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International Co-operative Programme
on Assessment and Monitoring of
Acidification of Rivers and Lakes:
8th Task Force Meeting, 1992

D. Smith and I. Davies

Central and Arctic Region
Department of Fisheries and Oceans
Winnipeg, Manitoba R3T 2N6

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**INTERNATIONAL CO-OPERATIVE PROGRAMME
ON ASSESSMENT AND MONITORING
OF ACIDIFICATION OF RIVERS AND LAKES:
8th TASK FORCE MEETING,
1992**

Edited by
D. Smith and I. Davies

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EDITOR'S COMMENTS

This volume contains papers presented at the 8th task force meeting of the International Co-operative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes. A list of attendants and their addresses have been included, as well as the agenda, which was sent out to each participant. The papers were not subject to a full formal review process, however, the editors did transcribe the papers into the format used by Can. J. Fish. Aquat. Sci., as much as possible. Editorial text changes, that were made to the manuscripts submitted for publication, were done to increase the readability of the papers. We have tried not to change the authors' original content or meaning, but we apologise if this has occurred. Questions on any individual contribution should be directed to the authors. Any statements or views presented here are totally those of the authors and are neither condoned or rejected by the editors. Mention of trade names or commercial products does not constitute endorsement or recommendation for use. Lastly, we the editors, regret that it has taken so long to assemble these proceedings. We are grateful to the authors for their patience.

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ABSTRACT

Smith, D. and I. Davies (ed.) 1997. International co-operative programme on assessment and monitoring of acidification of rivers and lakes: 8th task force meeting, 1992. Can. Tech. Rep. Fish. Aquat. Sci. 2155: iv+68 p.

The convention on long-range transboundary air pollution, 8th annual task force meeting of the International Co-operative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes was held at the Norwood Hotel in Winnipeg, Manitoba, Canada from September 14th to September 18th, 1992. There were 29 participants. Administrative reports and discussions are summarised in the minutes, which have been distributed separately. Many scientific presentations were given, 8 of which were submitted for publication and are collected in this volume. A tour of the Department of Fisheries and Oceans Experimental Lakes research station (ELA) was attended by the participants from 17th-18th September, 1992

The main focus of the meeting was to establish an international exchange of information on assessment, monitoring, and status of acidification of rivers and lakes. Task force meetings emphasise an informal exchange of ideas and knowledge among participants in the programme. This forum provides an annual focus on the principles, current problems and approaches in long-range transboundary air pollution, and its effect on aquatic systems. The meeting was jointly chaired by B. Kvaeven (Norwegian State Pollution Control Authority) and M. Johannessen (Norwegian Institute for Water Research) and organized by I. Davies (Department of Fisheries and Oceans) and D. Jefferies (Department of the Environment), representatives from the host country. Papers presented here were given at the meeting and submitted by the authors for publication in Can. J. Fish. Aqu. Sci. Technical Report Series. Publication costs were covered by the Department of Fisheries and Oceans.

Key words: Long-range, transboundary, air-pollution, ICP Waters, NIVA, Acidification, Winnipeg

RÉSUMÉ

Smith, D. and I. Davies (ed.) 1997. International co-operative programme on assessment and monitoring of acidification of rivers and lakes: 8th task force meeting, 1992. Can. Tech. Rep. Fish. Aquat. Sci. 2155: iv+68 p.

CRITICAL LOADS MAPPING FOR FRESHWATERS IN THE UK

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Introduction

The UK Critical Loads for Freshwater Programme is a collaborative exercise sponsored by the Department of Environment (Air Quality Division) and undertaken by the Environmental Change Research Centre, University College London; Institute of Hydrology (Wallingford, Oxfordshire); Scottish Office Agriculture and Fisheries Department, Freshwater Laboratory (Pitlochry, Perthshire); Institute of Terrestrial Ecology (Monks Wood, Huntingdon) and School of Applied and Pure Biology, University of Wales (Cardiff).

The critical acidity load for an aquatic ecosystem is exceeded when the first biological change that can be related to acid deposition occurs. As load increases through time sensitive surface waters become more acidic and many organisms progressively decline and eventually disappear. Target loads may be set according to the need to protect any selected species or groups of species and the need to enable such species to recolonize former habitats.

In the UK Freshwaters Programme two different, but complementary empirical models have been used to set critical loads for individual waterbodies. These are the diatom model (Battarbee et al. 1993) and the steady-state chemistry model (Henriksen et al. 1992). The diatom model is used to set the basic critical load for the site, while the steady-state chemistry model is used to set a critical load needed to maintain a pre-set alkalinity (ANC) to protect individual species or species groups. These two models have been used to map critical loads on a 10 km x 10 km grid square basis throughout the UK. In addition, MAGIC, a dynamic model (Cosby et al. 1985) has been used at selected sites to explore the effect of time-lags, land-use, and other factors in achieving or not achieving critical loads at different streams and lakes.

A detailed account of the UK critical loads results for freshwaters, including exceedance maps for the present and projected for 2005, is presented in Battarbee et al. (1992).

