0	1	19	14
	-	0	7	4

B) 1994-2004 (N=55)

		Trend in N deposition		
		+	0	-
Trend in N runoff	+	0	8	0
	0	1	30	3
	-	1	10	2

Figure 6. Trends in NO_3^- runoff vs. trends in N deposition (concentration of total inorganic N in precipitation) at 54 ICP Waters sites in Europe during two different time periods analysed by the Mann-Kendall test. Notation: + significant increase ($p < 0.05$); 0 No significant trend; - significant decrease ($p < 0.05$) (de Wit et al. 2007).

Data from ICP IM sites have also been used in trend assessments (Kleemola and Forsius 2006), and the ICP IM results are largely consistent with the ICP Waters data (Table 2). The most recent analysis uses the non-parametric Seasonal Kendall test for the period 1993- 2003. The trends confirm the previously observed regional-scale decreasing trends of S in deposition and runoff/soil water. Acid-sensitive ICP IM sites in northern Europe show recovery from acidification. The situation regarding N was quite different with few statistically significant decreasing trends of N in deposition and both decreasing and increasing trends of nitrate in runoff/soil water. Site-specific characteristics are important in determining the response to N (and S) emission reductions (Gundersen et al. 2006). It was concluded that the N issue thus clearly requires continued attention as a European air pollution problem.

3.1.2 Aquatic biota

There are good empirical relationships between water ANC and biological response. The widest used of these is the observed population status for brown trout and ANC in Norwegian lakes (Lien et al. 1996) (Figure 7). Similar relationships exist for other biological groups, such as invertebrates in running waters (Lien et al. 1996, Raddum 1999) and diatoms in lakes (Battarbee et al. 1996).

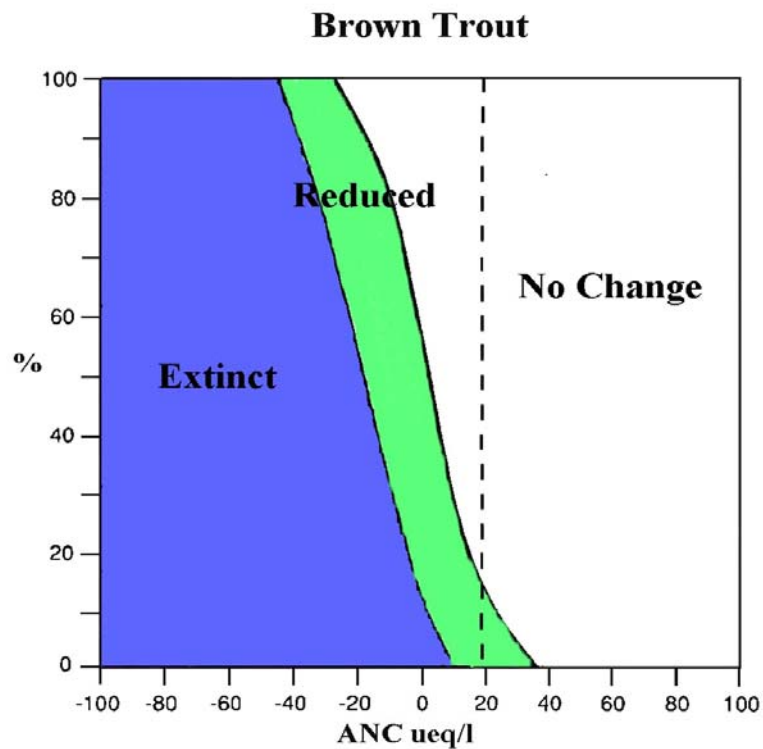


Figure 7. The observed relationship between brown trout status and surface water ANC at 827 sites in Norway (from (Lien et al. 1996).

In recent years there have been an increasing number of reports of biological recovery in acidified surface waters in response to decreasing acidity. Fish populations have begun to recover in lakes and rivers in southern Norway, for example (Figure 8) (see also Text box 2). Recovery of invertebrate fauna has also been documented (for example, at Farsund, southernmost Norway, Figure 8) (SFT 2006, Raddum et al. 2007). Recovery has also been reported from ICP Waters sites in Canada, Czech Republic, Sweden and the UK (Montieth et al. 2005, Skjelkvåle et al. 2003, deWit et al. 2007).

