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## Recovery from acidification in European surface waters

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### Abstract

Water quality data for 56 long-term monitoring sites in eight European countries are used to assess freshwater responses to reductions in acid deposition at a large spatial scale. In a consistent analysis of trends from 1980 onwards, the majority of surface waters (38 of 56) showed significant ( $p \leq 0.05$ ) decreasing trends in pollution-derived sulphate. Only two sites showed a significant increase. Nitrate, on the other hand, had a much weaker and more varied pattern, with no significant trend at 35 of 56 sites, decreases at some sites in Scandinavia and Central Europe, and increases at some sites in Italy and the UK. The general reduction in surface water acid anion concentrations has led to increases in acid neutralising capacity (significant at 27 of 56 sites) but has also been offset in part by decreases in base cations, particularly calcium (significant at 26 of 56 sites), indicating that much of the improvement in runoff quality to date has been the result of decreasing ionic strength. Increases in acid neutralising capacity have been accompanied by increases in pH and decreases in aluminium, although fewer trends were significant (pH 19 of 56, aluminium 13 of 53). Increases in pH appear to have been limited in some areas by rising concentrations of organic acids. Within a general trend towards recovery, some inter-regional variation is evident, with recovery strongest in the Czech Republic and Slovakia, moderate in Scandinavia and the United Kingdom, and apparently weakest in Germany.

**Keywords:** acidification, recovery, European trends, sulphate, nitrate, acid neutralising capacity

### Introduction

The emission, and subsequent deposition, of acidifying sulphur (S) and nitrogen (N) compounds has resulted in the acidification of surface waters in many geologically sensitive regions of Europe (Rodhe *et al.*, 1995). As a transboundary pollution issue, legislation to reduce acidifying emissions has taken place at an international level, through the Convention on Long-Range Transboundary Air Pollution of the United Nations Economic Commission for Europe (UN-ECE). The first agreement to restrict S emissions was reached in 1985 and called for a 30% cut relative to 1980 in all countries. Subsequent agreements have resulted in further emissions reductions for S, the introduction of emissions controls for N, and in a more targeted strategy aimed at reducing critical load exceedance for terrestrial and

freshwater ecosystems at the European scale. The most recent multi-pollutant, multi-effect (Gothenburg) protocol, signed in 1999, incorporates additional air pollutants and is intended to bring a 63% reduction in European S emissions relative to 1990 (approximately 80% relative to 1980), and a 41% reduction in oxidised N emissions relative to 1990 (UN-ECE, 1999). Current S emissions are less than half the 1980 levels, while oxidised N emissions have declined by about 20% (Verstreng and Støren, 2000).

Given these major reductions in emissions of the dominant acidifying pollutant over the last two decades, substantial improvements should have occurred in the affected ecosystems over the same period. Analysis of long-term trends in freshwater data provides an important means to assess whether this expected improvement has in fact

occurred and, if so, to what extent this represents a return to 'good ecological status' as mandated by the European Union Water Framework Directive (EU, 2000). Long-term data also aid understanding of the acidification process and contribute to the development of critical loads, dynamic models and prediction of the benefits of current and future emissions control agreements.

Long-term water quality trends have previously been examined for many different sites (e.g. Christophersen *et al.*, 1990; Reynolds *et al.*, 1997), regions (e.g. Harriman *et al.*, 1995; Soulsby *et al.*, 1997; Veselý *et al.*, 1998; Kopáček *et al.*, 1998; Mosello *et al.*, 2000) and countries (e.g. Mannio and Vuorenmaa, 1995; Skjelkvåle *et al.* 1998; Wilander and Lundin, 1999; Monteith and Evans, 2000). The co-ordinated international strategy on emission controls suggests that trends should be qualitatively similar from country to country; therefore, combining trend data for different countries should provide valuable additional information regarding the spatial extent, consistency and magnitude of recovery at the European scale. To date, studies of long-term trends in European surface waters have been undertaken within the framework of the International Co-operative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes (ICP-Waters), which compiles data from monitoring programmes throughout much of Europe and North America. Stoddard *et al.* (1999) analysed trends for 1980-1989 (41 sites) and 1990-1995 (74 sites) for the ICP-Waters network with additional data for the UK from the Acid Waters Monitoring Network. Sites were divided into three regions (Nordic Countries, United Kingdom, North/Central Europe). Skjelkvåle *et al.* (2000) updated this with an assessment of trends between 1989 and 1998 for 64 European surface waters. The regions selected for this analysis (Northern Nordic Countries, Southern Nordic Countries and UK, and Central Europe) differed from those selected by Stoddard *et al.* (1999). Both studies provided evidence for recovery from acidification at many sites but trends varied considerably between regions. In particular, few recovery trends were observed in the UK and in continental Europe, although large sulphate ( $\text{SO}_4$ ) reductions were recorded, Skjelkvåle *et al.* (2000) found significant heterogeneity in acidity trends.

The current study builds on the results of previous work by examining trends at 56 sites included in the RECOVER:2010 project. Most of these sites are not part of ICP-Waters and they, therefore, provide a validation of previous observations, as well as incorporating more recent data. Site trends are considered individually, but an attempt is made to identify general patterns of chemical change at the European scale and the extent to which systematic differences arise between trends in a total of six regions

(primarily individual countries). The European-scale assessment presented here is also supported by more detailed analyses of trends in each individual region by Alewell *et al.* (2001), Rogora *et al.* (2001), Moldan *et al.* (2001), Skjelkvåle *et al.* (2001), Evans and Monteith (2001) and Kopáček *et al.* (2001).

## Site description and methodology

The analysis of trends for RECOVER:2010 draws together data for a total of 56 lakes and streams from eight European countries (Fig. 1). These sites cover a wide range of acid-sensitive landscapes, from high-altitude alpine meadows in the Italian Alps and Tatra Mountains of Slovakia through Central European and Scandinavian forests to peaty moorlands in the UK (Table 1). A full list of the sites used is given in Appendix 1 and more detailed site descriptions are provided elsewhere in this volume.

Since these data have been collated from a large number of different monitoring programmes, there is inevitably some heterogeneity in dataset characteristics, with start dates ranging from 1971 to 1988, sampling frequencies from weekly to monthly at streams, and monthly to yearly at lakes, and varying sets of measured determinands. Here, a set of six key variables are considered: acid neutralising capacity (ANC), pH, non-marine sulphate ( $\text{xSO}_4$ ), calcium (Ca), nitrate ( $\text{NO}_3$ ), and soluble aluminium (Al). These variables were measured at all study sites with the exception of Al, for which data were unavailable for three of the five Italian sites. ANC was calculated as the difference in the equivalent sum of base cations (Ca, magnesium (Mg), sodium (Na), potassium (K)) and acid anions (chloride (Cl),  $\text{SO}_4$ ,  $\text{NO}_3$ ) in all regions except the UK, where it was calculated by adjusting measured alkalinity for inorganic aluminium and organic acids (Evans and Monteith, 2001). This alternative estimate is theoretically equivalent to ANC calculated from the difference between the sum of base cations and the sum of acid anions. It is used for sites in the UK, which have high sea-salt concentrations, because it is less influenced by measurement errors at high ionic strengths (Evans *et al.*, 2001). Non-marine  $\text{SO}_4$  was calculated by subtracting the marine  $\text{SO}_4$ , calculated based on the sea-water  $\text{SO}_4/\text{Cl}$  ratio, from total  $\text{SO}_4$ . In central Europe, where marine inputs are minor,  $\text{xSO}_4$  is approximately equal to  $\text{SO}_4$ , but in coastal areas of the UK and Scandinavia, it is necessary to consider  $\text{xSO}_4$  in order to distinguish changes in anthropogenic  $\text{SO}_4$  from climate-related variations in sea-salt deposition.