Mapping

Because of the inadequacy of existing water chemistry datasets, a new water chemistry survey of the whole UK was carried out. One site, chosen to be the most sensitive, from each grid square was sampled and analysed. To minimize problems of temporal variability standing rather than running waters were selected, but, where no suitable standing waters were available, low-order streams were used. Criteria for site selection are given in Kreiser and Battarbee (1993).*

* The editors are unsure as to the authors intended reference.

Critical Loads: The Diatom Model

As diatoms are among the most sensitive organisms to acid stress, the point at which the first evidence of acidification occurs in the diatom record in a lake sediment core can be regarded as the time at which the "critical load" for that lake was exceeded. Detailed examination of cores from 53 lakes with varying sensitivity to acidification throughout the UK uplands shows variations in the degree of acidification at any site according to base cation concentration of the water and the amount of sulphur deposition received by the site. This relationship can then be used to produce a dose-response model to predict the acidification status of UK freshwaters and it can be used to calculate the critical load for acidity for a site (Battarbee et al. 1993). A critical loads map for the United Kingdom calculated in this way is shown in Fig. 1.

Critical Loads: The Steady State Water Chemistry Model

The steady-state water chemistry (or Henriksen) model assumes a situation where ionic inputs are at equilibrium with outputs, allowing mean chemical data to be used (Henriksen et al. 1992). The method is based on the principle that excess base cation production in a catchment should be equal to or greater than the acidic anion input, thus producing a positive critical load.

Unlike the diatom model which is used to set the base, or site critical load, the steady-state chemistry model is designed to set critical loads for individual species or assemblages. If the relationship between the probability of occurrence of a taxon and the ANC of the water is established, critical ANC values can be derived and used in the model to calculate the critical loads needed to protect that species. Suitable chemical/biological datasets to explore these relationships for a range of species are currently being compiled.

So far in the UK, maps have been produced for $\text{ANC} = 0$ considered to be the critical ANC value in predicting brown trout populations. This value has also been adopted by the UNECE for international mapping purposes (UNECE 1990). The UK critical loads map for $\text{ANC} = 0$ is presented in Harriman and Christie (1993) and shown here in Fig. 2. As expected, a comparison of the two maps (Figs. 1 and 2) shows that critical loads for $\text{ANC} = 0$ are higher than the base critical load derived from the diatom model. This indicates that while $\text{ANC} = 0$ may offer protection to brown trout, higher alkalinities are required to protect many other aquatic organisms (Ormerod, personal communication).

Critical Loads Maps: Uncertainties

The maps produced by the two methods described above are based on the chemistry of single samples from single sites within each grid square. Consequently, despite a good agreement between the maps and the expected geographical patterns, there are several uncertainties in the data and the results still require validation. The main concerns are:

- a. problems of high sea-salt inputs in the north-west of Scotland;
- b. temporal variations in critical loads according to seasonality and stage, especially for stream systems;
- c. the relationship between the critical load for the site sampled and the spatial variation in critical loads across grid squares.

These issues are being considered in the current phase of research.

Reversibility

As acid deposition decreases in the future, towards the critical load, it is important to know whether streams or lakes will recover chemically and biologically. In the UK this issue is being addressed by using the predictive capacity of the MAGIC model and by monitoring.