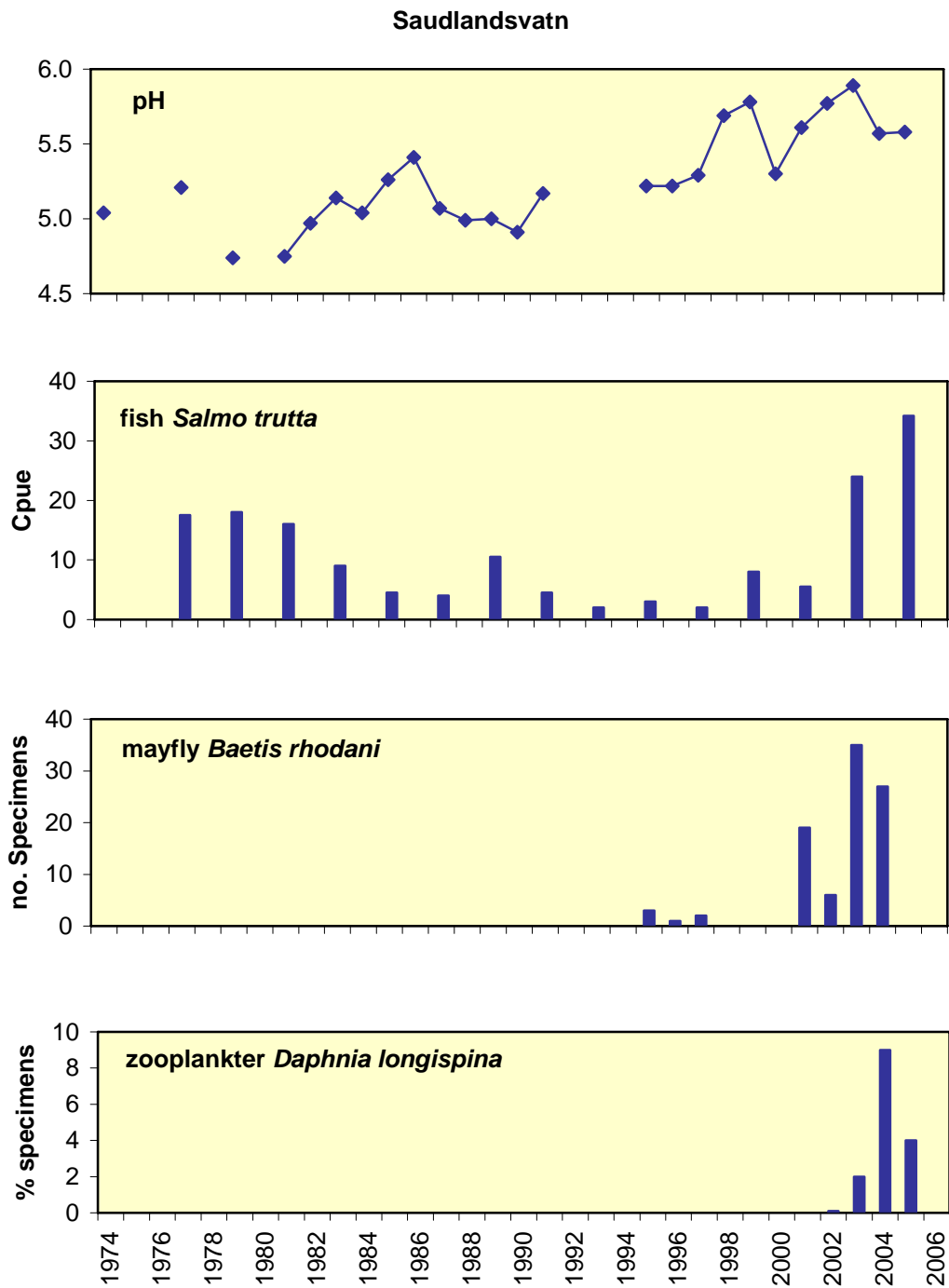


Figure 8. Recovery of pH, fish, benthic invertebrates and zooplankton fauna in the acidified lake Saudlandsvatn, southernmost Norway. Shown are data for one acid-sensitive species each of fish, benthic invertebrates and zooplankton. Data from NIVA, NINA and University of Bergen reported in SFT (2006).

Several other examples of biological recovery are found in the recent 2006 AMAP report (Skjelkvåle et al. 2006). Here a sensitive species of zooplankton has returned in an acidified

lake in northern Norway (Figure 9) (SFT 2006), and fish populations in several lakes in northern Finland also show signs of recovery.

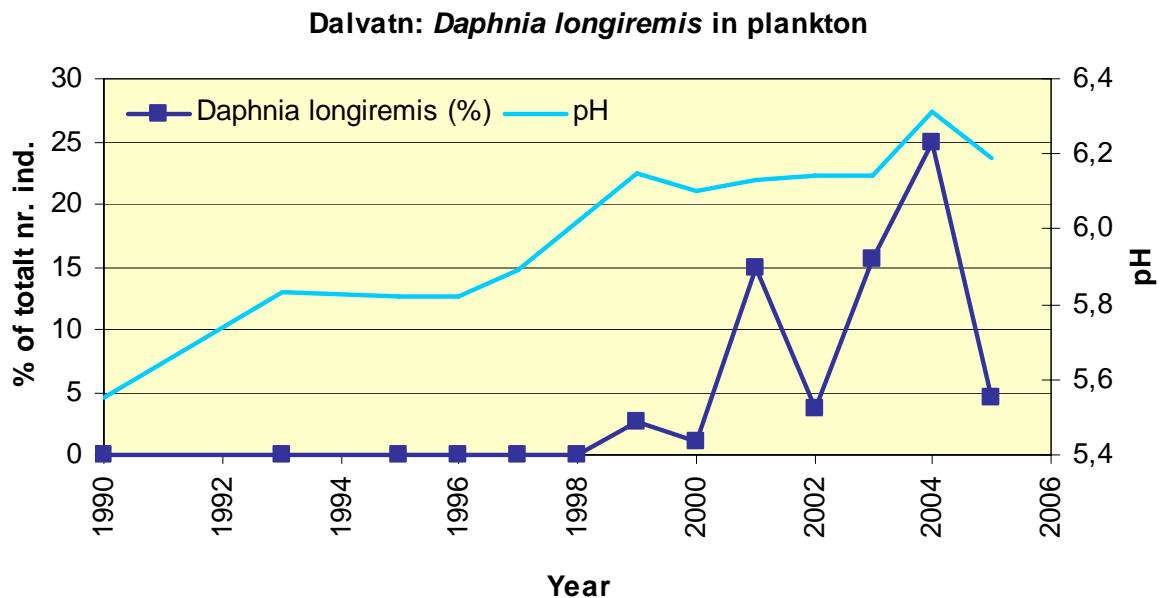


Figure 9. pH in lakewater and catch of the acid-sensitive cladoceran *Daphnia longiremis* in Lake Dalvatn (northern Norway). (SFT 2006).

3.2 Soils and vegetation (acidification S and N)

For forest soils the common chain of dose-response is (1) S+N deposition causes changes in soil acid base status (e.g. BC/Al ratio, %base saturation, pH), and then (2) these chemical changes cause biological response (e.g. crown condition, diversity of ground vegetation).

A report summarising available information from the ICP Forests and ICP IM programmes on cause-effect relationships of forest ecosystems has been prepared (de Vries et al. 2002). The results were also officially reported to the Working Group on Effects in 2002 (EB.AIR/WG.1/2002/15).

Ion mass and proton budgets have proved to be useful for evaluating the importance of various biogeochemical processes that regulate the buffering properties in ecosystems. Long-term monitoring of mass balances and ion ratios in catchments/plots can also serve as an early warning system to identify the ecological effects of different anthropogenic pollutants, and to verify the effects of emission reductions.

Proton budget calculations showed that there was a large difference between the ICP IM sites regarding the relative importance of the various processes involved in the transfer of acidity (Forsius et al. 2005). These differences reflected both the gradients in deposition inputs and the differences in site characteristics. The proton budget calculations indicated a clear relationship between the net acidifying effect of nitrogen processes and the amount of N

deposition. When the deposition increased also N processes became increasingly important as net sources of acidity. Sulphur budgets calculations indicated a net release of S from many ICP IM sites, indicating that the soils are releasing previously accumulated S. Similar results have been obtained in other recent European plot and catchment studies.

de Zwart (1998) carried out an exploratory multivariate statistical gradient analysis of possible causes underlying the aspect of forest damage at ICP IM sites. These results suggested that coniferous defoliation, discolouration and lifespan of needles in the diverse phenomena of forest damage are for respectively 18%, 42% and 55% explained by the combined action of ozone and acidifying sulphur and nitrogen compounds in air.

The epiphytic lichen flora of 25 European ICP IM monitoring sites, all situated in areas remote from local air pollution sources, was statistically related to measured levels of SO₂ in air, NH₄, NO₃ and SO₄ in precipitation, annual bulk precipitation, and annual average temperature (de Zwart et al. 2003, van Herk et al. 2003). It was concluded that long distance transport of nitrogen air pollution is important in determining the occurrence of acidophilic lichen species, and constitutes a threat to natural populations that has been underestimated so far.