The six variables selected illustrate the main surface water responses to changes in acid deposition, with  $\text{xSO}_4$  and  $\text{NO}_3$  representing the major acidifying anions, and ANC, pH and Al providing measures of surface water acidity and toxicity.

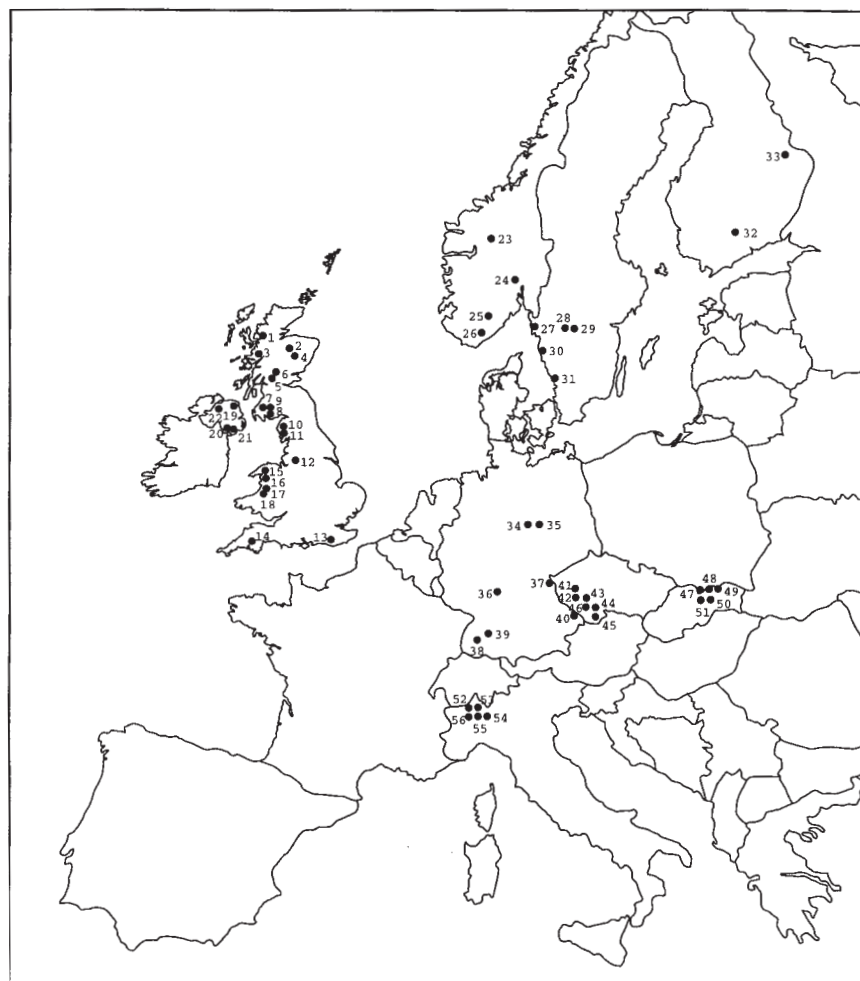


Fig. 1. Sites used in European trend assessment. Site numbers correspond to those in Appendix 1

Table 1. General characteristics of monitoring sites in each country

Country	Number of lakes	Number of streams	Altitude range	Land-cover	Soils	Geology	ANC range ( $\mu\text{eq l}^{-1}$ )
Italy	2	3	193 to 2269	Rock/alpine meadow (lakes) Mixed forest (streams)	Rankers	Granite, gneiss, micaschist	4 to 216
Slovakia	5	-	1477 to 2145	Rock/alpine meadow (3) coniferous forest (2)	Thin podzolic	Granite	-11 to 280
Czech Republic	6	-	1008 to 1096	Spruce forest	Podzols	Schist, gneiss, granite	-38 to 7
Germany*	-	7	385 to 1355	Spruce forest (6) Beech forest (1)	Cambisols, podzols	Granite, gneiss, sandstone	-140 to
Finland	-	2	150 to 165	Spruce, pine, birch, aspen	Podzols, peats, cambisols	Gneiss, granodiorite	71 to 105
Sweden	-	5	75 to 245	Spruce, pine, birch, aspen, heather	Podzols	Gneiss, granite, moraine	
Norway	-	4	200 to 580	Spruce, pine, birch, heather	Podzols, peaty rankers	Granite, gneiss	-38 to 2
United Kingdom	11	11	10 to 785	Grass/heather moorland (17) Coniferous forest (5)	Peat, podzol, gley, ranker	Granite, schist, sedimentary.	-32 to 168

\* Sampling at two German sites (Schluchsee and Villingen) ceased in 1996.

Calcium represents the major source of base cation buffering at most sites and unlike the other main base cations, Na and Mg, is not strongly affected by a large marine component. Chloride, which is primarily marine-derived, exhibits few long-term trends and was not therefore considered here. Other variables of interest, such as Dissolved Organic Carbon (DOC), alkalinity, and the inorganic (toxic) and organic (non-toxic) components of Al, could not be incorporated into the European-scale analysis as they were not measured at a sufficient number of sites. Additional variables are, however, considered where data are available in the accompanying regional studies.

For data series with limited data (e.g. annual), the Mann-Kendall Test (MKT) was applied (Mann, 1945; Kendall, 1975). MKT is a non-parametric method of trend analysis which is robust with regard to non-normality, missing or censored data. All data pairs within a time series ( $x_j$ ,  $x_k$ , where  $j > k$ ) are assigned a function  $\text{sgn}(x_j - x_k)$  with values of +1, 0 or -1 depending on the sign of  $x_j - x_k$ . These are then summed to give the Mann-Kendall statistic  $S$  (Eqn. 1):

$$S = \sum_{k=1}^{n-1} \sum_{j=k+1}^n \text{sgn}(x_j - x_k) \quad (1)$$

where  $n$  is the number of data values. The value of  $S$ , together with calculated variance,  $\text{Var}(S)$ , are used to derive a test statistic  $Z$ , which is positive if the trend is upward and negative if the trend is downward. The magnitude of  $Z$  is used to estimate trend significance. Trend slopes are obtained using the method of Sen (1968), as the median of all between-year differences between data values. MKT was used for the Czech, Slovak and Italian lakes.

For time series with a greater sampling frequency, the Seasonal Kendall Test (SKT) was applied. SKT is a refinement of MKT which is robust with respect to seasonality in time series, whereby data are first grouped into either monthly or quarterly blocks (Hirsch *et al.*, 1982).  $S$  and  $\text{Var}(S)$  are calculated within each seasonal block and summed for all seasons to give full dataset statistics  $S'$  and  $\text{Var}(S')$ , which are then used to compute  $Z$ . The estimate of  $\text{Var}(S')$  was obtained using a modified procedure presented by Hirsch and Slack (1984), which provides a more robust significance test with respect to serial correlation. Trend slopes are again calculated according to the method of Sen (1968), as the median of between-year differences in values within each seasonal block. SKT has been identified as a suitable method for analysis of water quality data (Taylor and Loftis, 1989) and applied in a number of monitoring programmes, including ICP-Waters (Stoddard *et al.*, 1999) and the United States Long Term Monitoring project (e.g.

Driscoll and van Drearon, 1993).

Given the close methodological similarities between MKT and SKT, the trend significances and slopes obtained at different sites should be approximately comparable. Both methods, however, are only able to detect monotonic trends within time series. As a consequence, and because the aim of this study was to assess whether recovery has occurred in response to falling levels of acid deposition, it was necessary to restrict analysis to the period at, or following, the peak in acid deposition. For this reason, and to limit the impact of varying monitoring periods at different sites, a 1980 start date was used for trend analyses at all sites with longer-term data. A significance threshold of  $p \leq 0.05$  was applied to both trend tests. A second threshold of  $p \leq 0.20$  was also used to identify weak, but potentially genuine, trends.