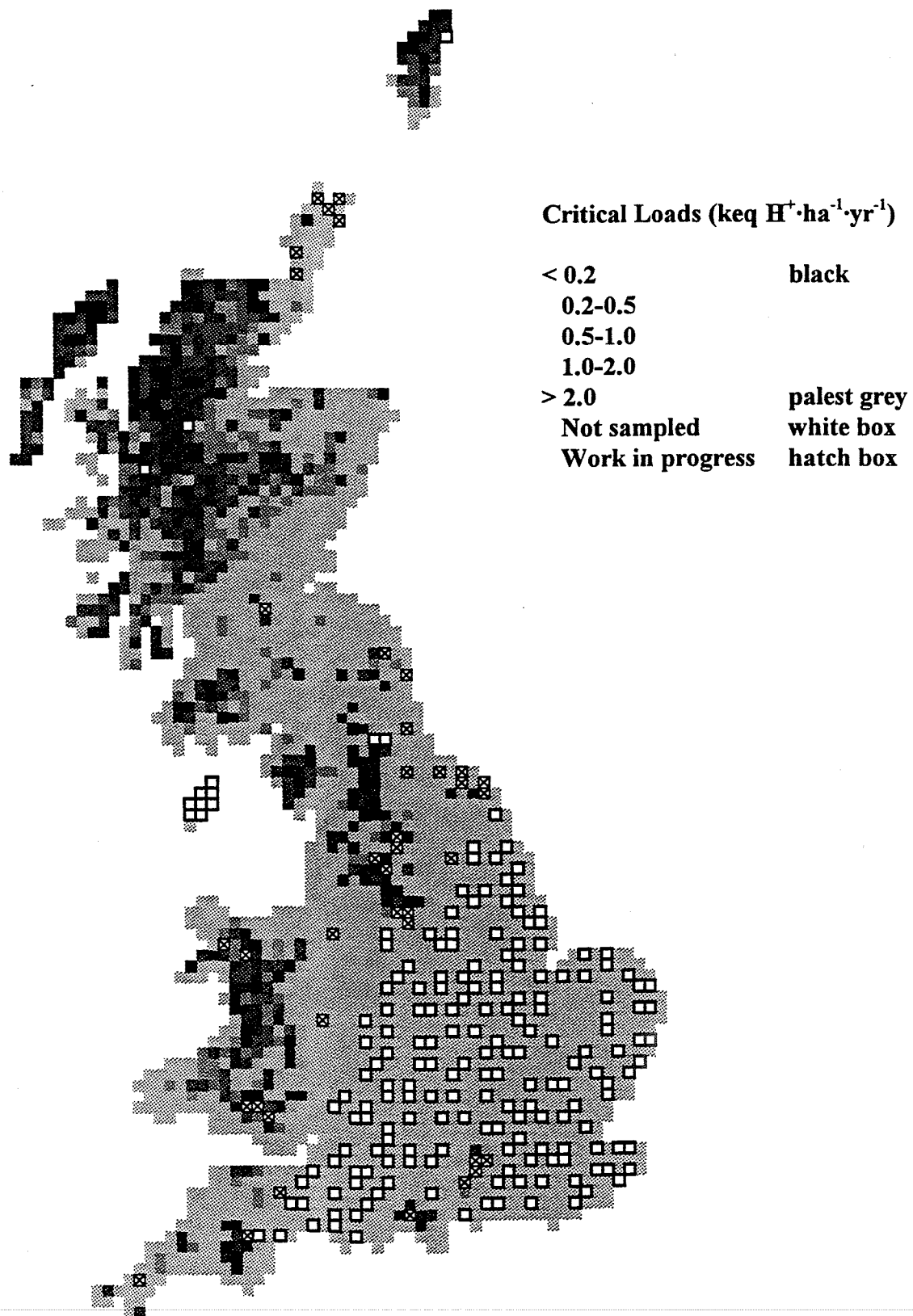


Fig. 1. Provisional Critical Loads for Sulphur for UK Freshwaters, 1992 (Henrikson Model).