Dynamic modelling of the response of S+N deposition impacts on soil (and surface water) chemistry has been carried out at ICP IM and ICP Waters sites. These results allow the assessment of past and future trends of soil chemical conditions (see section 4).

3.3 Soils and vegetation (eutrophication N)

The negative effect of N deposition for terrestrial biodiversity has received increasing attention during the last years (see review by Bobbink et al. 2003). This topic has been studied at the national level at several ICP IM sites (Ferretti et al. 2006, Dirnböck et al. 2007). Dirnböck et al. (2007) concluded that continuous eutrophication takes place in large parts of the Northern Limestone Alps in Austria where airborne pollution levels are elevated. If N emission is not abated efficiently, there is a risk of biodiversity loss and effects on ecosystem function in the future in this region.

4. Critical loads and exceedences

The critical load is the maximum amount of a pollutant that can be deposited on an ecosystem without adverse effects. The concept thus relates a chemical pollutant to a biological impact. In the case of acid deposition the critical load is the maximum deposition of acidity that can be deposited without adverse effect on the ecosystem. Application of critical loads involves identification of key organism (or organisms) to be protected, a “critical limit” for the concentration of, for example, ANC (termed ANC_{limit}), and a model to relate deposition rate to the concentration of ANC.

4.1 Waters

There is generally good agreement between exceedence of critical load for acidity and ANC in surface waters (Figure 10). Exceptions are those sites at which there are significant time delays between changes in S+N deposition and response in water chemistry. These time delays are well explained by known processes acting in catchments and waters, and can be modelled by dynamic models.

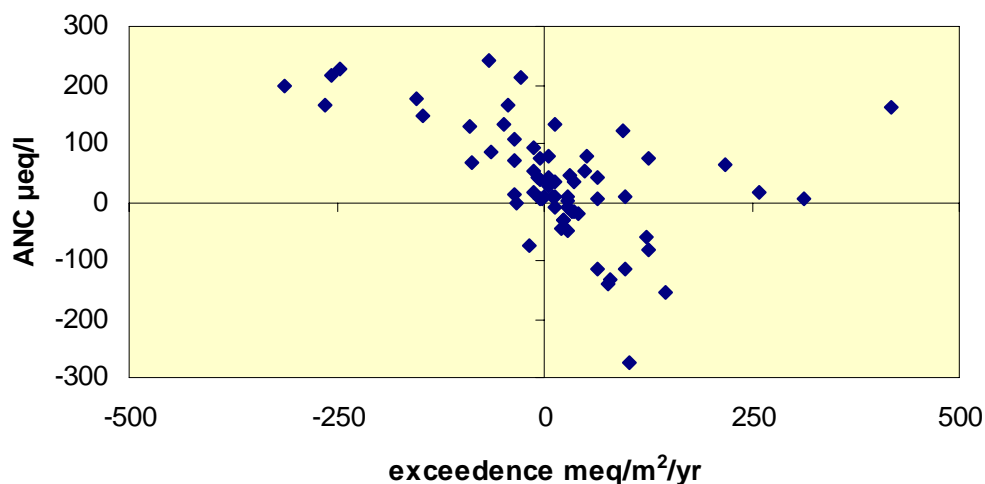


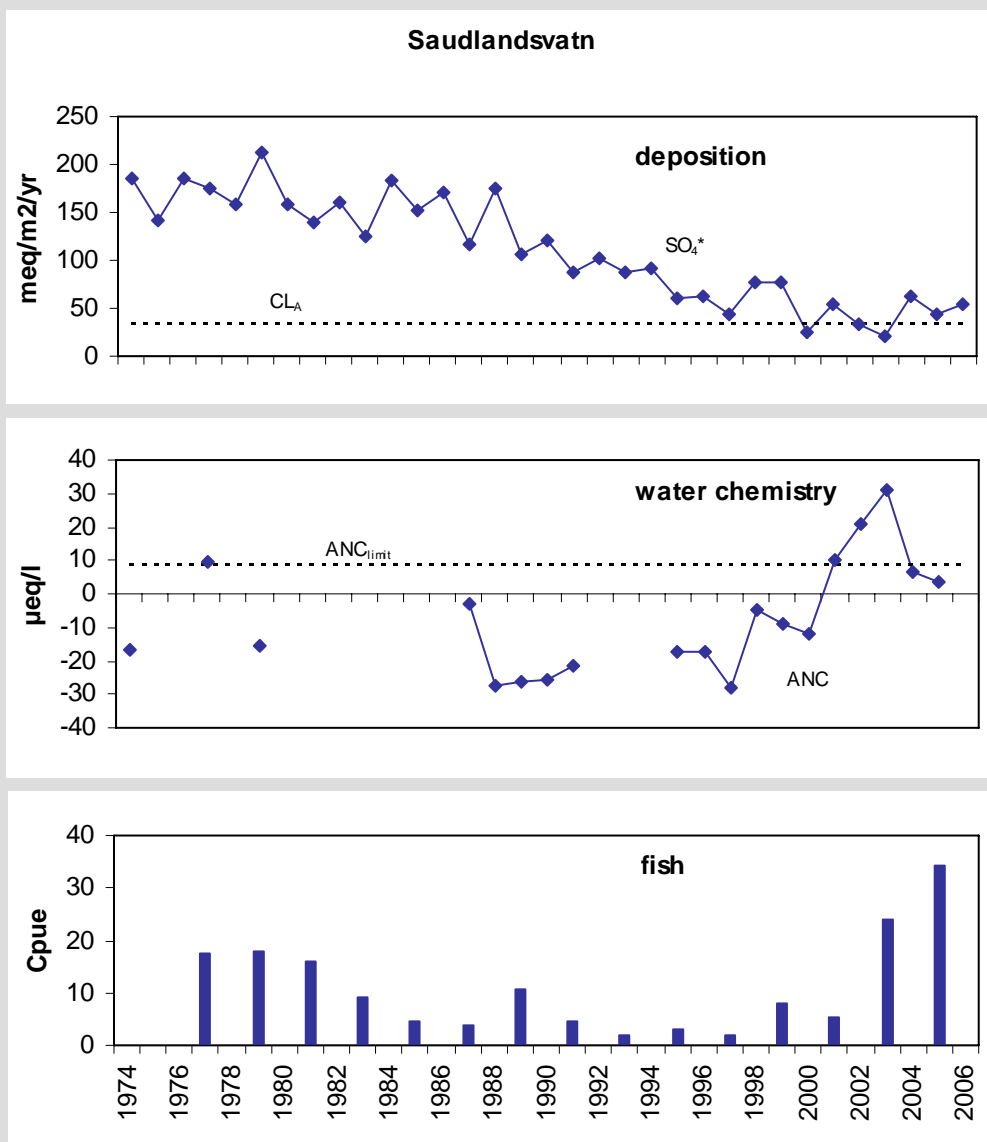
Figure 10. Exceedence (S deposition) of critical load of acidity and ANC in 1990 at ICP Waters sites in Europe. Sites in the lower right quadrant are exceeded and have ANC below ANC_{limit} . Sites in the upper left quadrant are not exceeded and ANC is above ANC_{limit} . Sites in both these quadrants are positioned as expected with respect to critical load, exceedence and response of water chemistry. Sites in the upper right quadrant are exceeded but have ANC above ANC_{limit} . These sites may show delayed response to S deposition. Data from Henriksen and Posch (1998).

