

# Convention on Long-Range Transboundary Air Pollution

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



ICP Waters Report 92/2008

Proceedings of the 23<sup>rd</sup> meeting  
of the ICP Waters Programme Task  
Force in Nancy, France

October 8 – 10, 2007



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CONVENTION ON LONG-RANGE  
TRANSBOUNDARY AIR POLLUTION

INTERNATIONAL COOPERATIVE PROGRAMME ON  
ASSESSMENT AND MONITORING OF ACIDIFICATION  
OF RIVERS AND LAKES

Proceedings of the 23<sup>rd</sup> meeting of the ICP Waters  
Programme Task Force in Nancy, France,  
October 8 - 10, 2007

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Oslo, February 2008



## Preface

The International Cooperative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes (ICP Waters) was established under the Executive Body of the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP) in July 1985. Since then ICP Waters has been an important contributor to document the effects of implementing the Protocols under the Convention. Numerous assessments, workshops, reports and publications covering the effects of long-range transported air pollution has been published over the years.

The ICP Waters Programme Center is hosted by Norwegian Institute for Water Research (NIVA), while the Norwegian Pollution Control Authority (SFT) leads the programme. The Programme Centres work is supported financially by SFT.

The main aim of the ICP Waters Programme is to assess, on a regional basis, the degree and geographical extent of the impact of atmospheric pollution, in particular acidification, on surface waters. More than 20 countries in Europe and North America participate in the programme on a regular basis.

ICP Waters is based on existing surface water monitoring programmes in the participating countries, implemented by voluntary contributions. The ICP site network is geographically extensive and includes long-term data series (more than 20 years) for many sites. The programme yearly conducts chemical and biological intercalibrations.

At the annual Programme Task Force, national ongoing activities in many countries are presented. This report presents national contributions from the 23<sup>rd</sup> Task Force meeting of the ICP Waters programme, held in Nancy, France, October 8 - 10, 2007.



Brit Lisa Skjelkvåle  
ICP Waters Programme Centre

*Oslo, February 2008*

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# 1. Introduction

The International Cooperative Programme on Assessment and Monitoring of Rivers and Lakes (ICP Waters) is a programme under the Executive Body of the Convention on Long-Range Transboundary Air Pollution. The main aims of the programme are:

- To assess the degree and geographic extent of the impact of atmospheric pollution, in particular acidification, on surface waters;
- To collect information to evaluate dose/response relationships;
- To describe and evaluate long-term trends and variation in aquatic chemistry and biota attributable to atmospheric pollution.

The national contributions on ongoing activities that were presented during the ICP-Waters Task Force meeting in Nancy, France in October 2007 are grouped thematically;

- i. Water chemistry – trends and status for S and N
- ii. Biological response
- iii. Heavy metals and POP's
- iv. Dynamic modelling / Critical loads

Under 'Water chemistry – trends and status of S and N', data on lake acidification from Swedish lake surveys from 1972 until now were presented (Chapter 2), in addition to water chemistry monitoring data from two Bulgarian catchments (Chapter 3).

Under the theme biological response, the UK results from the Acid Monitoring Network on chemical and biological recovery is presented (Chapter 4).

Findings from fish studies in the Central Asian countries Kazakhstan and Kyrgystan, where radioactivity and heavy metals is a serious environmental problem, was presented under the heading 'Heavy metals and POPs' (Chapter 5).

Under the theme Dynamic modelling /critical loads, modelling the effect of climate change on acidification recovery in Lake Paione Superiore in the Central Alps, Italy is adressed (Chapter 6).

Reports and publications from the ICP-Waters programme are given in Chapter 7.



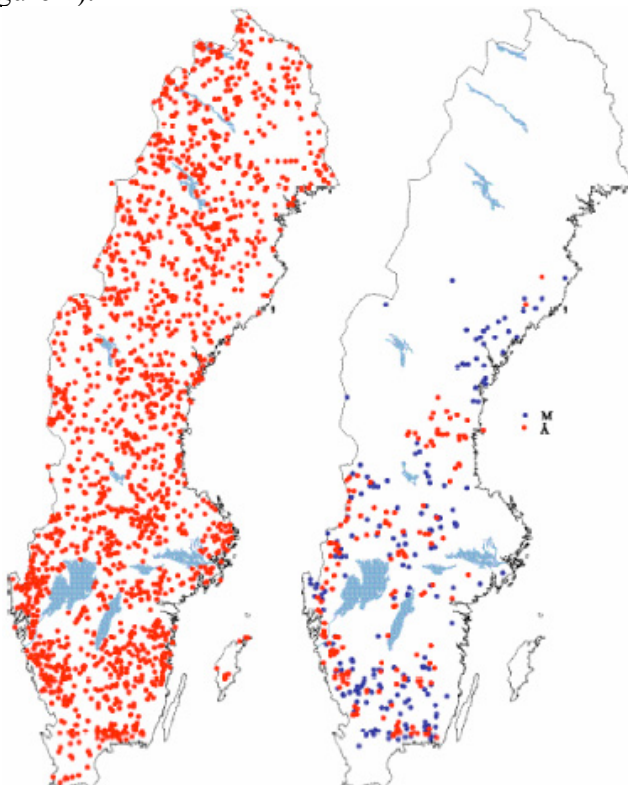
## 2. Swedish lake surveys: What do they tell about acidification?

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### 2.1 Introduction

In Sweden from 1972 until now, 8 lake surveys have been performed, with different motives and in various shapes. The latest three surveys were based on the joint work to perform a common Nordic lake survey in 1995 (Henriksen et al. 1998). Because the objective was to study the extent of acidification, a protocol dealing with how to randomly select lakes and perform the analysis was prepared. The selection was randomly stratified with respect to lake size (weighted towards a larger portion of larger lakes) and county (weighted with respect to acid deposition). For all three countries the basis was lake registers and thus after destratification the results were able to represent the conditions in the country. Russia also participated with a lake survey in the Kola Peninsula and parts close to the Finnish border (Skjelkvåle *et al.* 2001). Sampling was done in autumn during the circulation in order to obtain samples as representative as possible to the annual water chemistry conditions. Lake surveys in 2000 and 2005 in Sweden used the same protocol. However, the 1995 protocol was limited to lakes larger than 0.04 km<sup>2</sup>. Since nearly half the Swedish lakes have an area less than that, for the 2005 survey it was decided to randomly select a portion of small lakes (Figure 1).



Sampled lakes: 1709    Sampled limed lakes: 195 + 165

**Figure 1.** Location of sampled unlimed and limed lakes.

The Swedish population of unlimed lakes, with an area above 0,01 km<sup>2</sup>, is about 85000.

Limed lakes also have a lake register, and the population of limed lakes is:

Target lakes; 2943

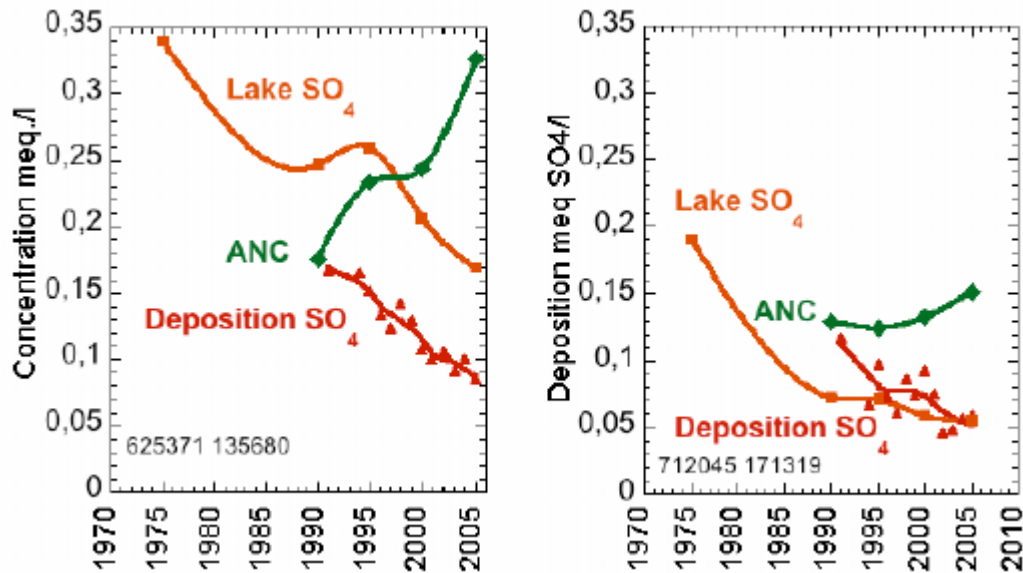
Storage lakes; 3243

*Target lakes* are limed for their own protection against acidification, while *storage lakes* are limed as reservoirs, to protect downstream rivers or lakes.