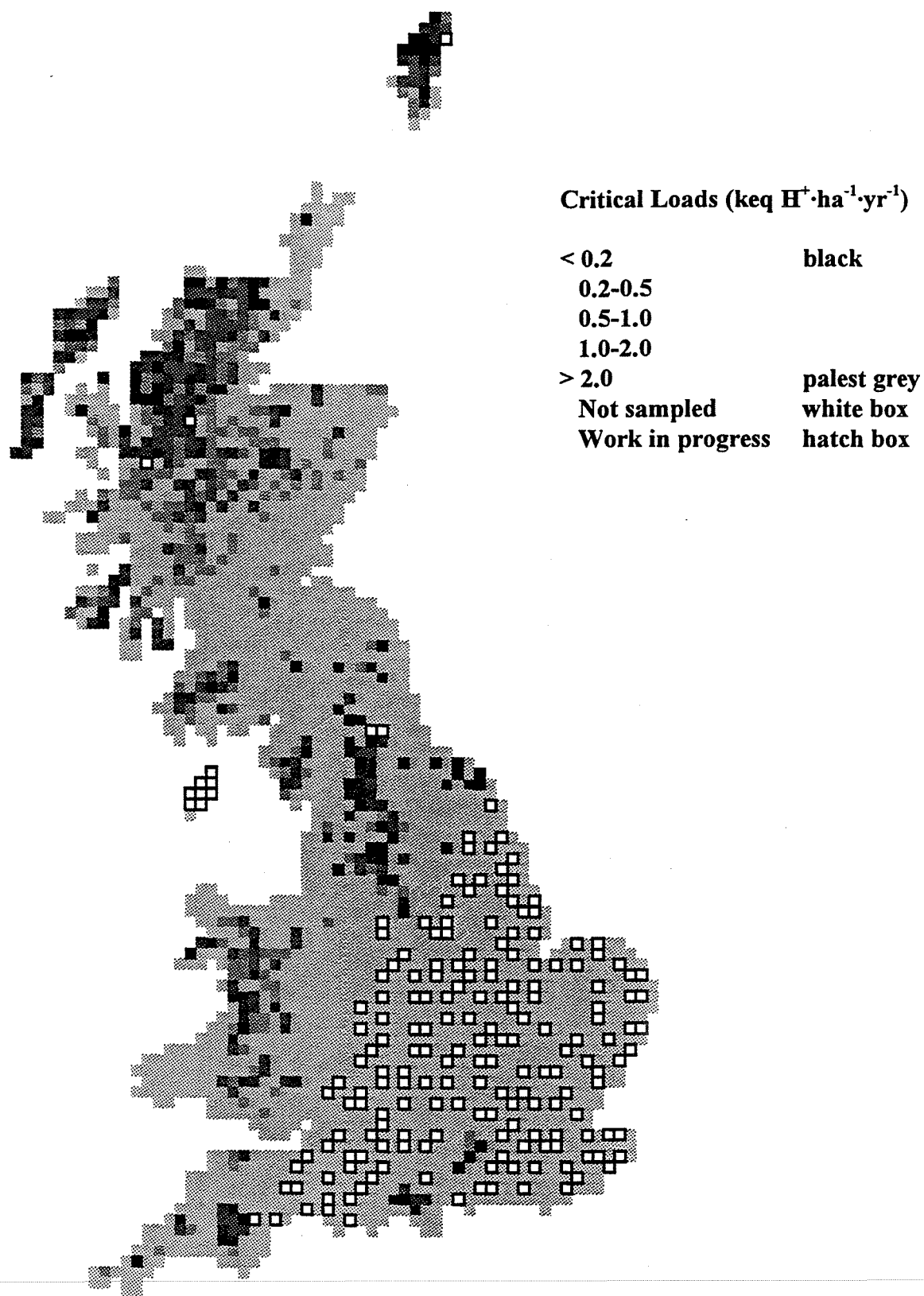


Fig. 2. Provisional Critical Loads for Sulphur for UK Freshwaters, 1992 (Diatom Model).

In an analysis of 39 lakes in the Galloway region of Scotland the MAGIC model predicts that acidified waters with negative alkalinities in the region will recover only marginally in response to a 60% reduction in UK emissions by the year 2005, and it indicates that the total percentage of lakes with negative alkalinity will remain the same (Jenkins and Shaw, in prep.).

These projections are somewhat pessimistic. Monitoring data so far available, however, are more encouraging, indicating that small but significant increases in the pH of Loch Enoch, Loch Valley, and the Round Loch of Glenhead took place between 1979 and 1990.

The extent to which these and other surface waters will respond further, as emissions begin to decrease again in the second half of the 1990's, is unknown, but excellent baseline data are available from the critical loads research programmes and from the Acid Waters Monitoring Network (Patrick et al. 1991) to enable a full evaluation to be made, and to allow the projections of the MAGIC model to be evaluated.

Acknowledgements

We would like to thank Ron Harriman, Alan Jenkins, Catherine Shaw, Jane Hall, Keith Bull, Steve Ormerod, Bob Wilson, Steve Juggins, Tim Allott, Annette Kreiser, and members of the Environmental Change Research Centre for their help and co-operation on the Critical Loads project. The project is funded by the UK Department of Environment, administered by the Air Quality Division.

References

- BATTARBEE, R.W. 1992. Critical loads and acid deposition for UK freshwaters. Environmental Change Research Centre, University College London, Research Paper No. 5. 115 p.
- BATTARBEE, R.W., T.E.H. ALLOT, A.M. KREISER, and S. JUGGINS. 1993. Setting critical loads for UK surface waters: the diatom model. In M. Hornung and R.A. Skeffington (ed.) Critical loads: concepts and applications. ITE Symposium No. 28, London, HMSO. In press.
- COSBY, B.J., G.M. HORNBERGER, J.N. GALLOWAY, and R.F. WRIGHT. 1985. Modelling the effects of acid deposition: assessment of a lumped-parameter model of soil water and streamwater chemistry. *Wat. Resour. Res.* 21: 51-63.
- HARRIMAN, R., and A.E.G. CHRISTIE. 1993. Critical loads and surface waters: evaluation of the steady-state water chemistry method. In M. Hornung and R.A. Skeffington (ed.) Critical loads: concepts and applications. ITE Symposium No. 28, London, HMSO. In press.
- HENRIKSEN, A., J. KAMARI, M. POSCH, and A. WILANDER. 1992. Critical loads of acidity: Nordic surface waters. *Ambio* 21: 356-363.
- JENKINS, A., and C. SHAW. In prep. Critical loads for surface waters: comparison of empirical and dynamic model approaches.
- KREISER, A.M., S.T. PATRICK, and R.W. BATTARBEE. 1993. In M. Hornung and R.A. Skeffington (ed.) Critical loads: concepts and applications. ITE Symposium No. 28, London, HMSO. In press.
- PATRICK, S.T., S. JUGGINS, D. WATERS, and S. JENKINS. 1991. The United Kingdom acid waters monitoring network: site description and methodology report. Report to the DoE and DoE(NI). ENSIS Ltd. 63 p.
- UNECE. 1990. Draft manual for mapping critical levels/loads. Task force on mapping. United Nations Economic Commission for Europe, Berlin.

CHEMISTRY TRENDS IN ATLANTIC CANADA LAKES AND RIVERS

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Abstract

Thirty-nine lakes from southwest Nova Scotia and 33 from Newfoundland, as well as 26 rivers from the whole of Atlantic Canada were sampled from the spring of 1983. Trends of acidification related water chemistry variables were evaluated using non-parametric techniques. Results showed that pH increased in almost half of Newfoundland lakes. Acid neutralization capacity (ANC) increased in one-third of Nova Scotia lakes and in six of the Newfoundland sites. Despite the strong evidence from pH and ANC which suggested a reduction in acidification effect, only a few lakes in Nova Scotia showed decreases in sulphate (SO_4^{2-}). Moreover, base cation concentrations also did not seem to vary much over time. The situation for the rivers was similar to that of lakes. Hydrogen ion concentrations decreased in 11 of the 26 rivers sampled, while ANC also increased at 11 sites. SO_4^{2-} actually increased at five sites, though NO_3^- decreased at three.