Text box 2:

Critical limit: links between S deposition, surface water chemistry, and fish in Lake Saudlandsvatn, southernmost Norway.

Saudlandsvatn, southernmost Norway, is an example of a lake that was acidified and had damage to the fish population in the 1980's, but then has shown significant chemical and biological recovery in response to decreased S deposition since 1990. Since about 2000 S deposition has fallen below the critical limit of acidity (CLA) (upper panel), water chemistry has improved with ANC above ANClimit (middle panel), and fish population (Cpue = catch per unit effort) has begun to show improved recruitment (lower panel).

Data from T. Hesthagen, NINA (fish), NIVA (water chemistry), NILU (deposition)



4.2 Soils/catchments

A critical deposition threshold of about 8-10 kg N ha⁻¹ a⁻¹, indicated by several previous assessments, was confirmed by the N input-output calculations with the ICP IM data (Forsius et al. 2005), as well as using a larger European database including these sites (MacDonald et al. 2002) (see above). The important parameters that determine N leaching (and thus N retention) are: N deposition, the organic layer carbon to nitrogen ratio (C/N ratio) and annual temperature. At low C/N ratios (below 23) N input determines N leaching. At higher C/N ratios both N input and temperature are important. Adding more sites did not change these relationships and they were robust in validation tests (Gundersen et al. 2006).

Model testing/development work indicated good consistency between calculations of critical loads of incoming acidity as estimated by dynamic modelling (MAGIC) and the FAB model (used for reporting surface water critical loads by some countries). Uncertainties over how N dynamics should be treated by dynamic models and uncertainties due to changing climate were also assessed (Hutchins and Jenkins 2006).

The critical load for acidity will still be exceeded in many areas and in many types of ecosystems after full implementation of the Gothenburg protocol (Figure 11) (Hettelingh et al. 2007). The European map for surface waters shows information for only the four countries that report data to the Coordination Centre for Effects (CCE). Several ICP Waters and Integrated Monitoring sites in other areas of Europe (and North America) will also continue to be acidified and critical loads for acidity exceeded in the future.

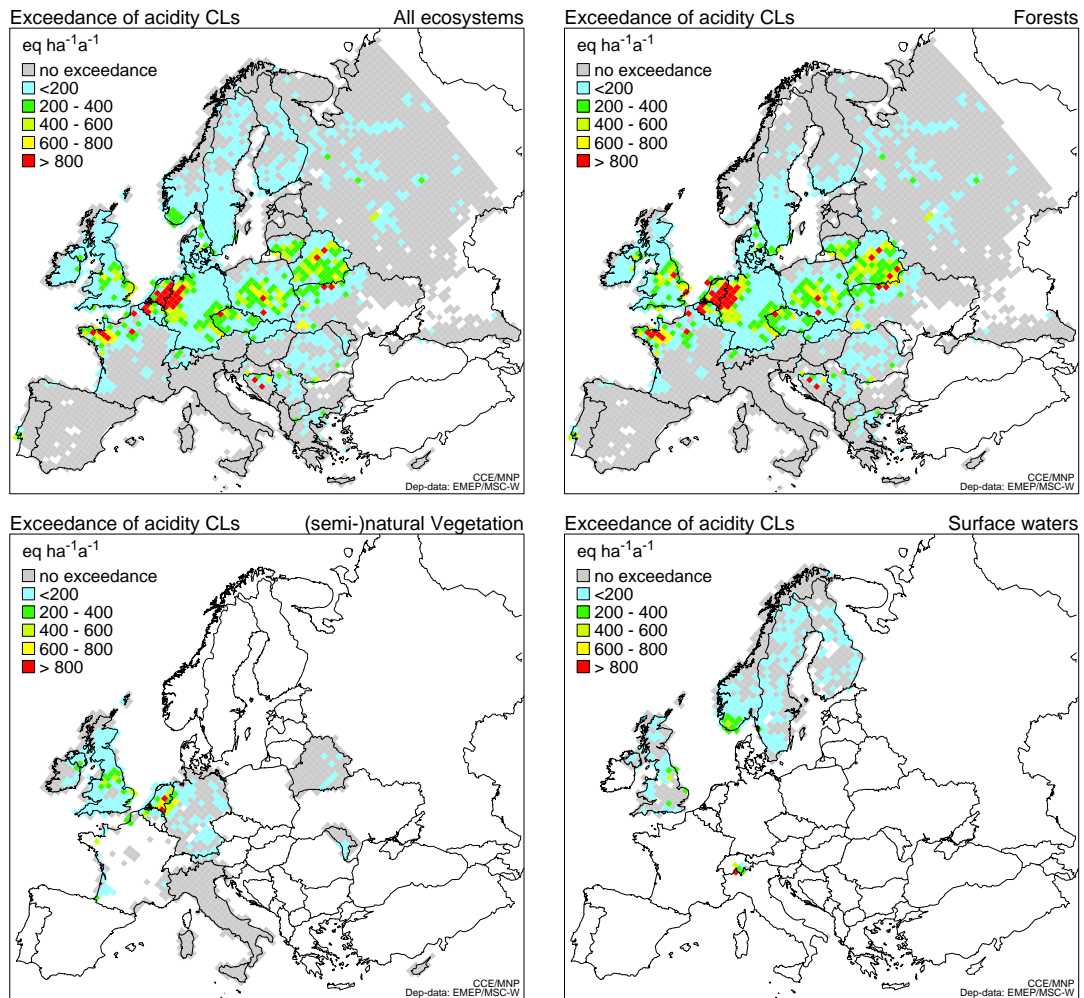


Figure 11. European maps of exceedance of critical loads in the year 2010 for several ecosystem types given full implementation of the Gothenburg protocol and other legislation (CLE scenario). The maps show that for all ecosystem types critical loads will still be exceeded in areas in several countries. White areas indicate 'no data' for the respective ecosystem class (courtesy of Coordinating Centre for Effects; see also Hetteling et al. 2007).