## Results

For each variable, estimated slopes for all 56 sites are ranked (Fig. 2) in order to identify overall patterns of concentration change during the study period. Given the potential dominance of the 22 UK sites within this assessment, sites were also subdivided into six regions, for which median and percentile slopes were calculated (Table 2, Fig. 3). Due to the small number of Finnish and Norwegian sites in the dataset, these were combined with those in Sweden to form a single 'Scandinavia' region of 11 sites. Trend directions are also mapped in Fig. 4.

### NON-MARINE SULPHATE

Clear decreases in surface water  $x\text{SO}_4$  concentrations have occurred throughout the areas of Europe included in this study, with significant ( $p \leq 0.05$ ) declining trends at 38 sites and weak ( $p \leq 0.20$ ) downward trends at a further 10. The largest median decreases have occurred in Slovakia and the Czech Republic, with large and widespread decreases also observed in Scandinavia and Germany (with the exception of one site, Lange Bramke). In all four of these regions, median  $x\text{SO}_4$  reductions have exceeded  $-2.5 \text{ meq l}^{-1} \text{ yr}^{-1}$ . Significant downward trends are observed at all five Italian sites, but are smaller in magnitude (median  $-1.0 \text{ meq l}^{-1} \text{ yr}^{-1}$ ). In the UK, although most sites have shown an overall decrease in  $x\text{SO}_4$ , a period of relatively stable concentrations between 1988 and 1995 (Evans and Monteith, 2001) resulted in fewer sites showing significant downward trends. European trends in  $\text{SO}_4$  are discussed in further detail by Prechtel *et al.* (2001).

Table 2. Summary of trends and median estimated concentration changes by region

	Italy	Slovakia	Czech R	Germany	Scandinavia	UK	All
Number of sites	5	5	6	7	11	22	56
<b>ANC</b>							
Rising ( $p \leq 0.05$ )	2	4	5		6	10	27
Rising ( $p \leq 0.20$ )			1	1	3	4	9
Falling ( $p \leq 0.20$ )	1	1				2	
Falling ( $p \leq 0.05$ )				1		1	
Median change ( $\mu\text{eq l}^{-1} \text{yr}^{-1}$ ) +0.24		+1.80	+4.49	+1.10	+1.33	+1.05	+1.29
<b>pH</b>							
Rising ( $p \leq 0.05$ )	2	1	4	1	5	6	19
Rising ( $p \leq 0.20$ )		2			1	3	6
Falling ( $p \leq 0.20$ )	2	1				1	4
Falling ( $p \leq 0.05$ )					1	1	2
Median change (pH unit $\text{yr}^{-1}$ ) -0.001		+0.013	+0.011	-0.001	+0.008	+0.004	+0.005
<b>xSO<sub>4</sub></b>							
Rising ( $p \leq 0.05$ )				1		1	2
Rising ( $p \leq 0.20$ )						0	
Falling ( $p \leq 0.20$ )			1			9	10
Falling ( $p \leq 0.05$ )	5	5	4	6	11	7	38
Median change ( $\mu\text{eq l}^{-1} \text{yr}^{-1}$ ) -1.00		-4.17	-3.47	-2.60	-2.76	-1.10	-1.92
<b>Ca</b>							
Rising ( $p \leq 0.05$ )			1				1
Rising ( $p \leq 0.20$ )	1						1
Falling ( $p \leq 0.20$ )	1				2	6	9
Falling ( $p \leq 0.05$ )	1	5	4	4	6	6	26
Median change ( $\mu\text{eq l}^{-1} \text{yr}^{-1}$ ) -0.30		-2.77	-0.46	-1.61	-0.83	-0.90	-0.90
<b>NO<sub>3</sub></b>							
Rising ( $p \leq 0.05$ )	4			1	2	3	10
Rising ( $p \leq 0.20$ )				1		3	4
Falling ( $p \leq 0.20$ )	1	1		2	2		6
Falling ( $p \leq 0.05$ )		2	3	2	4		11
Median change ( $\mu\text{eq l}^{-1} \text{yr}^{-1}$ ) +0.54		-0.78	-0.72	-0.61	-0.04	0	0
<b>Al*</b>							
Rising ( $p \leq 0.05$ )			1	1		2	4
Rising ( $p \leq 0.20$ )					1	2	3
Falling ( $p \leq 0.20$ )	1	2		2	4	2	11
Falling ( $p \leq 0.05$ )	1		5	2	1	4	13
Median change ( $\mu\text{g l}^{-1} \text{yr}^{-1}$ ) -1.4		+0.1	-20.5	-4.2	-0.1	+0.1	-0.6

\*Al data only available for two sites in Italy

## NITRATE

Nitrate trends are highly variable among the study sites, with no evidence of consistent Europe-wide changes in concentrations since the 1980s. A substantial number of

sites, mostly in low-deposition areas of the UK and Scandinavia, have near-zero NO<sub>3</sub> trends as a result of continuously low surface water concentrations. The only