Key words: Nova Scotia, Newfoundland, acidification, Acid Neutralizing Capacity, sulphate

Introduction

Atlantic Canada receives significantly higher than background levels of acidifying ions in precipitation (Sirois and Summer 1989). Clair and Whitfield (1983), using time-series analyses, showed that trends in sulfate (SO_4^{2-}) increased and pH decreased in sensitive rivers in the area from 1968 to the late 1970's. In contrast, Green et al. (1986), using a variety of statistical procedures on data from the same sites, found that pH and SO_4^{2-} were relatively stable from the mid-1960's to the mid-1980's. Part of the difference in interpretation between the two studies was likely due to inappropriate analytical methods used in the laboratory analysis of samples and differences in the time period studied. For example, SO_4^{2-} was measured using the methyl thymol blue method (Anonymous 1984) on most waters collected before 1981. Later work (Kerekes et al. 1984) eventually showed the method overestimates actual values due to interferences from natural organic acids. Another problem encountered was that the two end-point alkalinity approach (Anonymous 1984), with a high detection limit of $0.5 \text{ mg}\cdot\text{L}^{-1}$, was used on early samples. Many surface waters in Atlantic Canada, because of very low buffering capacities of soils and bedrock, have concentrations lower than the detection limit, thus generating large numbers of censored values.

In recent years, major advances have occurred in both laboratory and statistical approaches. Since the early 1980's, the ion chromatography (IC) and Gran titration approach have become commonly used for SO_4^{2-} and acid neutralisation capacity (ANC) respectively, so that the data accumulated are of higher quality. Secondly, important advances have occurred in trend detection techniques. Cluis (1988, 1989) and Loftis (1991) have summarized a number of non-parametric approaches which require fewer restrictions to assess data sets for trends. A combination of improvements in the actual data, as well as its interpretation, now allow a more rigorous assessment of trends in freshwater chemistry.

There were measurable decreases in emissions of acidifying substances from the United States and Canada from 1980 to 1985 (RMCC 1990a), which were reflected in decreased SO_4^{2-} deposition in

Nova Scotia from 1980 to 1987 (Sirois and Summers 1989; Underwood et al. 1989) and Newfoundland between 1982 and 1987 (Ryan et al. 1990). The estimated decrease in SO_4^{2-} was from $12\text{-}6\text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ in Newfoundland, but was much less precipitous in Nova Scotia. This report was prepared in order to see if the measured changes in precipitation chemistry could also be related to surface waters.

In 1983, the Conservation and Protection Service of Environment Canada began monitoring a number of lakes and rivers in the eastern portion of Canada, the Atlantic Provinces, which include New Brunswick, Prince Edward Island, Nova Scotia, and Newfoundland. Most of the sites were selected in 1982 using historic survey data (Howell 1986).

Many basins in Atlantic Canada are low in dissolved inorganic ions compared to those from other locations, making them sensitive to acidification effects. Many also have high dissolved organic carbon levels, makes it difficult to study trends in acidification. Not only does DOC itself tend to lower water pH from the dissociation of organic acids, but the organic anions so formed can then buffer the solution from mineral acids. In order to quantify the changes in pH, ANC, and other parameters with changes in acidification, the relationship between parameters was expressed following the electroneutrality principle where:

$$\sum C_b - \sum C_a = \text{ANC} = [\text{HCO}_3^-] + 2[\text{CO}_3^{2-}] + [\text{OH}^-] + [\text{O}_r^-] - [\text{H}^+] \quad [\text{Eq. 1}]$$

C_b and C_a denote the sum of the base cations (Ca^{2+} , Mg^{2+} , K^+ , Na^+) and acid anions (SO_4^{2-} , Cl^- , NO_3^-) respectively, ANC is the acid neutralisation capacity as measured using the Gran titration, HCO_3^- (bicarbonate) and CO_3^{2-} (carbonate) are measured as dissolved inorganic carbon (DIC) and estimated using pH (Stumm and Morgan 1981), OH^- (hydroxyl), and H^+ (hydrogen) are calculated from the pH measurement, and O_r^- is the organic anion component contributed to water from the breakdown of plant organic matter. Assuming that the organic anion contribution is independent of acid precipitation effects and thus will remain stable, changes in SO_4^{2-} which would reflect acid deposition variations, should then lead to changes in ANC and pH (Eq. 1). Changes in SO_4^{2-} deposition could also modify C_b , as acid deposition increases bedrock weathering and thus can increase base cations (Kilham 1982).

By using the information collected in the sampling programme and analysing it within an electroneutrality framework, we should be able to understand the causes of measured trends. The purpose of this report is to describe and explain results of the trend analysis conducted on Environment Canada's Atlantic lake monitoring network from 1983 to 1989. This work summarises lake trend results reported in Clair et al. (1992) as well as unpublished results from the Atlantic Canada river network.