5. Time lags – dynamic models

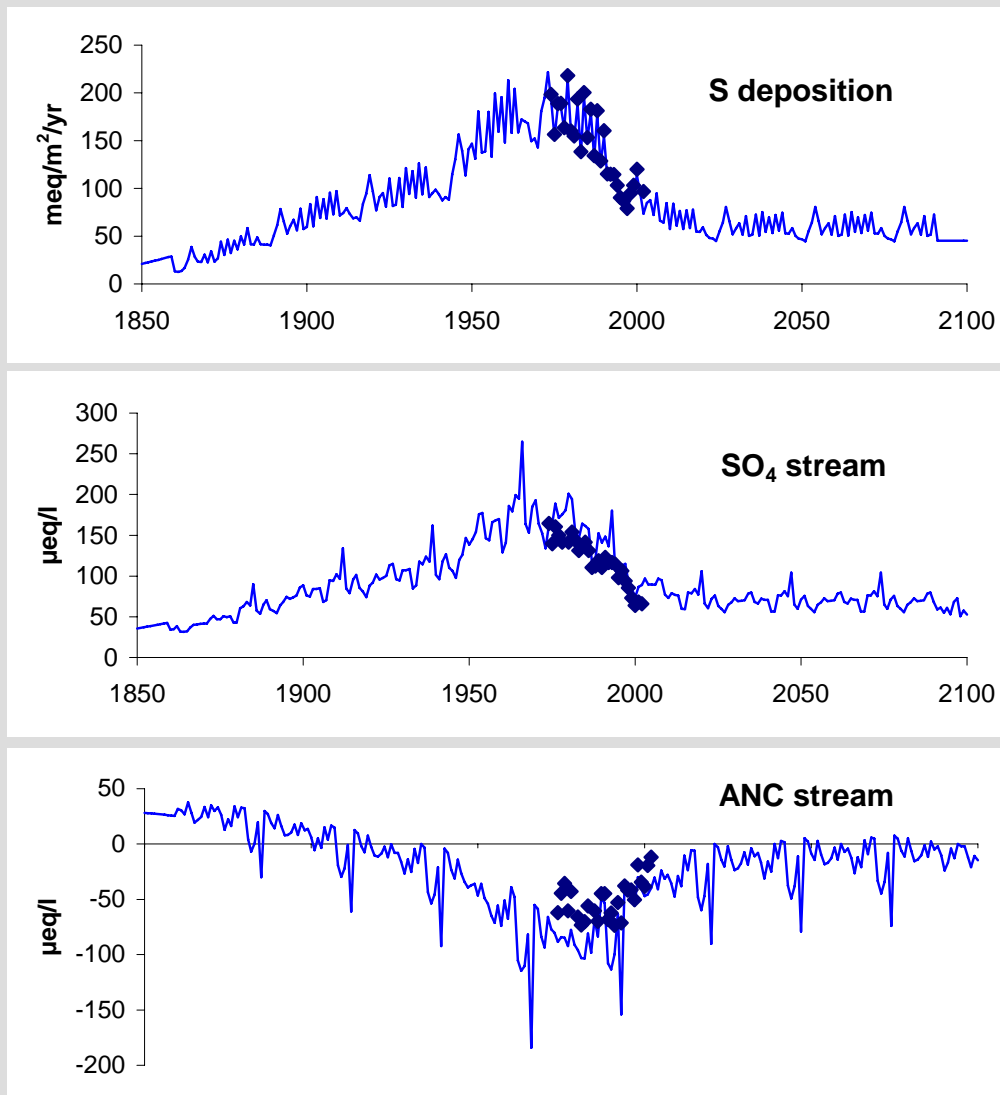
Dynamic modelling

In a policy-oriented framework, dynamic models are needed to explore the temporal aspect of ecosystem protection and recovery. The critical load concept, used for defining the environmental protection levels, does not reveal the time scales of impacts/recovery. Dynamic models have been developed and used for the emission/deposition scenario assessment at selected ICP IM sites (e.g. (Forsius et al. 1998, Jenkins et al. 2003, Jenkins et al. 2003), and applied on large surface water databases compiled by ICP W and national programmes (Wright et al. 2005). These models are flexible and can be adjusted for the assessment of alternative scenarios of policy importance. These modelling studies have shown that the recovery of soil and water quality of the ecosystems is determined by both the amount and the time of implementation of emission reductions. According to the models, the timing of emission reductions determines the state of recovery over a short time scale (up to 30 years). The quicker the target level of reductions is achieved, the more rapidly the surface water and soil status recover. The model simulations also indicate that N emission controls are very important to enable the maximum recovery in response to S emission reductions. Increased nitrogen leaching has the potential to not only offset the recovery predicted in response to S emission reductions but further to promote substantial deterioration in pH status of freshwaters and other N pollution problems in some areas of Europe (Wright et al. 2005).

For surface waters MAGIC (Cosby et al. 1985, Cosby et al. 1985, Cosby et al. 2001) is the most widely-used dynamic model. MAGIC has been applied and tested at in a large number of site-specific and regional applications in Europe, North America and other acid sensitive regions throughout the world. MAGIC has been shown to well simulate response of water chemistry to experimentally-manipulated changes in acid deposition (removal by roof and additions) such as those of the RAIN project in Norway, the roofed catchment experiment at Gårdsjön in Sweden, and the roof experiment at Klosterhede in Denmark (Wright and Cosby 1987, Beier et al. 2003). MAGIC well accounts for the observed recovery during the past 20-30 years (see Text box 3).

Text box 3.

Dynamic modelling of acidification and recovery. Example from Birkenes, a small acid-sensitive stream in southernmost Norway. The MAGIC model (solid line) was used to reconstruct the acidification history and to predict the future recovery given implementation of the Gothenburg protocol. Measurements (points) carried out since 1972 show significant recovery since the mid-1980s in sulphate deposition, streamwater sulphate and streamwater ANC. Modified from Larssen (2005).



Wright et al. (2005) applied the MAGIC and SMART models to 12 acid sensitive surface water regions in Europe (as part of the EU-project RECOVER: 2010), (Figure 12 and Figure 13). The model results indicated that even after complete implementation of the Gothenburg Protocol and other current legislation, acidification with commensurate adverse biological effects will continue to be a significant problem in southern Norway, southern Sweden, the Tatras, the Italian Alps, and the Southern Pennines in the United Kingdom. More than 5% of the ecosystems in each of the regions evaluated would not meet the ANC criterion to protect sensitive aquatic organisms (Figure 12). Additional mitigation measures would be required in these regions to meet long-term European policy objectives. The model simulations also indicated that, as expected, the percent base saturation (%BS) of soils decreased during the long period of acidification of 1860–1980. Between 1980 and 2000, the large reductions in sulphur deposition appeared in most cases to be sufficient to stop the decrease in %BS but still insufficient to allow %BS to recover. The prognosis for the future indicated little or no recovery of base saturation in the soil, and in one of the modelled regions (the Tatra Mountains in Slovakia) the soil would continue to acidify (Figure 13).