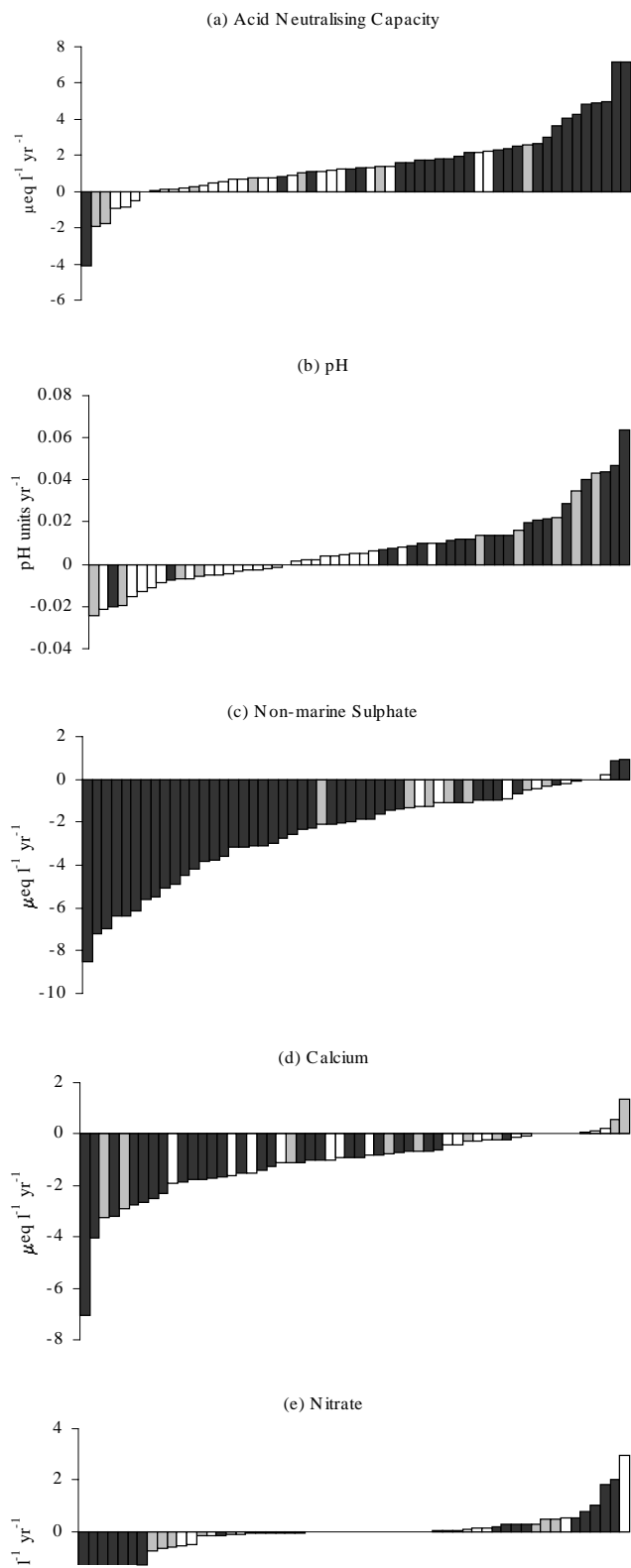


Fig. 2. Ranked trend slopes for all analysed surface waters

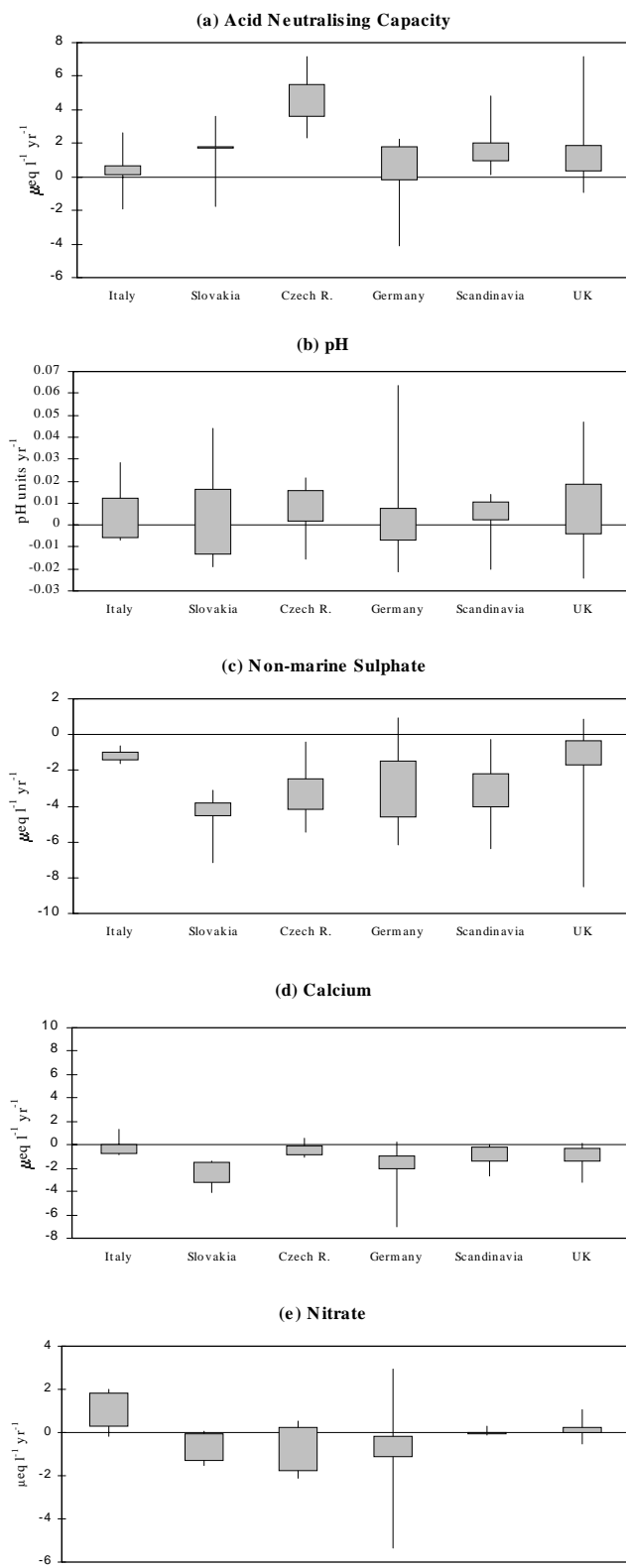


Fig. 3. Range of trend slopes observed within each region. Boxes represent interquartile range of slope estimates, lines show maximum and minimum observed slopes. Note that all slope estimates are included in calculation, regardless of significance.

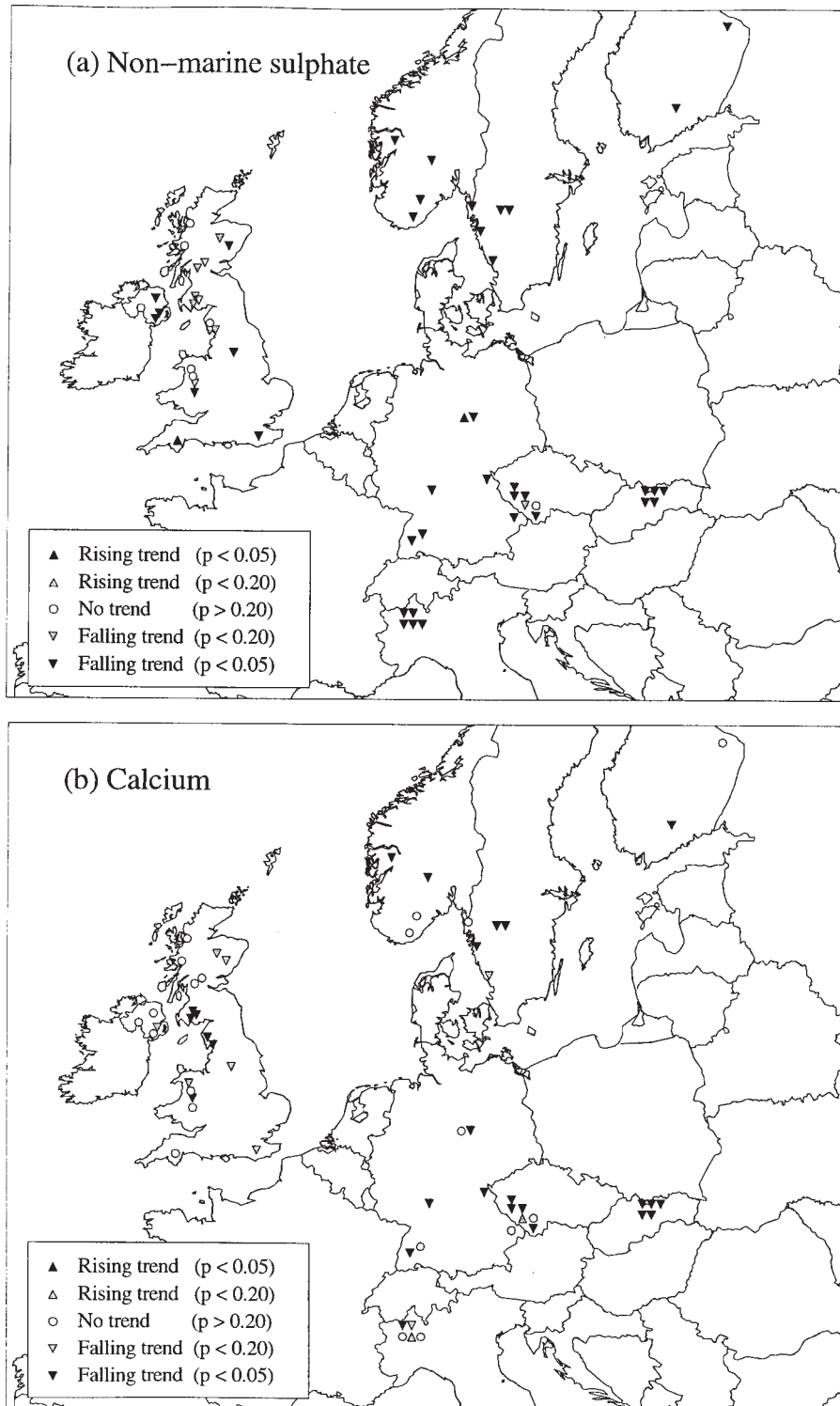


Fig. 4. Spatial distribution of observed trends

area with clear evidence of rising concentrations is Italy, where significant increases have occurred at four sites and may be indicative of decreasing catchment N retention (Mosello *et al.*, 2001). A small number of sites in higher-deposition areas of the UK have also shown significant

increases, but most trends here appear to result from a climate-related  $\text{NO}_3$  peak in 1996 (Evans and Monteith, 2001). The largest increase in  $\text{NO}_3$  occurred at Markungsgraben, following severe forest damage caused by a bark beetle infestation starting in 1993 (Alewell *et al.*,