Area and Methods

Water Sampling and Analysis

Monitoring of precipitation chemistry in the Atlantic Region was done at a number of sites (Vet et al. 1988), though the data have only been analysed for trends at Kejimikujik National Park, in the western portion of the study area and at Baie D'Espoir, Newfoundland, located at the eastern end of the study area. A continuous trend of decreasing SO_4^{2-} deposition in precipitation was measured from 1979 to 1987 (RMCC 1990b). The situation was somewhat different for NO_3^{2-} deposition, as it seemed to increase slightly during the same period. However, overall deposition of acidifying anions showed a decrease at this site. Amounts of acidifying substances deposited in Newfoundland are approximately half those in Nova Scotia, due to the greater distance of this region from pollution sources.

Since the spring of 1983, 72 lakes have been sampled twice a year (during the spring and fall lake overturn periods), in the two Atlantic Provinces (Figs. 1 and 2). Contemporaneously, 26 rivers were sampled monthly. The lakes and rivers were chosen because their basin geology and geography made them sensitive to acid precipitation and because of their low anthropogenic development (Howell 1986).

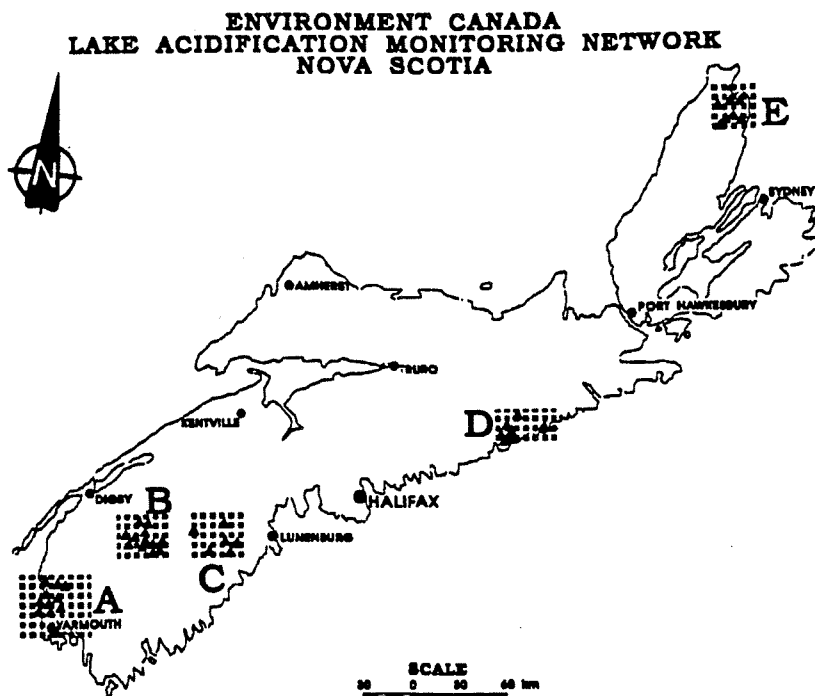


Fig. 1. Location of Nova Scotia lake sampling sites.

Lake sampling was conducted only in Nova Scotia and Newfoundland using a helicopter at most sites from the spring of 1983 to fall 1989. Sampling procedure involved manual collection of water at 30 to 50 cm depth from the surface. Rivers were sampled using a sampling iron. At randomly chosen sites, triplicate samples were collected to measure reproducibility. The samples were shipped to the Environment Canada regional laboratory in Moncton, N.B., where they were analysed for major ions and other parameters using standard analytical methods (Anonymous 1984). Regular review of triplicate data showed that most values lay within $\pm 10\%$ of the mean.

SO_4^{2-} is the most important tracer of acidification of surface waters in the region. It, as well as Cl^- , were measured using the ion chromatography (IC) method, which avoids colour interference. Acid neutralisation capacity (ANC), which is sometimes called alkalinity, was measured by Gran titration. Calcium and magnesium were analysed by atomic absorption spectroscopy and potassium and sodium by flame photometry.

Dissolved organic carbon (DOC) is a major component of water chemistry of approximately half the lakes sampled. DOC is produced by soil, wetland, and planktonic biological activities and is transported to water courses by hydrological processes. This parameter is thus an indicator of basin background processes which should only vary with climatic changes. For trend analysis of lakes we used colour (measured against the platinum-cobalt relative scale) as a surrogate for organic content. Values ranged from less than 5 units for clear waters, to greater than 100 for brown-water draining bogs and other wetlands.

Dissolved organic carbon was analysed by the persulfate-UV digestion method and was used for trend analysis of the river data.