For each size class and county we know the number of lakes sampled and the population, thus by destratification we can estimate the status of the Swedish lake population.

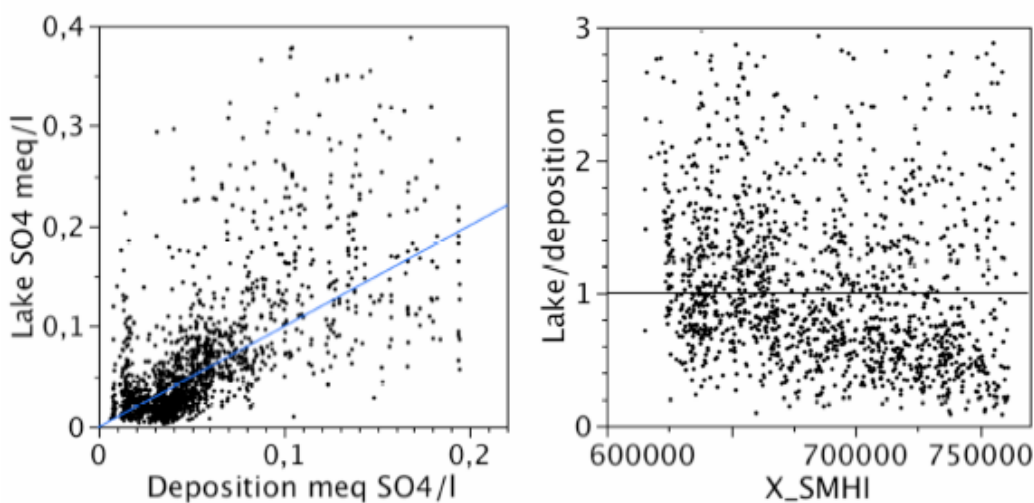
## 2.2 Lake sulphate concentrations depend on deposition but also on soil conditions

In general lake sulphate concentrations reflect deposition concentrations (adjusted for evapotranspiration), as is the case for two lakes sampled at several lake surveys (Figure 2).



**Figure 2.** Relationship between sulphate concentrations in deposition and lake. Also development in ANC is shown. Lakes sampled at several lake surveys. Deposition obtained from modelled data (MATCH of Persson et al. 1996).

The southern lake (left in Figure 2) has a concentration higher than the deposition, while the northern lake (right) indicate a balance between deposition and lake concentration. It is well known that inorganic adsorption/desorption as well as the more dominant binding of sulphur in organic matter take place. The surveyed lakes indicate storage of sulphur at low deposition, which is governing in the north, and more common losses from the drainage areas at higher deposition in the south (Figure 3).

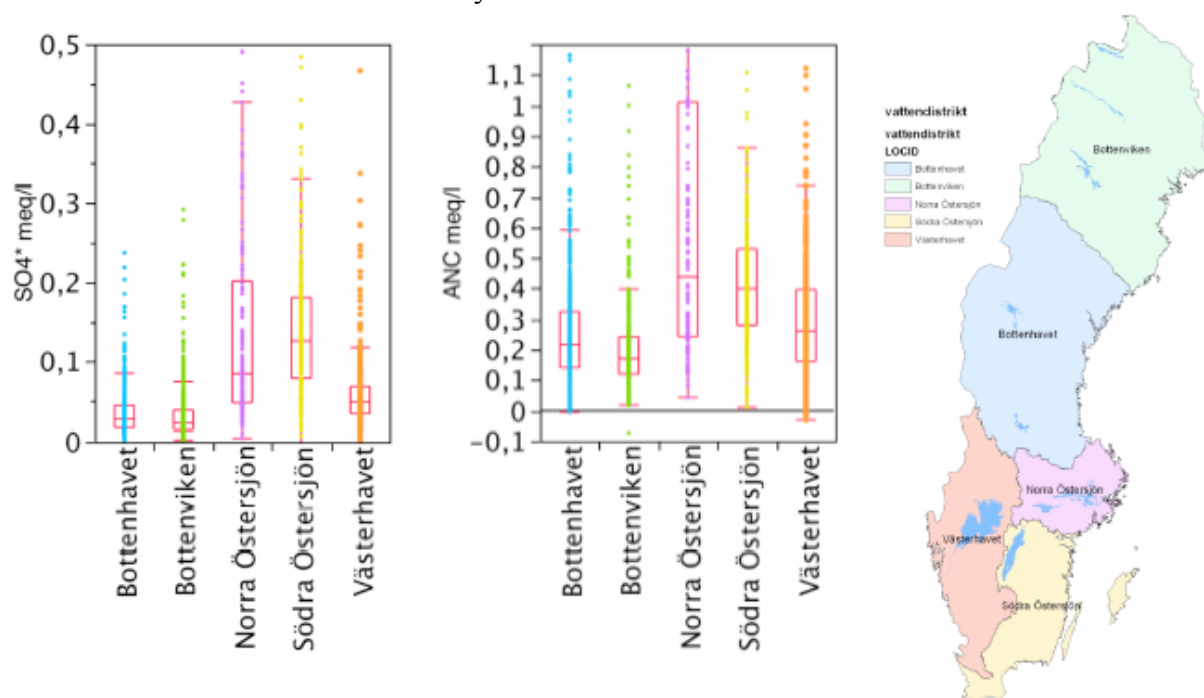


**Figure 3.** The relationship between deposition of sulphate (total) during 2005 and lake concentrations. Right Quotient lake/deposition concentrations along a gradient from south to north. The vertical line indicates equilibrium between deposition and lake concentration.

## 2.3 Status and trends for surveyed lakes

Highest sulphate concentrations are found in south-eastern Sweden (“Östersjön”), caused by relatively high deposition, and concentrated through high evapotranspiration (Figure 4). The western part has the lowest ANC values as extremes, caused by acid deposition.

135 lakes have been sampled 7 – 8 times since 1972, and trends for these were calculated using Sen’s slope (Table 1). In all lakes the sulphate concentration has decreased. In the non-limed lakes alkalinity increased as a median with 0.005 meq/L/year. About two thirds of the lakes showed an increase in alkalinity.



**Figure 4.** Concentrations of non-marine sulphate and ANC for five water districts.

**Table 1.** Trends for non-marine sulphate and alkalinity for surveyed lakes. (n= 135).

Parameter	Bottenviken and Bottenhavet	Norra and södra Östersjön	Västerhavet
Sulphate non-marine	-0,007	-0,031	-0,037
Alkalinity	-0,001	0,005	0,010

## 2.4 Limed lakes play an important role when acidification is evaluated

In Sweden about 6000 lakes are limed as a remedy for acidification. These lakes constitute a large part of the acidified lakes and have to be treated separately. The effect of liming was adjusted for by the use of a Ca\*/Mg\* for nearby non-limed lakes and the Mg\* concentration in the limed lake (Göransson *et al.* 2005).