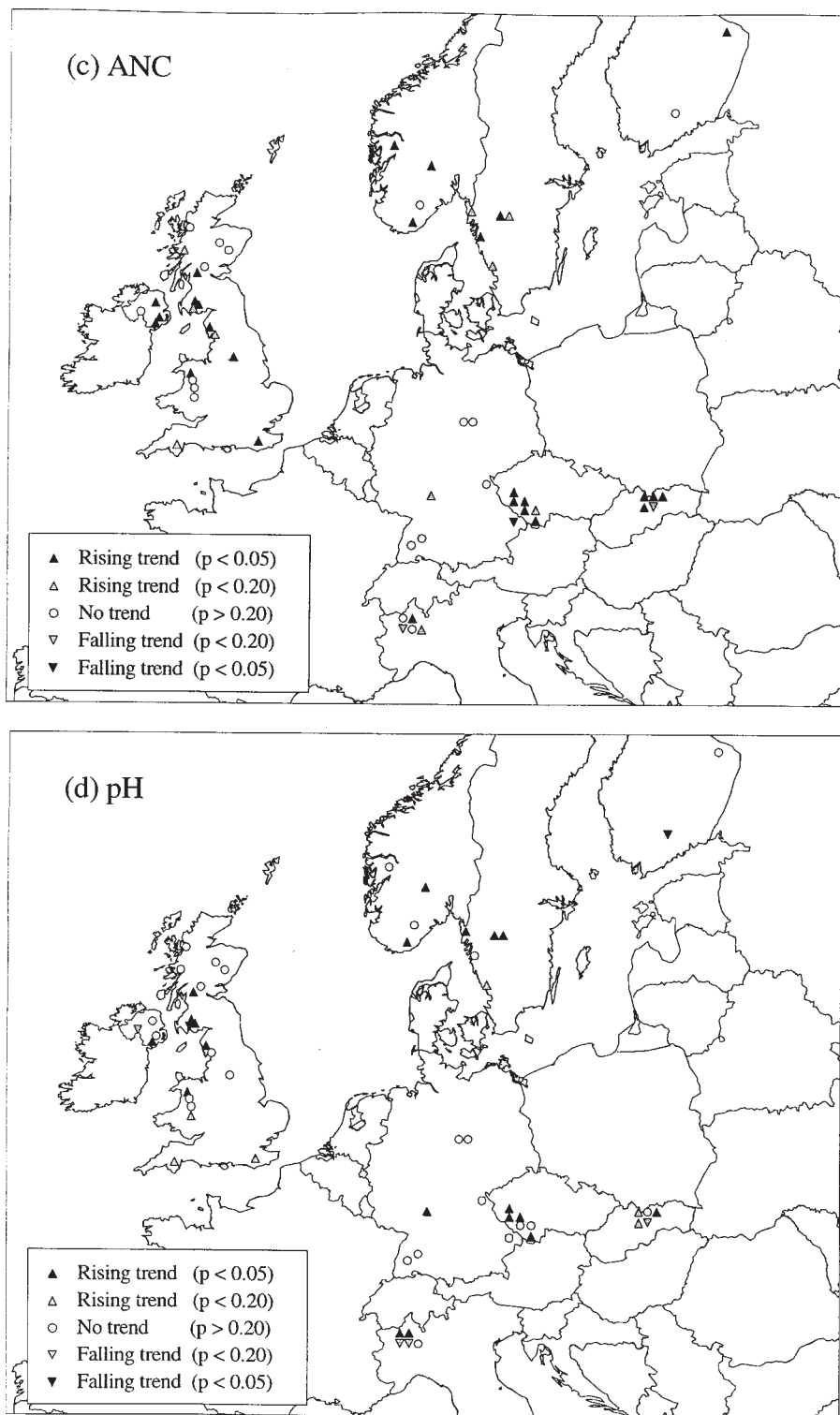


Fig. 4 (contd). Spatial distribution of observed trends

2001). Many sites in Central Europe (Germany, Slovakia and the Czech Republic), however, recorded decreases in  $\text{NO}_3$ , probably reflecting reductions in N deposition during the 1990s (Verstreng and Støren, 2000; Barret *et al.*, 2000).

Smaller but significant downward trends were observed at four sites in Scandinavia. European trends in  $\text{NO}_3$  are discussed in further detail by Wright *et al.* (2001).