ENVIRONMENT CANADA
LAKE ACIDIFICATION MONITORING NETWORK
NEWFOUNDLAND

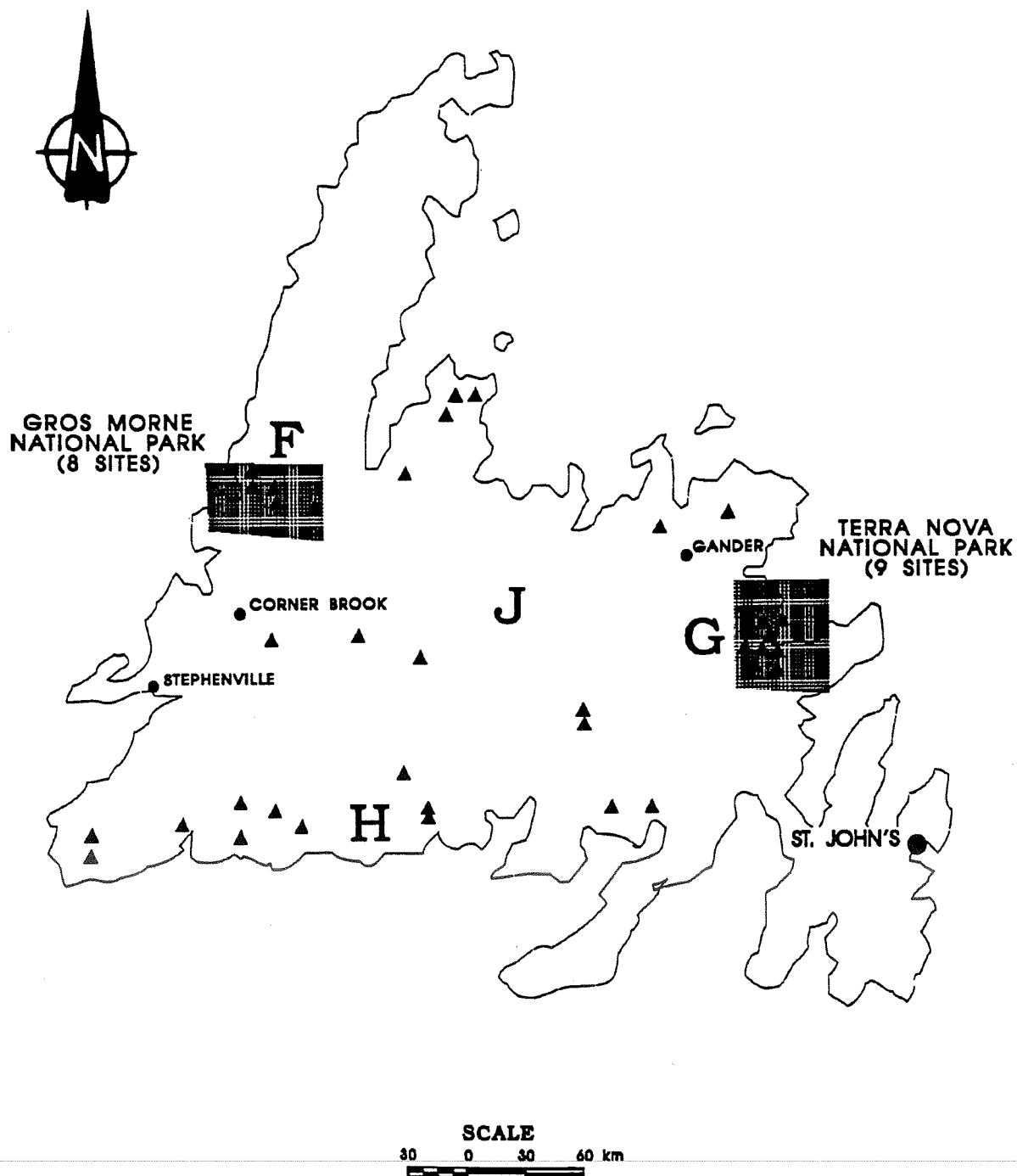


Fig. 2. Newfoundland lakes sampling locations.

In Atlantic Canada, Cl^- is often found at high concentrations in surface waters due to seaspray contributed by the nearby ocean. As Cl^- is thought not to be involved in basin biological, geological, or chemical processes, cations and anions contributed from seaspray were corrected for seasalt using this ion (Watt et al. 1979).

Statistical Analysis

Trend analysis was done on each parameter from each individual lake or river. Based on data characteristics, one of a series of non-parametric tests was used to analyse for trends in the data, as well as for seasonality and serial correlation (Markovian persistence). The trend tests which were selected are shown in Table 1 (Cluis et al. 1988).

Table 1. Statistical methods used to analyse lake data for monotonic trends (Cluis et al. 1988).

Persistence	Seasonality	Test to be used
No	No	Kendall
No	Yes	Kendall Seasonal
Yes	Yes	Hirsh and Slack
Yes	No	Spearman/Lettenmaier

Due to the intensive nature of the sampling programme, less than 1% of the sampling points were missing. These were replaced by temporal interpolation using calculated seasonal means (Cluis et al. 1989).

After all data sets were analysed, results showed that either the Kendall seasonal or the Spearman/Lettenmaier test provided the best results for all cases studied. Trend analyses were carried out on pH and H^+ , Gran alkalinity (or ANC), the sum of seasalt corrected base cations (Ca, Na, K, Mg), colour, Cl^- and SO_4^{2-} . The river data were analysed for H^+ , ANC, seasalt corrected Ca, SO_4^{2-} , and river discharge (Q).