## 2.5 Present level of acidification

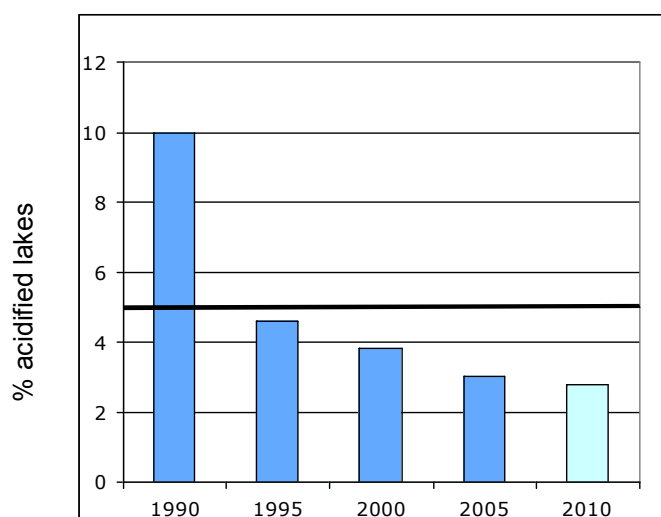
New water quality criteria for acidification are based on a library of MAGIC modelled lakes (calculations made by IVL, Gothenburg). The “most similar” lake is selected as equal and a pre-industrial pH-value is calculated. Also a present pH-value is calculated using today’s ANC and TOC. A delta-pH  $\geq 0.4$  units is taken as the limit for an acidified lake. The result for

each evaluated lake is destratified with respect to the entire lake population (Table 2). It presents data for all acidified lakes and for non-limed lakes in the Swedish lake population.

**Table 2.** Percentage of acidified lakes in each lake size. Calculated according to Swedish water quality criteria.

Lake area km <sup>2</sup>	Acidified lakes Incl. limed	% Acidified lakes Excl. limed
>100	0.0	0.0
10-100	8.1	0.0
1-10	12.3	0.2
0.1-1	13.5	3.7
0.04-0.1	17.0	8.9
0.01-0.04	21.0	17.3

The largest portion of acidified lakes is found in the lake size 0.01–0.04 km<sup>2</sup> that was studied for the first time in the 2005 survey. The acidification of Swedish lakes can be followed using lake surveys (Figure 5). Reduced sulphate deposition has led to a substantial reduction in the number of acidified lakes in Sweden, but still a number of lakes need liming in order to mitigate acid deposition.



**Figure 5.** Development of acidified lakes. Lake surveys 1990, 1995, 2000 and 2005 and prognosis for 2010. The Swedish environmental goal of 5 % acidified lakes is shown (area > 0.04 km<sup>2</sup>).

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### **3. Hydrochemical Monitoring in Forested Catchments in Bulgaria for Assessment of the Risk of Stream Water Acidification**

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#### **Abstract**

Acidifying pollutants and base cation concentrations in bulk precipitation, throughfall and stream water have been monitored for 22 years in two forested catchments in Bulgaria in order to document effects of atmospheric deposition on surface waters. It was found that the concentration of acidifying sulfates was lower in the stream water than in throughfall, and the concentration of the neutralizing calcium higher. This contributed to neutral pH-values in the surface waters. Critical loads of acidity, sulfur and nitrogen have been used to identify the sensitivity of soils and water for acidification. The results obtained in this study were then compared with similar parameters from an acid stream in the Vosges Mountains (France). It was demonstrated that the sulfate:calcium ratio is lower in stream water than in throughfall in unacidified catchments, while it is the contrary in acidified catchments. Critical loads of acidity, sulfur and nitrogen were higher and were not exceeded in neutral stream water in Bulgaria contrary to the acid stream in the Vosges Mountains.

*Key words: Hydrochemical monitoring, acidification and critical loads of acidifying pollutants*

#### **Introduction**

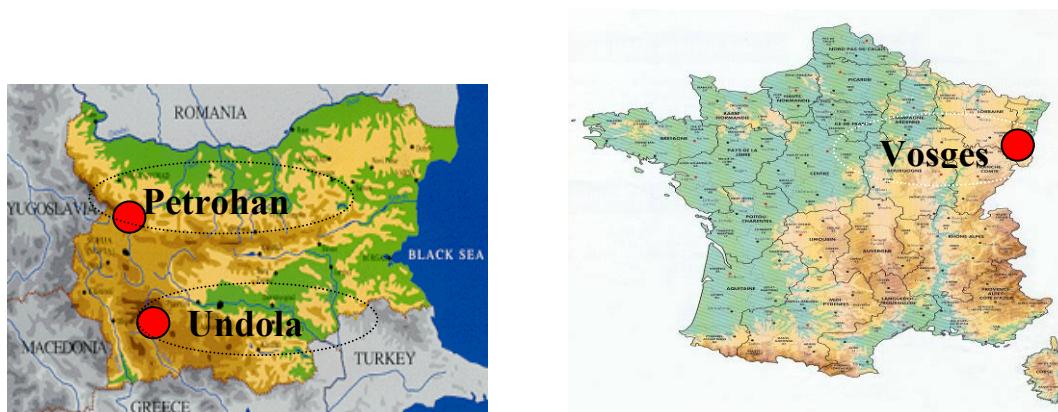
Although the atmospheric pollution has decreased in Europe in the last 20 years and catchment monitoring studies have shown that the decrease of atmospheric deposition has led to a decrease in sulfate and base cation concentrations in streams (Probst et al., 1992; Moldan et al., 2001; Schopp et al., 2003), there is an agreement that there still are many acidified streams in Europe. In addition, the acidity of some stream waters that originate from catchments situated closely to the acid ones remains neutral. It means that the stream water acidification depends not only on acid depositions but also on the background substrate, the buffering capacity of the soils in the catchment area, the vegetation cover, the climate etc. The concentration of base cations can also give an indication of the acid sensitivity of the surface water. To evaluate anthropogenic acidification of surface water, the water chemistry must be evaluated together with deposition input. The relationship between the risk of water acidification and pollution, and the environmental conditions is illustrated by the critical loads of acidifying pollutants and their exceedances by the real depositions. The main objective of this study is to carry out longtime hydrochemical investigations in forested catchments in order to determine the ratio between the acidifying and neutralizing ions in precipitation and surface water. The critical loads of acidity, sulfur and nitrogen will be determined as well.



Critical loads are thresholds of air polluting compounds which should not be exceeded in order to protect aquatic ecosystems from the risk of acidification.

## Materials and Methods

The evolution of the chemical composition of two streams has been studied the last 22 years (1986-2007) in response to changes in acid deposition. One of the catchment areas (Undola site) is situated in the Southwestern part of Bulgaria at 1500 m.a.s.l. on granite and Dystric Euthric Cambisols with 596 mm of precipitation and a Spruce, Fir and Pine forest cover. The second one (Petrohan site) is situated in the Northwestern part of the country, and differs from the first site only by the amount of precipitation (1026 mm) and the type of forest cover (mainly beech).



**Figure 1** Location map of sites for the longtime hydrochemical monitoring in forested catchments in Bulgaria (Petrohan and Undola), compared with an acid stream in the Vosges Mountains in France

Sampling of bulk precipitation, throughfall and stream water has been performed every 14 days, and pH,  $K^+$ ,  $Na^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $NH_4^+$ ,  $NO_3^-$ ,  $SO_4^{2-}$  and  $Cl^-$  was analyzed. The first results showed a dramatic increase for the period 1988-1992 in both acidifying anions and base cations in depositions and stream water for the Undola and Petrohan sites. This is probably due to the lack of investments for reducing emissions of pollutants to the atmosphere at during the beginning of the economic changes in Bulgaria. However, the values of pH in stream water remained neutral (Ignatova, 1994, 1995 Ignatova & Dambrine, 1996). In addition the ratio between  $SO_4^{2-}$  and  $Ca^{2+}$  concentrations in all water compounds has been calculated. The effect based Steady-state mass balance model was used to calculate the critical loads of acidity C L (A), sulfur C L max (S) and nitrogen [C L max (N), C L nut (N)], according to the follow equations:

$$\begin{aligned}
 C L (A) &= B C w + Q [ H ] \text{ crit} + R Al/Ca ( B C \text{ dep} + B C w - B C u) \\
 &= 2.5 B C w + 0.09 Q + 1.5 B C \text{ dep} - 1.5 B C u \\
 C L \text{ max} (S) &= C L (A) + B C \text{ dep} - B C u \\
 C L \text{ max} (N) &= C L \text{ min} (N) + C L \text{ max} (S) \\
 C L \text{ nut} (N) &= N u + N i + N \text{ le} (\text{crit})
 \end{aligned}$$