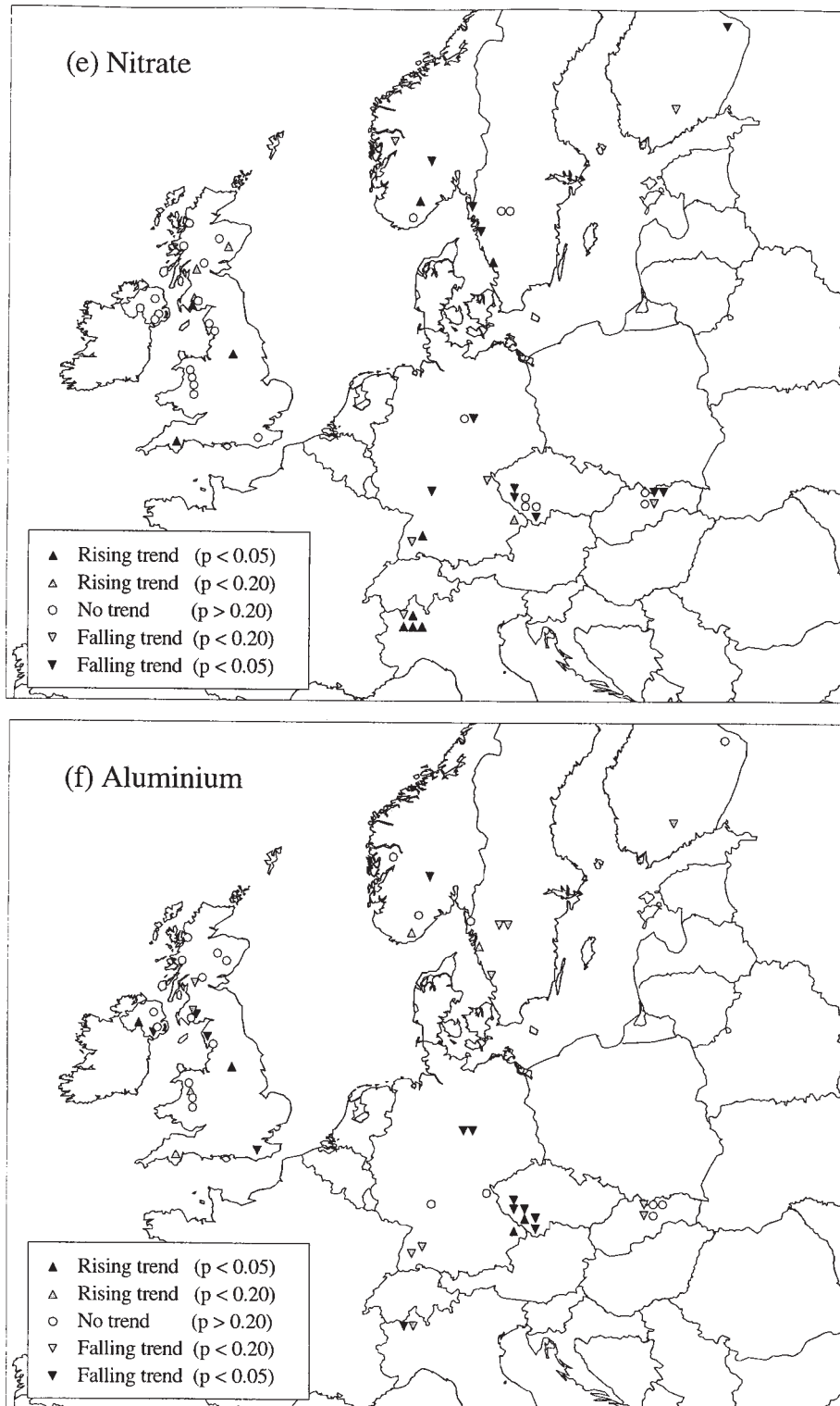


Fig. 4 (contd). *Spatial distribution of observed trends*

#### ACID NEUTRALISING CAPACITY

There is a clear tendency towards rising ANC in lakes and streams throughout the study regions; of the 56 sites, 27 exhibit significant rising trends, and weak increases are

observed at a further nine. The majority of sites show increasing ANC in all regions except Germany, where only one weak rising trend is recorded (Table 2). In no region do more than two sites show an ANC decline and the only site

at which ANC has decreased significantly is Markungsgraben in Germany, due to the  $\text{NO}_3$  pulse described above. The greatest median ANC increase ( $4.5 \mu\text{eq l}^{-1} \text{yr}^{-1}$ ) has been observed in the Czech Republic and the smallest ( $0.7 \mu\text{eq l}^{-1} \text{yr}^{-1}$ ) in Italy; overall the median increase has been  $1.3 \mu\text{eq l}^{-1} \text{yr}^{-1}$ .

## CALCIUM

Changes in Ca appear to correspond closely to those in  $\text{xSO}_4$ , with significant downward trends at 26 sites and weak downward trends at nine additional sites. The overall median trend slope of  $-0.9 \mu\text{eq l}^{-1} \text{yr}^{-1}$  is just under half of the median slope for  $\text{xSO}_4$  ( $-1.9 \mu\text{eq l}^{-1} \text{yr}^{-1}$ ), but substantial inter-regional variations are apparent, with much smaller proportional Ca reductions in the Czech Republic, and to a lesser extent Scandinavia and Italy, and larger proportional reductions in Slovakia, Germany and the UK. Observed decreases can be explained in part by reductions in Ca deposition, particularly in Central Europe, but at most sites a reduction in leaching from catchment soils is considered to be the main cause (discussed below).

## pH

Nineteen of the 56 sites exhibit significant rising trends in pH and another six show weak increases. Median slope estimates are positive for five regions, but narrowly negative in Germany. As for ANC, the results indicate widespread recovery, but fewer sites show increasing pH than show increasing ANC. The reasons for this apparent discrepancy are discussed below.

## ALUMINIUM

Significant downward trends in Al have been observed at 13 sites and negative slopes at another 15. Only four sites have shown significant rising trends. Although data are limited to two sites for Italy, results for all regions suggest a tendency towards declining Al concentrations (Figs. 3 and 4). Examination of individual regions shows a high degree of heterogeneity, however, with by far the largest decreases observed in the Czech Republic. At five of the Czech lakes, strong recovery from very acidic conditions has resulted in large reductions in Al ( $-13$  to  $-34 \mu\text{g l}^{-1} \text{yr}^{-1}$ ). Trends in Slovakia are generally weak, possibly due to limited Al data for these sites. In Germany, most streams exhibit falling concentrations, but Markungsgraben is again an outlier with sharply rising Al concentrations correlated with the rise in  $\text{NO}_3$ . In the UK, few trends are observed in Al, but this apparent lack of change masks a major shift in speciation at

many sites, from the toxic inorganic forms to non-toxic organic species (Evans and Monteith, 2001). Similar trends in Al species at the four Norwegian sites (SFT, 1999; Lydersen, 1998) indicate that this transition has also occurred in at least part of Scandinavia.

## Discussion

Taking the 56 study sites as a whole, results provide clear evidence for recovery from acidification in terms of falling  $\text{xSO}_4$  concentrations, rising ANC, rising pH and falling Al at a high proportion of sites. Decreases in  $\text{xSO}_4$  are particularly strong and widespread, confirming that surface waters have responded to reductions in anthropogenic S deposition throughout the areas of Europe studied, although release of stored S from catchment soils may have damped this response in some regions, notably Germany (Alewell *et al.*, 2001; Prechtel *et al.*, 2001). With relatively little change in  $\text{NO}_3$  at most sites,  $\text{xSO}_4$  decreases have led to an overall reduction in acid anion concentrations, generating a range of chemical responses. Although qualitatively similar at the majority of sites, quantitative differences in trends can be identified both within and between regions.

As might be expected, the magnitude of ANC trend is negatively correlated with the sum of trends in  $\text{xSO}_4$  and  $\text{NO}_3$ , although there is considerable scatter in this relationship (Fig. 5a;  $R^2 = 0.27$ ,  $p < 0.0001$ ). A slope coefficient of  $-0.49$  implies that, on average, approximately half of the observed reduction in  $[\text{xSO}_4 + \text{NO}_3]$  has translated into increased ANC. Clearly, therefore, part of the acid anion reduction has been balanced by changes in other ions and this is demonstrated by a strong positive correlation between  $\Delta\text{Ca}$  and  $(\Delta\text{xSO}_4 + \Delta\text{NO}_3)$  (Fig. 5b;  $R^2 = 0.59$ ,  $p < 0.0001$ ). Over the timescales involved in this study, the primary mechanism of recovery is thought to be decreasing ionic strength, whereby reduced concentrations of mobile acid anions lead to reduced leaching of both acid cations (H and inorganic Al) and base cations from soils, and consequently to increases in ANC (Reuss and Johnson, 1986). Clearly, this process must involve all base cations to some extent, but a slope coefficient of  $0.41$  for  $\Delta\text{Ca}$  v.  $(\Delta\text{xSO}_4 + \Delta\text{NO}_3)$  suggests that Ca reductions alone can account for around 80% of the imbalance between acid anion reductions and ANC increases when considering the dataset as a whole (Fig. 6). Since ANC increases due to declining ionic strength can only occur while acid anion concentrations continue to fall, longer term recovery must involve an increase in soil base saturation, with base cations from weathering and deposition replacing H and Al on exchange sites (Galloway *et al.*, 1983). However, such a base saturation increase is neither