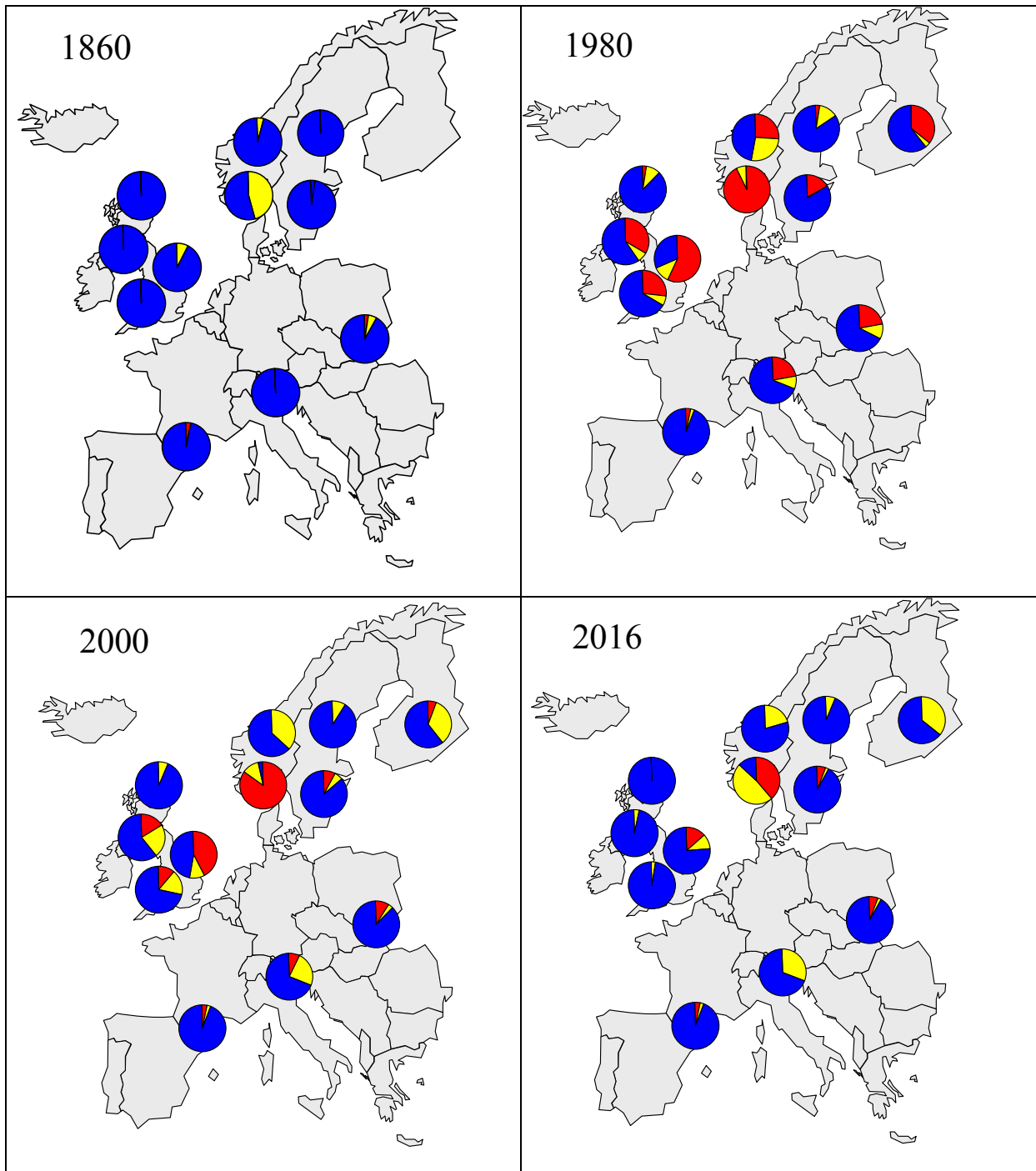


Figure 12. Simulated ANC concentrations in acid-sensitive surface waters in 12 regions of Europe. The three ANC classes correspond to the probability of viable populations of brown trout and other key indicator organisms. Red: ANC < 0 $\mu\text{eq/l}$ and barren of fish; yellow: ANC 0-20 $\mu\text{eq/l}$ and sparse population of fish; blue: ANC > 20 $\mu\text{eq/l}$ and good population of fish. Simulations made using the acidification model MAGIC (SMART in Finland). Four key years are shown: 1860 pre-acidification; 1980 maximum acidification; 2000 present; 2016 complete implementation of emission reduction protocols (Gothenburg and other current legislation). Results from the EU project RECOVER:2010 (Wright et al. 2005).