*Exceedances of critical loads* of acidity Ex (A), sulfur Ex (S) and nitrogen Ex (N) by present depositions were defined as follows:

$$\text{Ex (A)} = \text{P L (S)} + \text{P L (N)} - \text{B C dep} - \text{B C u} - \text{N u} - \text{C L (A)}$$

$$\text{Ex (S)} = \text{P L (S)} - \text{Sf (B C u} - \text{B C dep)} - \text{C L (S)}$$

$$\text{Ex (N)} = \text{P L (N)} + (1 - \text{Sf}) (\text{B C u} - \text{B C dep}) - \text{C L (N)}$$

where:

B C w - input of base cations by weathering;

B C dep - input of base cations by deposition;

B C u and N u - uptake of base cations and nitrogen by the biomass;

N i - nitrogen immobilization;

N le (crit)- nitrogen leaching ;

$$\text{Sf} = \text{P L (S)} / [\text{P L (S)} + \text{P L (N)} - \text{N u} - \text{N i}];$$

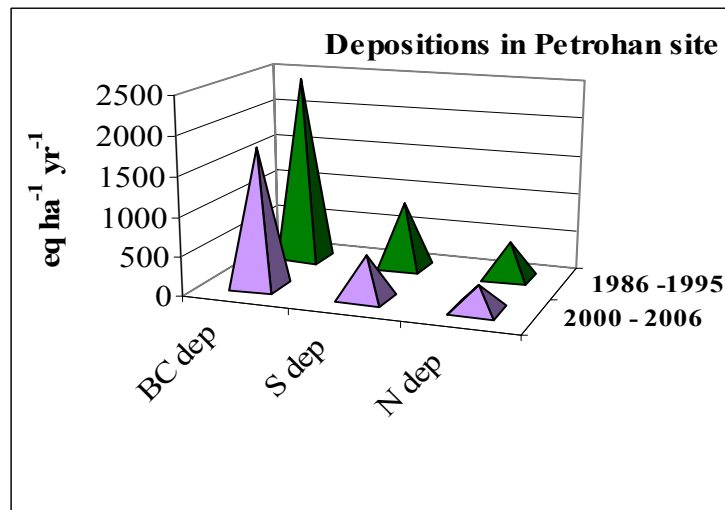
$$\text{P L (S)} = \text{P L (S-SO}_2) + \text{P L (S-SO}_4^{2-})$$

$$\text{P L (N)} = \text{P L (N-NO}_2) + \text{P L (N-NO}_3^-) + \text{N-NH}_4^+$$

In the absence of acid streams in Bulgarian forested areas, the results obtained in this study have been compared with data from acid stream water (pH 4.5) from the Vosges mountains, France, produced on quartzitic sandstone and acid soils (Angeli et al., in press; Probst et al., 2005; Posch et al., 2005).

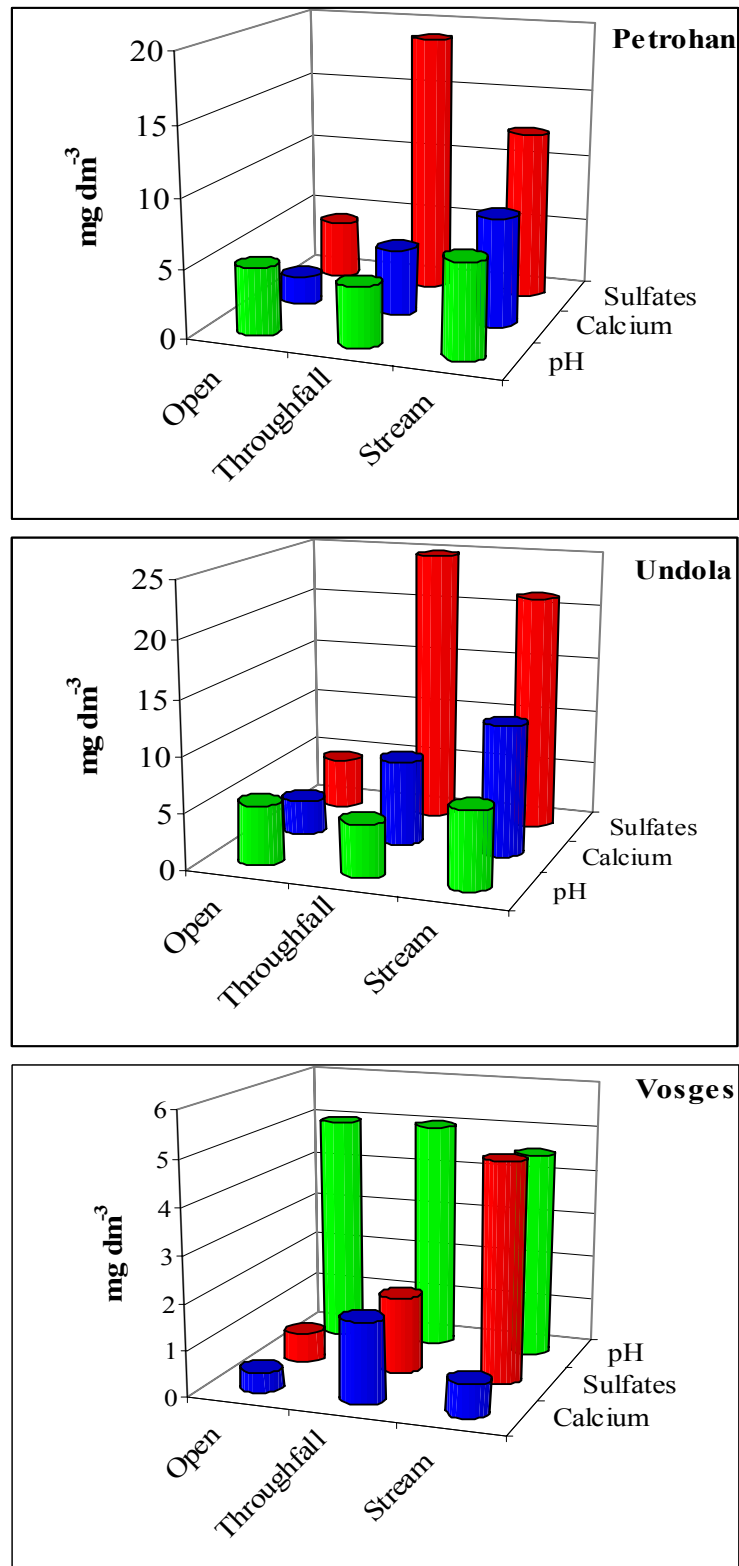
## Results and Discussion

Comparing the deposition data for the period 1986-1995 with those from 2000- 2006 it can be seen that there is a significant decrease in the deposition of both acidifying anions and neutralizing base cations in the Bulgarian sites (Figure 2), keeping the neutral values of pH for surface waters. Although a similar decrease in deposition can be seen in Western Europe (Hettelingh et al., 2004; Probst et al., 2005; Posch et al., 2005) there are still acid streams in the Scandinavian countries, Canada and Western Europe. This also includes the Vosges Mountains, where the source, stream and surface water are more acid than the bulk precipitation and throughfall. The concentration of acidifying sulfates in the precipitation and throughfall in the Vosges catchment is lower than in the Bulgarian sites. However, the stream water is much more acid in the Vosges catchment, not only compared to the Bulgarian streams but also compared to the bulk precipitation and throughfall. On the contrary, in both Bulgarian sites (Petrohan and Undola) the stream water pH is higher than the bulk precipitation and throughfall (Figure 3). This means that the actual acidification of the stream water is not only related to the acid deposition. Comparing the concentrations in Figure 3 it can be seen that the concentration of calcium is much higher in the stream water than in throughfall in the Petrohan and Undola sites, while the stream water in Vosges has only low concentrations of Ca. At the same time the concentration of acidifying sulfates is more than twice as high in the stream water than in the throughfall in the Vosges catchment, this is however not the case in the Bulgarian sites. This indicates that the stream water acidification depends not only on the acidifying pollutant concentration, but also on their ratio with base cations such as  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{K}^+$ . The ratio between the concentration of sulfates and calcium in precipitation and stream water (Figure 4) shows that this ratio is lower in stream water than in throughfall in both the Petrohan and the Undala site. However, the sulfate to calcium ratio of the stream water in the Vosges Mountains is about 5 times as high as in the open field and throughfall precipitation (Figure 4).