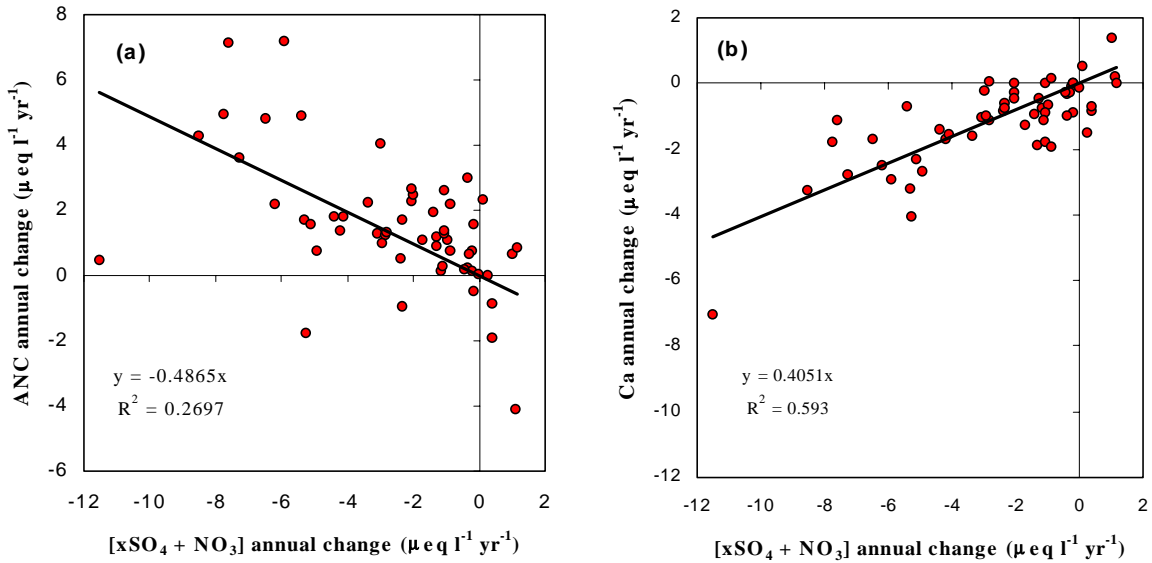


Fig. 5. Relationships between trend slopes for ANC, Ca and  $[xSO_4 + NO_3]$

required nor implied by the short-term surface water trends observed. If present or future acid anion deposition continues to outstrip base cation supply, soil base saturation may continue to fall at some sites.

As noted earlier, the generalised relationship observed between acid anion and base cation changes conceals significant inter-regional variation, with the Czech Republic and Scandinavia showing a proportionally large ANC response to falling acid anion concentrations, and other areas (notably Germany) relatively small ANC responses. This variability can be linked to the buffering capacity of catchment soils; at the highly impacted Czech sites, for example, soils with a severely depleted base saturation have little capacity to buffer changes in acid anions and most of the decrease in acid anions has been accompanied by decreases in acid cations. At poorly buffered or severely depleted sites, therefore,  $\Delta ANC$  will tend towards  $(\Delta xSO_4 + \Delta NO_3)$ , whereas at sites where soil base cations are able to neutralise fully incoming acidity,  $\Delta ANC$  will tend towards zero (Henriksen *et al.*, 1986). At the latter extreme, the absence of ANC ‘recovery’ is thus due to the absence of past freshwater acidification.

The smaller proportion of sites showing increases in pH compared to the proportion at which ANC has risen (Table 2) appears to result from a number of factors. In part it is a function of the logistic relationship between ANC and pH, with pH changes smaller, and harder to detect, relative to changes in ANC at more alkaline sites. However, discrepancies between ANC and pH trends are also observed at some acidic sites. At these sites, ANC increases are comprised both of a decrease in H ion (i.e. an increase in

pH) and a decrease in inorganic Al (Fig. 6). Consequently, trends in each acid cation may be weaker than the trend in ANC. In addition, it appears that in some regions of Europe, pH recovery may have been suppressed by increases in organic acidity. These increases, inferred from rising trends in DOC, are particularly evident in the UK (Evans and Monteith, 2001) and are also observed at a number of streams in Scandinavia. Although data are not available for most other regions, rising trends in chemical oxygen demand

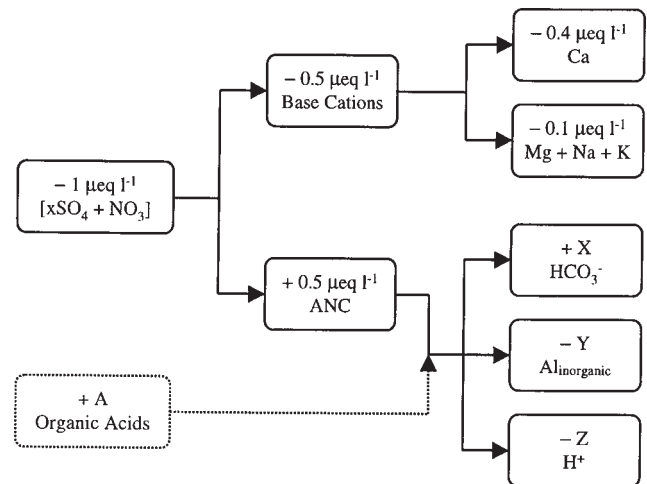


Fig. 6. Schematic view of changes in water chemistry caused by changes in acid deposition. Values given are an ‘average’ case based on trend analyses of the RECOVER:2010 sites, and will vary from site to site. Organic acid increases, observed in the UK and Scandinavia, are assumed here to have occurred independently of reductions in acid deposition. In the scheme given,  $X + Y + Z = \Delta ANC - A$ .

at lakes in the Bohemian Forest and Tatra Mountains (Kopáček, unpublished data) suggest that similar changes may have occurred here. Increases in organic acidity, while lowering pH, do not affect ANC and can in part, therefore, account for the weaker observed increases in pH.

The evidence from this study of recovery trends in freshwater chemistry and of the importance of base cation changes in regulating this recovery, largely supports that of previous European-scale assessments by Stoddard *et al.* (1999) and Skjelkvåle *et al.* (2000). Because the recovery trends continue through the 1990s, the longer records used here generally result in a more consistent and stronger pattern. This is particularly true for the UK, where both previous studies found little evidence of recovery; the new data included in this study suggest that  $x\text{SO}_4$  is declining and ANC and pH increasing, at rates comparable with those observed elsewhere in Europe. These results thus emphasise the value of sufficiently long datasets in distinguishing long-term changes from short-term (e.g. climatic) fluctuations. The finding of Stoddard *et al.* (1999) that European surface water trends shifted from falling base cation concentrations and largely stable ANC in the 1980s toward stable base cations and rising ANC in the 1990s, implying that base cation supplies might now exceed acid inputs, was not widely observed at sites in this study.

The results of this study also demonstrate that, whilst some general inferences can be drawn regarding the nature of chemical change, spatial heterogeneity cannot be ignored. For example, the four 'Central European' areas in this study (Northern Italy, Slovakia, the Czech Republic and Germany) have exhibited substantially differing trends over the last two decades despite their relative proximity. This spatial variability is by no means surprising given that the 23 sites in this region comprise both lakes and streams and a wide range of deposition, topography, vegetation and geological sensitivity, confirming the need for care when extrapolating from individual sites to regional and continental scales.

## Summary and conclusions

A number of general patterns of chemical change can be clearly identified from the 56 European long-term monitoring datasets analysed within this study:

- Non-marine  $\text{SO}_4$  concentrations have fallen at the majority of sites and in all regions. Surface waters throughout the studied areas of Europe have, therefore, clearly responded to reductions in S emissions since the 1980s. However Prechtel *et al.* (2001) show that reductions in surface water  $\text{SO}_4$  have often been

significantly smaller than those in deposition, due to the release of S stored in catchment soils.