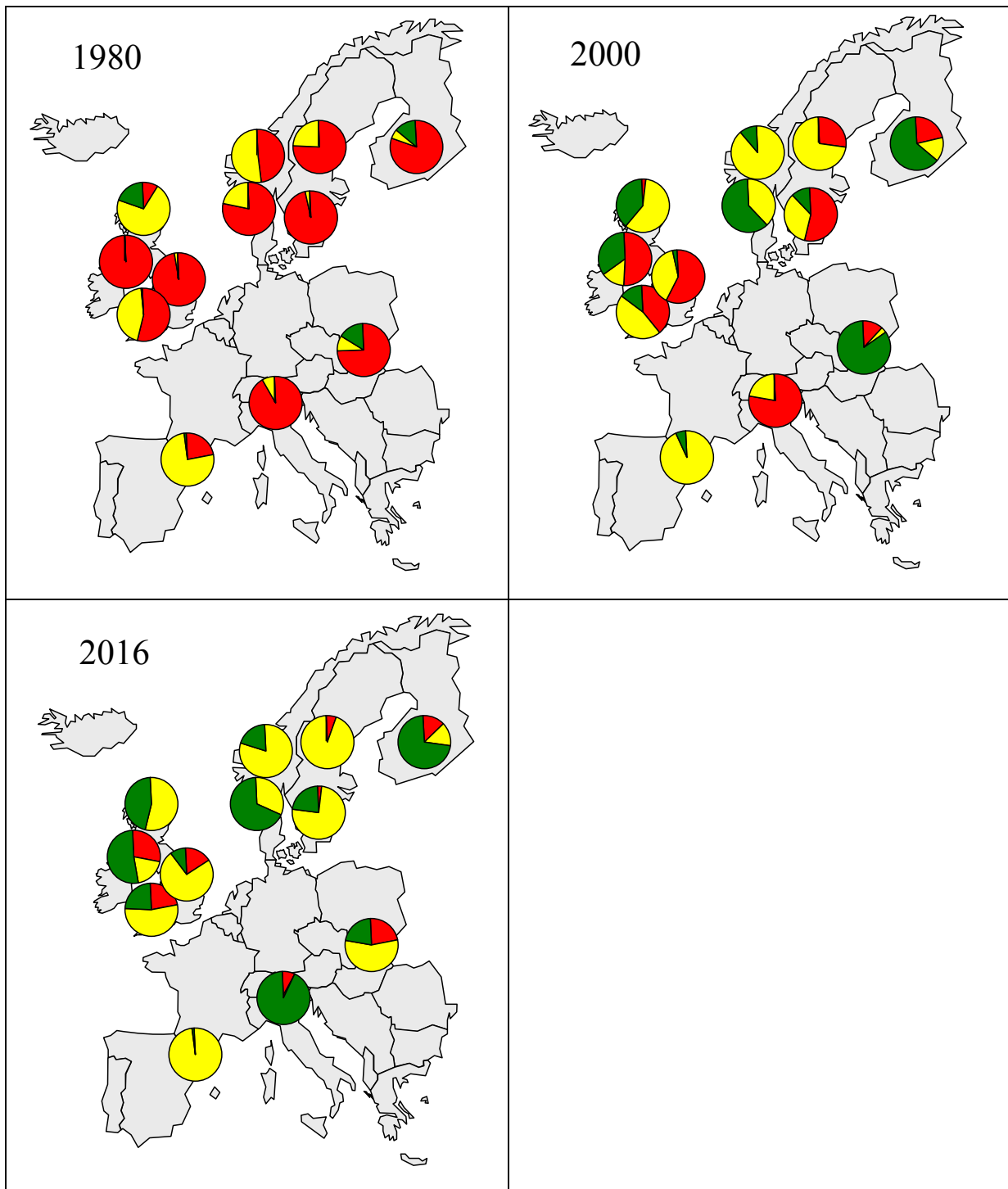


Figure 13. Simulated incremental changes $[(year_{n+1} - year_n)/year_n]$ in base cation pools in catchment soils of acid-sensitive lakes in 12 regions of Europe. Red: depletion by $> 0.1\%/yr$; yellow: negligible change; green: replenishment by $> 0.1\%/yr$. Simulations made using the acidification model MAGIC (SMART in Finland). Three key years are shown: 1980 maximum acidification; 2000 present; 2016 complete implementation of emission reduction protocols (Gothenburg and other current legislation). Results from the EU project RECOVER:2010 (Wright et al. 2005).

Reducing depositions to (or below) critical loads ensures that an ecosystem recovers, but critical loads do not give any information on when a recovery will occur. If the goal is to ensure recovery by a given year (and the time thereafter) one has to determine a so-called target load (for a specified target year). Target loads can only be determined by dynamic models, and meaningful target loads are always smaller than the critical load for the same ecosystem. In fact, a critical load can be viewed as the target load for target year ‘infinity’.

For more information on the concept of target loads and their calculation see (Jenkins et al. 2003, Posch et al. 2003, UBA 2004, UNECE 2005, Hettelingh et al. 2007).

Within the LRTAP Convention, target loads have been calculated by a number of countries, and the results are collated by the Coordination Centre for Effects (CCE) under the ICP on Modelling & Mapping and reported to the Working Group on Effects for use in integrated assessment (Posch et al. 2005, UNECE 2005, Hettelingh et al. 2007). European maps of target loads have been presented, but since only a selected percentile can be shown for every EMEP grid cell, comparisons between target loads for different target years and with the corresponding critical load are difficult. Alternatively, Figure 14 shows examples of comparing these quantities for a selected region (here: Czech Republic and France) by multiple correlations (‘wind mill’ plots). These plots show in a quantitative way the differences between target loads for different target years as well as between critical loads and target loads. The larger these differences (i.e. the further from the 1:1 line) the more stringent are the target loads compared to critical loads. Here the plots for the Czech Republic and France are similar in that both indicate that the target loads are lower than the critical loads (as expected) and that the target load increases with time (i.e. if the system is given longer time to respond to a reduction in acid deposition). Thus such a windmill plot allows a quick semi-quantitative assessment of the additional emission reductions needed to guarantee ecosystem recovery at an early date. A more comprehensive discussion can be found in Posch et al. (2005).

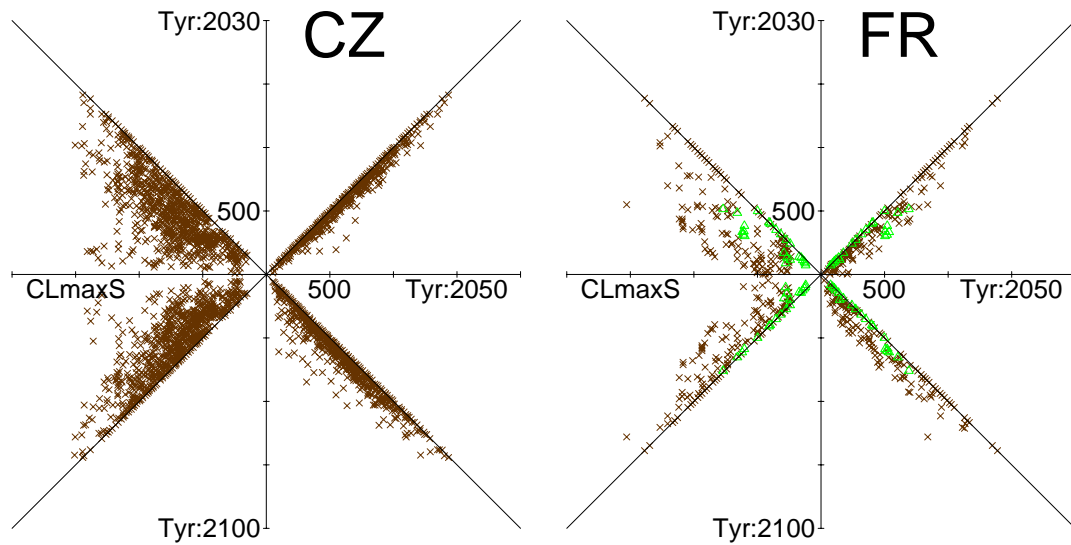


Figure 14. *Quadruple correlations ('windmill plots') between acidity target loads for the target years (Tyr) 2030, 2050, 2100 and the corresponding critical load, CLmaxS, modelled for 1667 sites in the Czech Republic (left) and 386 sites in France (right) (brown: forest; green: semi-natural vegetation; from Posch et al. (2005)).*

6. Climate change as confounding factor

Environmental factors other than acid deposition – so-called ‘confounding factors’ – are expected to affect chemical and biological recovery of freshwaters in response to reduced acid deposition. Predictions made with dynamic models make several assumptions with respect to environmental conditions in the future. Predictions are usually based on assumed scenarios for future emissions of S and N and the resulting acid deposition, and also often on scenarios of future land-use practices, such as forest cutting and replanting. There are, however, other environmental factors that may change in the future and that may affect recovery of ecosystems. These “confounding factors” add to the uncertainty in predictions. Climate change is one of these confounding factors. Others may include land-use change, changes due to other pollutants such as heavy metals and toxic organic pollutants, as well as shifts in the biological components of the ecosystems caused by, for example, invasion of exotic species.