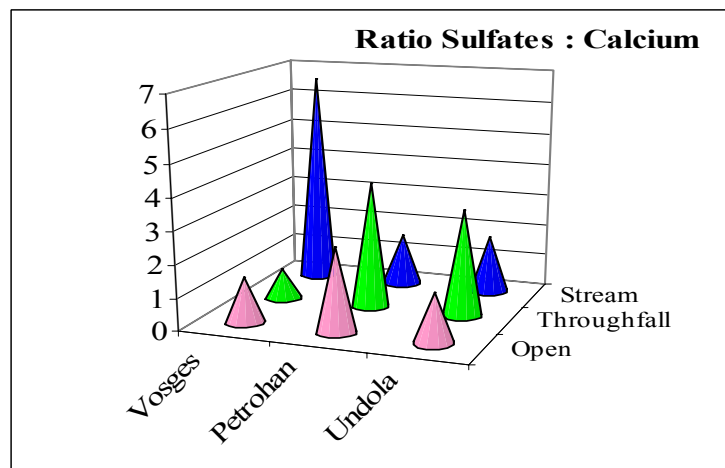


**Figure 2** Deposition of base cations and acidifying sulfur and nitrogen in the forested catchment of the Petrohan site, eq ha<sup>-1</sup> yr<sup>-1</sup>.

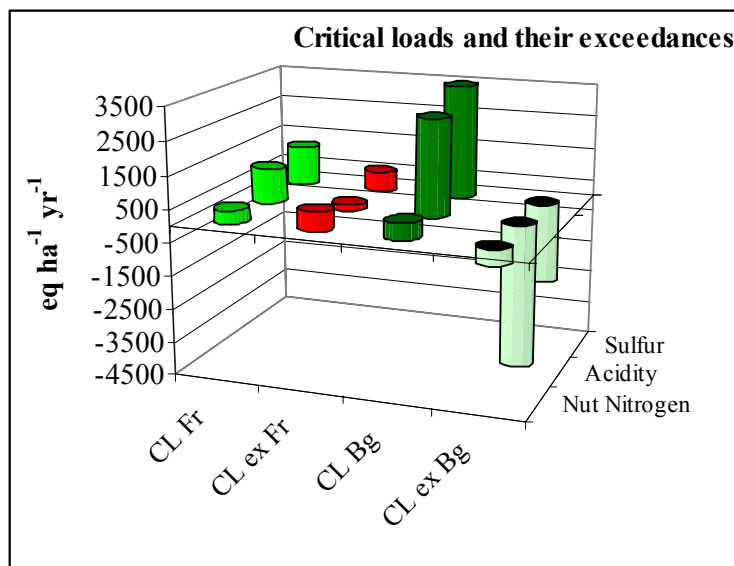
In general the sulfate to calcium ratios in the bulk precipitation in the Vosges Mountains and the Bulgarian sites are comparable, however, they are quite different when stream water is concerned. That illustrates the role of the catchment soils in neutralizing acid deposition. Critical loads of acidifying pollutants (acidity, sulfur and nitrogen) have been computed to assess the risk of stream water acidification (Figure 5.). The critical loads of acidity, sulfur and nitrogen are high (2500- 3500 eq ha<sup>-1</sup> yr<sup>-1</sup>) for the Bulgarian catchments, illustrating a high buffering capacity of their soils and water. The critical loads are not exceeded by the actual deposition of acidifying pollutants in the Undola and Petrohan monitoring plots, where the stream water chemistry is neutral. Critical loads of acidity and sulfur in the Vosges Mountains are about 1000 eq ha<sup>-1</sup> yr<sup>-1</sup>, much lower than for the Bulgarian sites, showing that the soil and water here have a much higher sensitivity to acidification. More importantly, the critical loads of acidifying pollutants are exceeded in the Vosges site.



**Figure 3.** Concentration of Calcium, Sulfates and pH of open field precipitation, throughfall and stream water in the Petrohan (top) and Undola (middle) sites in Bulgaria for the period 1994-2005 and the Vosges site (bottom) for the period 2002-2005 (Angeli et al., in press)



**Figure 4.** The ratio of sulfate to calcium in open field precipitation, throughfall and stream water in sites of Vosges (acid stream), Petrohan and Undola (neutral streams).



**Figure 5.** Critical loads (CL) of Sulfur, Acidity and Nutrient Nitrogen and their exceedances (CL ex) for acid (Fr) and neutral (Bg) streams

## Conclusion

1. By means of long term hydrochemical monitoring it has been shown that the surface stream water in two Bulgarian sites has remained neutral due mainly to the high concentration of base cations.
2. The sulfate to calcium ratio in precipitation and stream water is a good indicator for assessing the risk of surface water acidification.

3. Critical loads of acidifying pollutants and their exceedances by acid deposition can be successfully used to evaluate the risk of stream water acidification in forested catchments.
4. The critical loads of acidity, sulfur and nitrogen are high for both Bulgarian catchments studied, showing a good buffering capacity. Critical loads of deposition are not exceeded by the actual deposition in the Yundola and Petrohan monitoring plots, where the stream water chemistry is neutral.
5. In cases where the sulfate to calcium ratio in stream water is high, and the critical loads of acidifying pollutants are exceeded, the buffering capacity of the water and soils can be increased by liming the whole catchment area.

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## 4. An update on the state of chemical and biological recovery in the UK (1988-2006)

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The UK Acid Waters Monitoring Network (AWMN) was established in 1988 by the UK Department of the Environment to determine the effects of acid emission controls on acid sensitive surface waters. For most of its operation it has comprised 22 lakes and streams that have been monitored for water chemistry and a range of freshwater biological groups (i.e. epilithic diatoms, aquatic macrophytes, macroinvertebrates and fish. Diatom monitoring also includes the assessment of the species composition of lake sediment collected annually in traps that can be compared directly with historical changes recorded in sediment cores from these sites. In more recent years monitoring has been extended to include the assessment of carbonaceous particles and metals (including, uniquely, mercury) in lake sediment and mosses in AWMN catchments, and lake water temperature. At Lochnagar the metals sampling programme has been enhanced to cover deposition and various components of the biota.

A recent study of AWMN data, published as a Special Issue of the journal *Environmental Pollution* (See Monteith and Evans, 2005), covering the period 1988-2003, provided the first evidence for a national reduction in the non-marine sulphate ( $xSO_4$ ) concentration of UK surface waters. Concentrations fell sharply between 1996 and 2000, in line with similar reductions in  $xSO_4$  in deposition over the same period. Reductions were largest in southeast and central England and declined in strength along a northwesterly gradient. In the far northwest of Scotland and Northern Ireland the reductions were very slight, but here  $xSO_4$  deposition has always been very low and sites here have not acidified. There was no evidence for any regional decline in the concentration of nitrate ( $NO_3$ ). Water acidity, as reflected by pH, alkalinity and labile aluminium concentration, was found to have declined in proportion to the decline in  $xSO_4$  and links were demonstrated between chemically recovering sites and those where acid-sensitive plant and animal species were returning and/or becoming more abundant.