- Falling surface water  $x\text{SO}_4$  has led to widespread improvements in ANC. Increases in ANC have commonly been smaller than decreases in  $x\text{SO}_4$ , however, due to compensating changes in other ions.
- Much of the imbalance between  $x\text{SO}_4$  reductions and ANC recovery can be explained by widespread decreases in Ca. The magnitude of Ca reductions and their impact on ANC, varies considerably between regions, for example having little impact in the Czech Republic, but severely limiting ANC recovery in Germany.
- Increases in pH, although largely corresponding to those in ANC, have tended to be somewhat weaker, since part of the ANC increase has been comprised of reductions in inorganic Al rather than in H ion. Increases in organic acidity, which do not influence ANC, have also suppressed pH recovery at a significant number of sites, particularly in the UK.
- Consistent trends were not observed for  $\text{NO}_3$ . Whilst concentrations have generally fallen in the Czech Republic, Slovakia and Germany, a substantial number of rising trends were observed in Italy and the UK. Overall, therefore, there is little evidence to suggest either a widespread response to falling N deposition, or a general increase in catchment N leakage.
- Few sites appear as clear outliers with respect to chemical trends. An exception is Markungsgraben, in Germany, where the elevated  $\text{NO}_3$  leaching and re-acidification caused by insect damage underlines the continued susceptibility of many sensitive ecosystems to biological or climatic 'shocks' under current levels of deposition loading.

Overall, the long-term monitoring data presented here provide clear evidence that reductions in the emission of acidifying pollutants during the last two decades have had a significant beneficial impact on water quality across a wide range of countries and ecosystems within Europe. These improvements in water quality nonetheless represent only a partial recovery of these systems, many of which remain acidic. Given negative feedback processes such as S adsorption/desorption and soil cation exchange, chemical recovery has not been instantaneous and it is likely that biological recovery will lag further behind improvements in water quality. Future emissions reductions agreed under the Gothenburg Protocol should increase both the magnitude and rapidity of change, but it may be many years before European surface waters recover fully from acidification.

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## Appendix I Sites used in European trend analysis

<i>Site</i>	<i>Country</i>	<i>Latitude</i>	<i>Longitude</i>	<i>Start date</i>	<i>End date</i>	<i>Type</i>	<i>Mean ANC (meq<sup>l</sup><sup>-1</sup>)</i>
1. Loch Coire nan Arr	UK (Scotland)	57° 25' N	5° 39' W	1988	2000	Lake	42
2. Allt a' Mharcaidh	UK (Scotland)	57° 07' N	3° 50' W	1988	2000	Stream	46
3. Allt na Coire nan Con	UK (Scotland)	56° 45' N	5° 36' W	1988	2000	Stream	35
4. Lochnagar	UK (Scotland)	56° 57' N	3° 13' W	1988	2000	Lake	3
5. Loch Chon	UK (Scotland)	56° 12' N	4° 32' W	1988	2000	Lake	23
6. Loch Tinker	UK (Scotland)	56° 13' N	4° 30' W	1988	2000	Lake	54
7. Round Loch of Glenhead	UK (Scotland)	55° 05' N	4° 25' W	1988	2000	Lake	-2
8. Loch Grannoch	UK (Scotland)	55° 00' N	4° 16' W	1988	2000	Lake	-27
9. Dargall Lane	UK (Scotland)	55° 04' N	4° 26' W	1988	2000	Stream	9
10. Scoat Tarn	UK (England)	54° 28' N	3° 17' W	1988	2000	Lake	-14
11. Burnmoor Tarn	UK (England)	54° 25' N	3° 15' W	1988	2000	Lake	49
12. River Etherow	UK (England)	53° 29' N	1° 49' W	1988	2000	Stream	31
13. Old Lodge	UK (England)	51° 02' N	0° 04' E	1988	2000	Stream	-17
14. Narrator Brook	UK (England)	50° 30' N	4° 01' W	1991	2000	Stream	18
15. Llyn Llagi	UK (Wales)	53° 00' N	4° 00' W	1988	2000	Lake	12
16. Llyn Cwm Mynach	UK (Wales)	53° 47' N	3° 57' W	1988	2000	Lake	9
17. Afon Hafren	UK (Wales)	52° 28' N	3° 42' W	1988	2000	Stream	2
18. Afon Gwy	UK (Wales)	52° 27' N	3° 42' W	1991	2000	Stream	14
19. Beaghs Burn	UK (N.Ireland)	55° 07' N	6° 05' W	1988	2000	Stream	92
20. Bencrom River	UK (N.Ireland)	54° 10' N	5° 56' W	1988	2000	Stream	3
21. Blue Lough	UK (N.Ireland)	54° 11' N	5° 54' W	1990	2000	Lake	-32
22. Coneyglen Burn	UK (N.Ireland)	54° 40' N	7° 03' W	1990	2000	Stream	168
23. Kaarvatn	Norway	62° 47' N	8° 53' E	1980	1999	Stream	26
24. Langtjern	Norway	60° 22' N	9° 39' E	1980	1999	Stream	22
25. Storgama	Norway	59° 01' N	8° 32' E	1980	1999	Stream	-23
26. Birkenes	Norway	58° 23' N	8° 15' E	1980	1999	Stream	-37
27. Ringsmöbäcken	Sweden	58° 59' N	11° 45' E	1985	1999	Stream	8
28. Lommabäcken	Sweden	58° 42' N	14° 38' E	1985	1999	Stream	-6
29. Bratångsbäcken	Sweden	58° 42' N	14° 38' E	1985	1999	Stream	39
30. Gårdsjön F1	Sweden	58° 03' N	12° 01' E	1980	1999	Stream	-111
31. Pipbäcken	Sweden	57° 03' N	12° 47' E	1985	1999	Stream	1
32. Valkeakotinen	Finland	61° 14' N	25° 04' E	1988	1999	Stream	71
33. Hietajärvi	Finland	63° 10' N	30° 43' E	1988	1999	Stream	105
34. Lange Bramke	Germany	51° 52' N	10° 26' E	1980	1999	Stream	-67
35. Dicke Bramke	Germany	51° 52' N	10° 26' E	1980	1999	Stream	64
36. Metzenbach	Germany	49° 54' N	9° 26' E	1987	1999	Stream	97
37. Lehstenbach	Germany	50° 09' N	11° 52' E	1987	1999	Stream	-36
38. Schluchsee	Germany	47° 49' N	8° 06' E	1987	1996	Stream	33
39. Villingen	Germany	48° 03' N	8° 22' E	1987	1996	Stream	15
40. Markungsgraben	Germany	48° 57' N	13° 25' E	1987	1999	Stream	10
41. ěerné	Czech R.	49° 11' N	13° 11' E	1984	1999	Lake	-16
42. ěertovo	Czech R.	49° 10' N	13° 12' E	1984	1999	Lake	-38
43. Prašilské	Czech R.	49° 05' N	13° 23' E	1984	1999	Lake	-17
44. Zdarské	Czech R.	48° 56' N	13° 39' E	1984	1999	Lake	73
45. Laka	Czech R.	49° 07' N	13° 20' E	1984	1999	Lake	16
46. Plešné	Czech R.	48° 47' N	13° 52' E	1984	1999	Lake	-10
47. Slavkovské	Slovakia	49° 09' N	20° 11' E	1984	1999	Lake	-9
48. Batizovské	Slovakia	49° 09' N	20° 07' E	1982	1999	Lake	22
49. Vyšné Wahlenbergovo	Slovakia	49° 10' N	20° 01' E	1980	1999	Lake	9
50. Malé Hincovo	Slovakia	49° 10' N	20° 03' E	1984	1999	Lake	266
51. Jamské	Slovakia	49° 08' N	20° 00' E	1984	1999	Lake	-17
52. Lago Paoine Superiore	Italy	46° 11' N	8° 11' E	1980	1999	Lake	4
53. Lago Paoine Inferiore	Italy	46° 10' N	8° 11' E	1980	1999	Lake	29
54. Pellesino	Italy	45° 47' N	8° 04' E	1986	1999	Stream	173
55. Pellino	Italy	45° 48' N	8° 04' E	1984	1999	Stream	199
56. Cannobino	Italy	46° 04' N	8° 42' E	1980	1999	Stream	216