Climate contributes considerably to variability in surface water chemistry. Climate change may both enhance and delay recovery depending on region and variable considered. Future global change introduces another uncertainty to the predictions of acidification recovery. In a joint study modelling study of ICP IM and ICP W (conducted under the framework of the EU project EUROLIMPACS) the relative sensitivity of different climate change related processes affecting acidification recovery was investigated (Figure 15) (Wright et al. 2006). The results showed that several of the factors are of only minor importance (increase in partial pressure of CO₂ in soil air and runoff, for example), several are important at only a few sites or specific to particular regions (e.g. seasalts at near-coastal sites), and several are important at nearly all sites (increased concentrations of organic acids in soil solution and runoff, for example). In addition changes in forest growth and decomposition of soil organic matter are important at forested sites and sites at risk of nitrogen saturation. The trials suggested that in future modelling of recovery from acidification should take into account possible concurrent climate changes and focus specially on the climate-induced changes in organic acids and nitrogen retention.

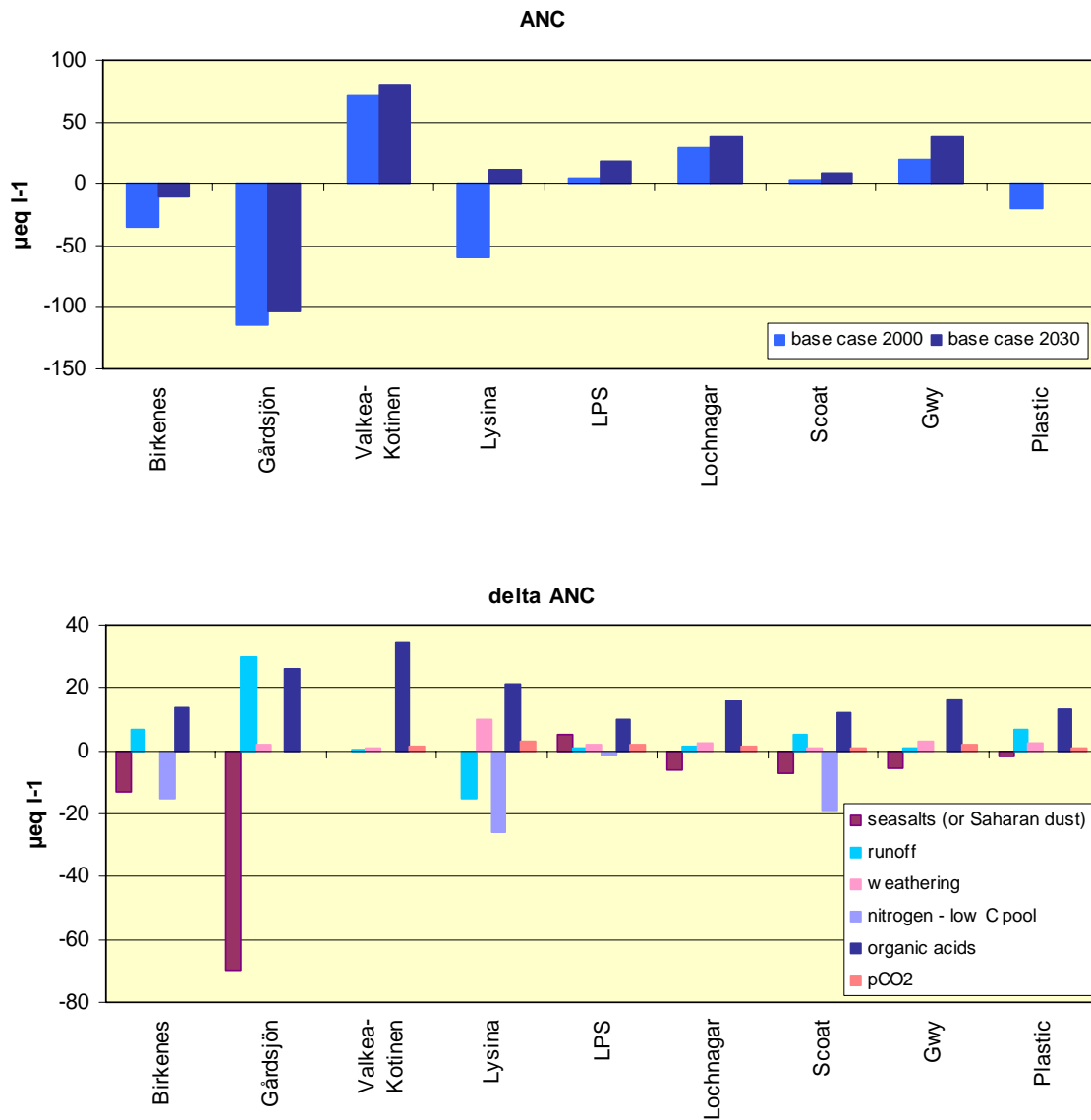


Figure 15. Top panel: ANC (volume-weighted annual mean) concentrations in runoff at 9 sites (8 in Europe, 1 in Canada) for the calibration year (2000) and predicted (using the dynamic model MAGIC) for the year 2030 assuming no climate change (base scenario). Bottom panel: Change in ANC predicted for the year 2030 relative to the base scenario (Wright et al. 2006).

7. Conclusions and recommendations

- Monitoring data from EMEP, ICP Forests, ICP IM and national sources show clear reductions (60-80%) in sulphur deposition in Europe since the peak years in the early 1980s. Nitrogen deposition has also declined, but to a much lesser extent
- Monitoring data from ICP W and ICP IM as well as other sources show clear and large regional trends in surface water chemistry in response to the large decreases in deposition of sulphur since the mid-1980s.
- Waters have become less acidic and less toxic to biota. At many sites sulphate concentrations now approach new levels expected following full implementation of the Gothenburg protocol.
- Nitrate does not show consistent trends, and most sites are far from steady-state conditions. The nitrogen issue clearly remains as a key air pollution issue.
- In 2000 critical loads of acidity were exceeded in freshwater ecosystems in many parts of Europe
- Dynamic models indicate that a significant number of sites in several regions of Europe will continue to be acidified after 2010.
- Biological recovery has begun in many regions, but lags behind chemical recovery.
- Further improvement is predicted with full implementation of the Gothenburg protocol, but verification requires continued chemical and biological monitoring.
- Future climate change will affect acidification and recovery.
- The long-term comprehensive data sets from background sites collected by ICP W and ICP IM are very valuable also for detecting impacts of climate and global change. The use of the data in this context is likely to increase in the future. They also provide the basis more model development and applications.

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