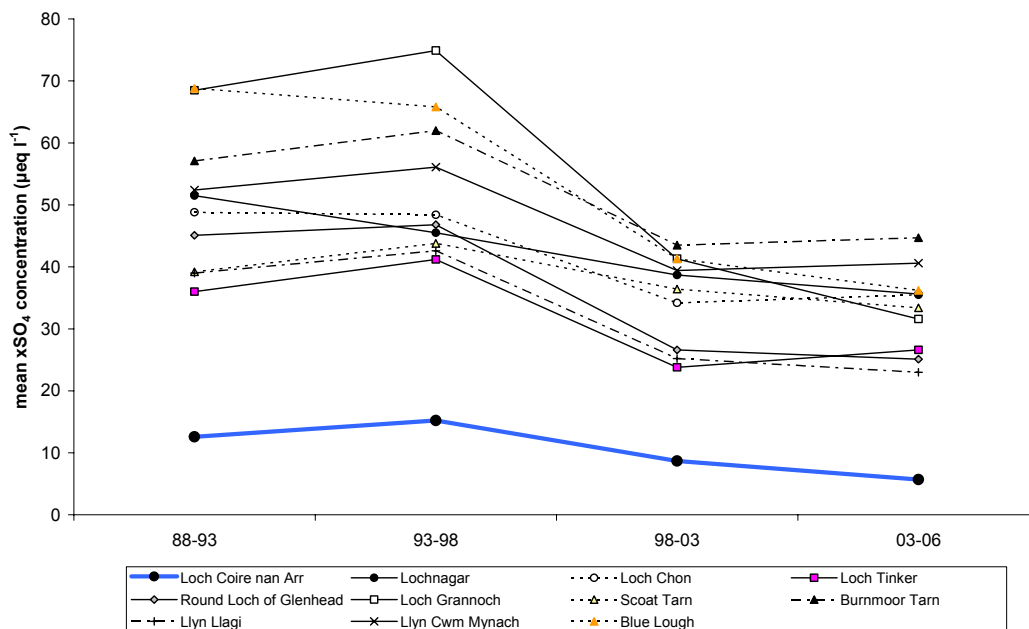
In our most recent assessment, covering the period 1988-2006, we found that recovery has stalled since around the turn of the century. Non-marine sulphate concentrations in most sites reached minimum levels around the year 2000 and have since stabilised or, in a minority of sites, have increased slightly (Figure 1). Over the period 2003-2006 the majority of AWMN lakes and streams showed mean  $xSO_4$  concentrations between 4 and 7 times that of the low deposition control lake in northwest Scotland (Loch Coire nan Arr). Concentrations in Old Lodge (southeast England) and the River Etherow (southern Pennines) were 23 and 29 times higher respectively.

Most sites continued to show no long-term trend in  $NO_3$  concentration and year to year variability in peak concentrations continued to be dominated by variability in climate (cf. Monteith et al., 2000). However, a minority of sites showed significant increases in



concentrations over the full 18 year period. These trends are consistent with the “nitrogen saturation” model that assumes that pollutant nitrogen will accumulate in soils to a point where it can no longer be retained by biological cycling and starts to “leak” into surface waters (cf. Stoddard, 1995). Alternatively, it is feasible that the relatively low concentrations of  $\text{NO}_3$  in these sites at the onset of monitoring were due to a climatic anomaly at the time (i.e. a series of unusually warm winters). These increases are of concern as they potentially threaten improvements in acidity gained by the reduction in  $\text{xSO}_4$ . Further monitoring and analysis is required to determine the drivers and the likely spatial extent of these changes in other surface waters. As  $\text{xSO}_4$  deposition falls and  $\text{NO}_3$  concentrations remain stable (or increase)  $\text{NO}_3$  is becoming increasingly important in slowing further recovery. At Scoat Tarn, in the English Lake District,  $\text{NO}_3$  is occasionally as important an acidifying agent (i.e. it reaches similar concentrations) as  $\text{SO}_4$ .

Over the full period labile aluminium ( $\text{Al}_{\text{lab}}$ ) concentrations have fallen markedly at several of the more acidified sites on the AWMN. In some cases mean levels have fallen below those believed to be toxic for fish. However, approximately half of all AWMN sites showed mean concentrations above a theoretical lower toxicity limit of  $25 \mu\text{g l}^{-1}$  between 2003-2006. Labile aluminium concentrations of other sites rise above this limit occasionally and this could still result in limitation of recovery of aquatic biota by episodic extremes. Hydrogen ion ( $\text{H}^+$ ) concentration has declined in several AWMN lakes. There is less evidence for improvements in mean  $\text{H}^+$  in AWMN streams, but most show a progressive decline in  $\text{H}^+$  maxima (i.e. pH minima) over the full record and this should be biologically beneficial.

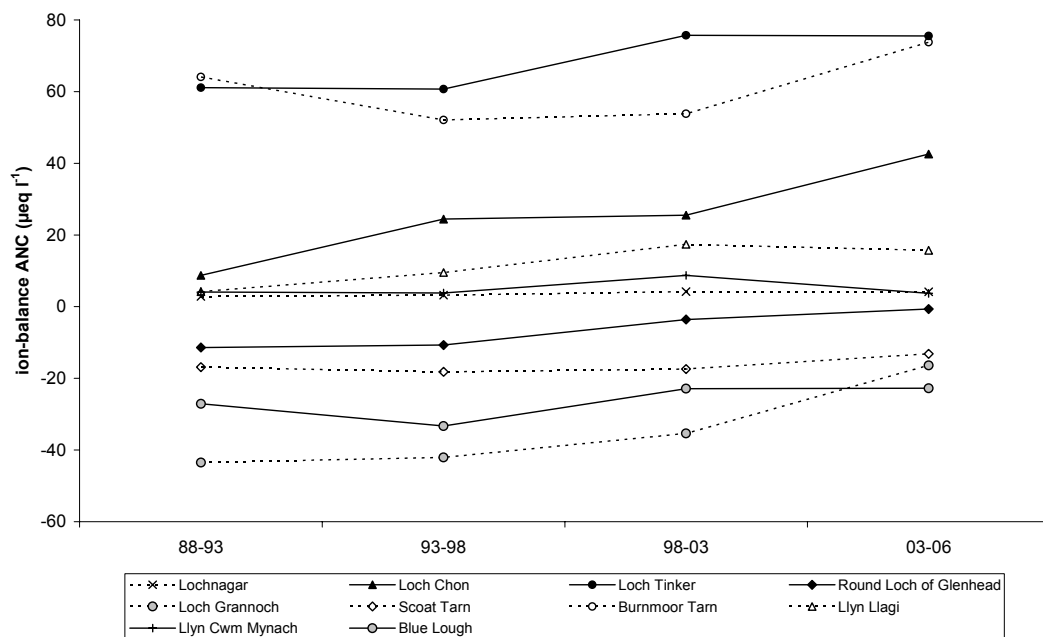


**Figure 1.** Five year mean concentrations of non-marine sulphate in UK AWMN lakes. The thick blue line represents the low deposition (control) site in north-west Scotland, Loch Coire nan Arr. Note last period is four years only.

Acid neutralising capacity (ANC – as determined by the difference in the sum of base cations and the sum of acid anions in terms of equivalence) has increased at several sites where  $x\text{SO}_4$  concentrations have fallen. UK government emissions policy (based on the concept of Critical Loads for Acidity) is based on the premise that an ANC of  $20 \mu\text{eq l}^{-1}$  (i.e.  $20 \mu\text{eq l}^{-1}$  higher base cation than acid anion concentration) provides protection for aquatic biota against the effects of acid deposition. However, between 2003-2006 only four AWMN lakes had mean ANC greater than  $20 \mu\text{eq l}^{-1}$ , and three of these are unlikely to have ever acidified below this level historically. Scoat Tarn, the Round Loch of Glenhead, Loch Grannoch and Blue Lough still show mean ANC of less than zero. If these sites are representative of their heavily acidified sub-regions in the Lake District, Galloway and Mourne Mountains, the implication is that the target of  $20 \mu\text{eq l}^{-1}$  has far from been met in these areas by the reductions in sulphate deposition achieved to date (Figure 2).

The 15 year interpretative exercise demonstrated a large national increase in concentrations of dissolved organic carbon (DOC) in common with many other monitored regions in northern Europe and North America (Evans et al., 2005; Skjelkvåle et al., 2005). Concentrations have stabilised over the past 3 years. Given that there has been no change in  $x\text{SO}_4$  at most sites, this observation is consistent with the hypothesis that the acidity and/or ionic strength of deposition exerts a dominant control on the solubility of the soil organic matter. The hypothesis has recently been tested and supported using an international dataset developed under the auspices of the ICP Waters Programme (see Monteith et al., 2007).

The 2003-2006 period has been unusual due to the relative absence of winter storms and accompanying seasalt deposition events that cause temporary elevations in  $\text{H}^+$  and  $\text{Al}_{\text{lab}}$  concentration (as a result of the temporary displacement of these acid cations by marine base cations from soils). However, in the longer term (i.e. since the 1970s), there has been a tendency for increased storminess, and this has been linked elsewhere to global warming. It is likely that the prolonged periods of seasalt deposition (reflected by very high chloride concentrations) experienced during the early 1990s will eventually recur. At current acid deposition levels this is likely to bring about the return of higher  $\text{Al}_{\text{lab}}$  and  $\text{H}^+$  concentrations in the more acidified sites and could retard or even reverse biological recovery. In several forested AWMN catchments particularly, these seasalt episodes are sufficient to raise  $\text{Al}_{\text{lab}}$  concentrations and depress pH to levels that are known to be toxic to fish.



**Figure 2.** Five year mean concentrations of Acid Neutralising Capacity in UK AWMN lakes. Note last period is four years only.

The increased proportion of relatively acid-sensitive diatom and macroinvertebrate species at sites showing chemical improvement documented in the last report have mostly been sustained in the last 3 years. Acid-sensitive aquatic macrophyte species only recently detected have been re-recorded, as have juvenile brown trout in some of the most acid sites on the Network. There is little indication of further biological improvement since the last report however. Changes observed to date at the more acidified sites represent the early stages of improvement only, and densities of brown trout at several sites remain very low.

Comparisons of time tracks of diatom assemblages in sediment cores (showing ecological changes over the last 200 years) and sediment traps from AWMN lakes provide clear evidence for partial recovery in communities toward previous (less acidified) ecological states. However, in some cases, community composition appears to be developing along new trajectories, with increase in acid-sensitive taxa which were not common historically (Battarbee et al., in prep). Reasons for these species shifts are currently unclear but may reflect the eutrophying influence of nitrogen or long term increases in water temperatures in response to global warming.

The AWMN is probably unique internationally as a long-term fully integrated chemical and biological assessment network for source waters, primarily influenced by depositional and meteorological effects. As such it represents a national resource that has wide application in the biogeochemical sciences. AWMN data are central to other research programmes funded by the Department for Environment Food and Rural Affairs, the UK National Environment Research Council and the European Union Framework programmes. At its current size and breadth of measurements it has huge potential to address issues not only relating to acidification, but also eutrophication by deposited nitrogen, climate change, biodiversity and upland land management in the UK source water regions.

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## 5. Fish studies in Central Asia (Kazakhstan and Kyrgyzstan)

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A Joint Norwegian Kazakh and Kyrgyz project has been established related to the contamination associated with uranium (U) mining and tailing in the former USSR republics. The aims of the project are to assess long-term consequences from radioactive and trace metal contamination associated with U mining and tailing at selected sites in Kazakhstan and Kyrgyzstan, to evaluate the needs for alternative countermeasures, and to strengthen the scientific competence as well as the national authorities with respect to infrastructure and legislation for radiation protection. The project is a joint collaboration between responsible scientists and authorities in Norway, Kazakhstan and Kyrgyzstan. Participants from Norway include scientists from the University of Life Sciences in Norway (UMB) and the Norwegian Radiation Protection Authority (NRPA). Participants from Kazakhstan include scientists from Al-Farabi Kazakh National University (Al-Farabi University), Institute of Nuclear Physics of the National Nuclear Centre of the Republic of Kazakhstan (INP NNC KZ) and Central Scientific Research Analytical Laboratory, Volkhovgeologia. Participants from Kyrgyzstan include The Central Research Laboratory (CRL).

The present paper describes some data from the fish population studies in the first joint field work, which took place in June 2006, at the U mining area at Kurday, Kazakhstan, and in August 2007 in Kadji-Say, Kyrgyzstan. Radon detectors were placed indoor and outdoor in local villages. A series of samples were collected (soil, plants, water, sediments, macrophytes and fish) and in situ fractionation of radionuclides and metals in lake, river and well water were performed. A broad analytical programme has been implemented.

### **Kurday mining site, Kazakhstan**

Kurday is a small village situated in the south-east of Kazakhstan, not far from the border of Kyrgyzstan. The U mining site at Kurday opened in 1953, and was closed in the end of the 1950s. During mining, the U containing rocks were crushed and transported by train from the site, and the site has been left without remediation until recently. Thus, the radionuclide and metal contamination associated with the mining activities within the site and a possible impact of the environment have never been investigated. The field investigated included Dead Lake, an artificial lake formed by rainwater filling the mining site. Russian soldiers and later the local populations have stocked the lake with fish.

## Kadji-Say mining and processing site, Kyrgyzstan

Kadji-Say is situated at the southern shore of Issyk-kul, the worlds second largest high mountain lake. In the mountain facing the lake, U mining and processing took place. As in Kurday, remediation has only recently been done, covering the uppermost tailings with ash, clay and soil, but large areas were left with little remediation. Issyk-kul has a traditional fishing, and over the years many new species for Kyrgyzstan have been introduced.

## Sampling and results

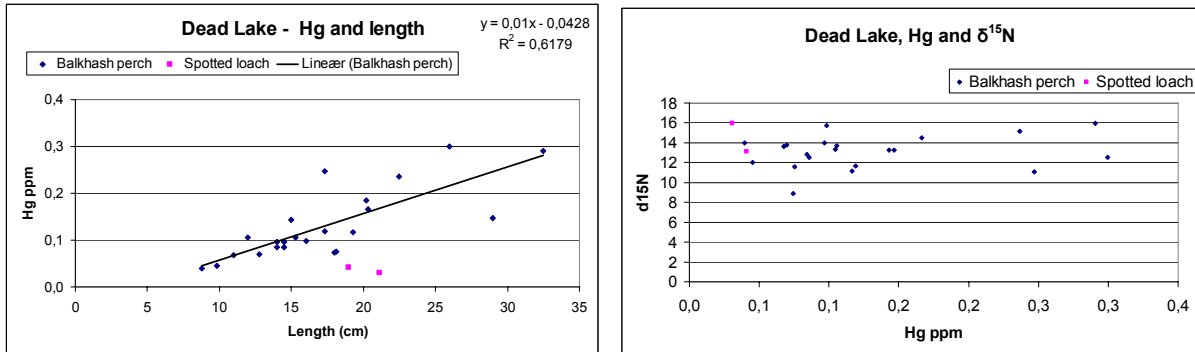
Testfishing, using the Nordic multifilament gillnet series were used in June 2006 in Dead lake, Kazakhstan (Fig. 1), and in August 2007 in Issyk-kul, Kyrgyzstan. Live fish were kept in a keepnet, and later sampled for blood and organs following the EMERGE protocol (Rosseland et al. 2003). In Dead Lake, two species of fish were captured; numerous Balkhash perches (*Perca schrenkii* Kessler) and two spotted loach (*Triplophysa strauchii*). In Issyk-kul, three fish species were analysed; pike perch (*Stizostedion lucioperca* L.), Issyk-kul Chebachok (*Leuciscus bergi* Kacharov), and White fish (*Coregonus lavaretus* L.). Filet of all species were analyzed for total mercury (THg) and stable isotopes of nitrogen ( $^{14}\text{N}$  and  $^{15}\text{N}$ ) and carbon ( $^{13}\text{C}$  and  $^{12}\text{C}$ ). The analysis of THg (wet weight, ww) was carried out by using the Perking-Elmer model FIMS 4000 Flow Injection Mercury System at the Environmental Chemistry Section, Norwegian University of Life Sciences. The stable isotope analyses were carried out at the Isotope Laboratory, Department of Plant and Environmental Sciences, UMB using IRMS (Thermo). Nitrogen and carbon were analysed from homogenized and freeze-dried muscle samples subjected to combustion in a Flash Elemental Analyser (EA). The isotopic ratios ( $^{15}\text{N}/^{14}\text{N}$ ,  $^{13}\text{C}/^{12}\text{C}$ ) were expressed as delta-values ( $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$ ), and a  $\delta^{15}\text{N}$  difference of 3.4 ‰ indicate a shift in trophic level.



**Figure 1.** Sampling of fish organs at Dead Lake, Kazakhstan. (Photo: G. Strømman)

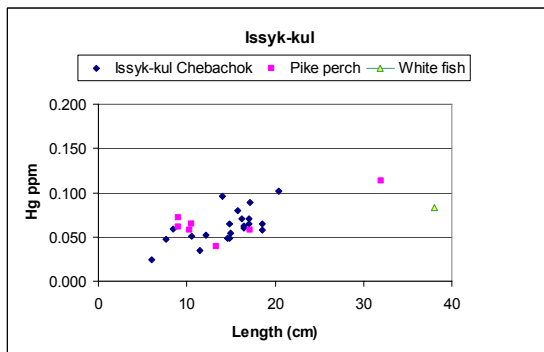
The total mercury and isotope ratios of N and C in relation to length classes of fish from Dead Lake are shown in Fig. 1. The Balkhash perch represented age classes from 1-12 years, thus the growth of the fish population was extremely slow, Fig.1. Both species had the same  $\delta^{15}\text{N}$

signal, and were thus on the same trophic level. The pike perch showed a clear bioaccumulation of Hg with size and thus age (Fig. 2, left), but biomagnification could not be observed (not piscivores) as there were no clear changes in  $\delta^{15}\text{N}$  with increasing mercury concentrations (Fig. 2, right).



**Figure 2.** THg in muscle tissue of the two species from Dead Lake, Kazakhstan, in relation to length (left) and  $\delta^{15}\text{N}$  (right).

THg in the three fish species in Issyk-kul, Kyrgystan, is shown in Fig.3. The general level of THg was low. Only one specimen of White fish was analysed. Both pike perch and Issyk-kul Chebacheck showed bioaccumulation properties.



**Figure 3.** Total Hg in muscle tissue of three species from Issyk-kul, Kyrgystan, in relation to length.

## Conclusion

We have a series of validated biomarkers for metals, heavy metals, radionuclides and POPs which we use to identify effects of environmental pollution on fish. In Dead Lake, Kazakhstan, the THg content in muscle of Balkhash perch at length > 17 cm exceed consumer's advice for human groups at risk (0.2 ppt Hg, FAO/WHO 2003). In Issyk-kul, Kyrgystan, no elevated mercury levels were found for the three species analysed in 2007.



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## 6. Modelling the effect of climate change on the acidification recovery of an alpine lake

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Lake Paione Superiore (LPS) is a high-altitude lake (2269 m.a.s.l.), located in the Central Alps, Italy (Marchetto *et al.* 2004). It is an acid-sensitive site that has been showing signs of recovery the recent period (increasing pH and alkalinity) as an effect of the decreasing SO<sub>4</sub> deposition (40% reduction in the Alps in the last two decades).

LPS is one of the study sites included in the EU Project EUROLIMPACS (Evaluating the Impacts of Global Change on European Freshwater Ecosystems; <http://www.eurolimpacs.ucl.ac.uk/>), more specifically in Work-package 4, dealing with climate-acidification interactions.

MAGIC (Model of Acidification of Groundwater in catchments; Cosby *et al.*, 2001) has been used within WP4 to test the relative sensitivity of individual processes to climate change. The aim was to evaluate several possible climate-induced effects on the recovery of soil and surface water from acidification at 14 intensively-studied sites in Europe and Eastern North America, including LPS.

Results of this modelling exercise were presented in a paper by Wright *et al.* (2006) and included in a Special issue of the EUROLIMPACS project. The trials suggested that in future modelling of recovery from acidification, possible concurrent climate changes should be taken into account and focus specially on climate-induced changes in organic acids and nitrogen retention. As for LPS, change in runoff and eventual increase in Saharan dust deposition proved to be the major drivers of change (Wright *et al.* 2006).

According to measured data, decreasing precipitation and snowmelt are the main drivers of change for alpine lake chemistry in the Central Alps, together with long-term change in atmospheric deposition (Rogora *et al.* 2003). In order to obtain a more refined and realistic prediction of LPS response to future change in climatic drivers, we made a preliminary application of climate change scenarios (HadCM3 model, A2 and B2 experiments), coupled with deposition scenarios.

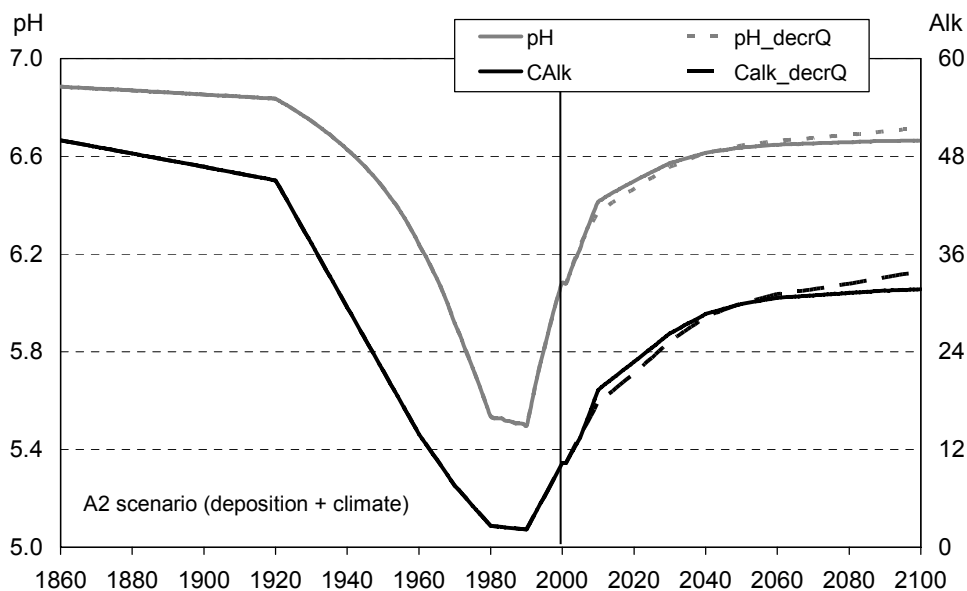
MAGIC has previously been calibrated at LPS using measured data from the period 1984-2000 (Rogora, 2004). Statistical DownScaling Model (Wilby *et al.* 2002; <https://co-public.lboro.ac.uk/cocwd/SDSM/>) was used to generate climate scenarios (temperature and precipitation) for LPS from General Circulation Models (GCMs). Precipitation and temperature data were used to calculate scale factors for runoff for the period 2000-2100, calculating evaporation by the Haude formula. A worst case scenario was generated (DecrQ A2), and used in MAGIC for the forecast. Future deposition of S- and N-compounds at LPS was modelled based on the implementation of the current legislation scenario (CLE), which is

based on the Gothenburg protocol under the UN-ECE Convention on Long-range Transboundary Air Pollution.

According to this scenario, SO<sub>4</sub> deposition in Northern Italy will continue to decrease in the coming years, though less pronounced than the reduction observed in the last 20 years. Nitrate deposition is supposed to decrease by 40% by 2100 with respect to 2000 levels, while no variation is envisaged for ammonium (Rogora, 2004).

Model output is presented in Figure 1 and is concerned with long-term change in pH and alkalinity values. The overall effect of runoff change at LPS will be a slight delay in the recovery pattern until 2040. The effect will be very small with respect to the base case scenario (deposition change only). The limited changes foreseen for pH and alkalinity result from the compensating effect of increasing base cation (Ca, Mg, K) and acid anion concentrations, mainly NO<sub>3</sub>, from 2010 onwards. The former trend will be an effect of concentration, because of increasing evaporation and decreasing runoff in a warmer and drier climate. The same process would also determine an increase of acid solutes such as sulphate and nitrate. Furthermore, previous simulations by MAGIC at LPS highlighted how the changes in N deposition, predicted under the CLE scenario, will not be enough to get a substantial decline of NO<sub>3</sub> in lake water, and allow for a considerable chemical recovery (Rogora, 2004).

The LPS case-study is a preliminary example of how climate scenarios from GCMs can be applied to simulate the simultaneous effects of deposition and climate change in acidification recovery. The next step in sophistication of modelling climate-induced effects will be to use long-term data series to generate empirical relationships between climate variables and water chemistry, or catchment processes.



**Figure 1.** Time series for pH and ANC ( $\mu\text{eq l}^{-1}$ ) at LPS as simulated by MAGIC for the period 1860–2100 under base case (deposition only) and climate (decreasing runoff, DecrQ) scenarios. Climate scenario derived from HadCM3 model, A2 experiment.

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## 7. Reports and publications from the ICP-Waters Programme

All reports from the ICP Waters programme from 1987 up to present are listed below. All reports are available from the Programme Centre. Publications from 2002 to present can be found at <http://www.iis.niva.no/icp-waters/> and the new web page <http://www.icp-waters.no/>

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