Results

Each data set was tested for the null hypothesis of no significant trend at the 95% confidence level. Acidity measured as pH or H^+ was remarkably stable in Nova Scotia lakes (Table 2). Almost half of all Newfoundland lakes showed pH increases. One lake showed a pH decrease and the remaining (16) showed no trend. H^+ trends were less clear-cut. Four sites showed increased values, eight decreased, and 21 showed no change. It seems that the normalisation which occurs in the calculation of pH tended to smooth out the data and reduce background noise. It was also noted that pH and H^+ trends were not necessarily detected in the same lakes. River hydrogen ion concentrations decreased throughout the region, with no local patterns detected.

In Nova Scotia, 13 of the 42 lakes showed an increase in ANC; none showed a decrease. ANC in six Newfoundland lakes increased; none decreased. There were no obvious differences in trends among clusters in either region, as three of five Nova Scotia and four of five Newfoundland clusters showed lakes with increasing trends. As with pH, river ANC increased across the region with no detectable regional variations. ANC increases ranged from 1.1 to 3.2 $\mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$. The number of sites with evaluated ANC suggests that the basins are responding to the lower deposition rates measured.

Table 2. Summary of trends in sampling lakes of the Atlantic region.

	Nova Scotia			Newfoundland		
	↑	↓	↔	↑	↓	↔
SO ₄ ²⁻	2	6	32	2	3	27
pH	0	1	39	15	1	16
ANC	13	0	27	6	0	26
C _b [*]	1	2	37	2	3	27
Colour	2	0	38	5	34	24
Cl	0	3	26	6	0	19

Based on Equation 1, there are two possible explanations for an increase in ANC, either a decrease in ΣC_a due to reduced SO₄²⁻ in precipitation, or an increase in ΣC_b . Analysis of these two components was also done. In Nova Scotia, C_b increased in only one lake, as opposed to two where it decreased. In Newfoundland, two increased while three decreased. Overall, the low number of lakes which showed any trend, either up or down, suggests that this parameter is very stable, perhaps because changes in base cations, if they occur, are too subtle to allow successful trend detection. Because C_b is composed of calcium, potassium, magnesium, and sodium, which are all measured separately (then corrected for seasalt), the sum of errors inherent in multiple analyses would tend to mask any potential trends. The rivers showed six sites with increasing calcium, and only one with decreasing concentrations (Table 3). Lake sulphate concentrations decreased at four sites in Nova Scotia. All Nova Scotia sites which showed this trend are located in the extreme western part in an area of complicated geology and are strongly impacted by seaspray (Howell 1986). The trends measured here could be due to factors unrelated to acid precipitation. The high base cation content at some of the sites (Clair and Esterby 1991) also suggests that evaporates, such as gypsum (CaSO₄) or anhydride (CaSO₄*nH₂O) in glacial till, or highway salting activities could be modifying SO₄²⁻ in water without affecting pH or ANC.

The Newfoundland lake situation is more complicated. Overall, SO₄²⁻ increased at two sites, decreased at three, and remained unchanged at 27. Difficult geology probably explains the inconsistent trends. The SO₄²⁻ results are not consistent with the increases in pH and ANC observed in the study lakes.

Table 3 shows that SO₄²⁻ values increased at five sites (as did pH and ANC) while it remained unchanged at other locations, possible due to a hydrological concentration effect. A study of trends in discharge from the rivers, though, showed only two of the study rivers with significantly decreasing trends in discharge (Q), thus neither supporting nor rejecting this possibility.

The length and sampling frequency of the data set are important considerations in determining whether or not trends in chemistry could be measured. For a number of reasons, mostly logistical, sampling of the lakes was only conducted twice yearly, which has been shown to be useful in trend detection by Taylor and Loftis (1989). This result was also confirmed by our work, as a number of trends were detectable at the 95% confidence level.

By studying all of the lakes in the data set, it can be seen that 29 showed evidence of improved water quality: that is, increases in pH or ANC, or a decrease in SO₄²⁻. One lake showed a pH decrease and 42 showed no clear trends.

Table 3. Variable trends in the rivers under study in Atlantic Canada.

River	H ⁺	ANC	Ca ²⁺	SO ₄ ²⁻	NO ₃ ⁻	Q
Lepreau	↓	↑	⇔	⇔	⇔	↓
Point Wolfe	↓	↑	⇔	↑	⇔	⇔
Oromocto	⇔	↑	⇔	⇔	⇔	⇔
Salmon	⇔	⇔	⇔	⇔	⇔	⇔
Meteghan	⇔	⇔	⇔	⇔	⇔	⇔
Kelley	↓	↑	⇔	↑	↓	↓
Wallace	⇔	⇔	⇔	⇔	⇔	⇔
Tusket	↓	⇔	↑	↑	⇔	⇔
Roseway	⇔	⇔	⇔	⇔	⇔	⇔
Mersey	↓	⇔	↓	⇔	⇔	⇔
Medway	⇔	⇔	⇔	⇔	⇔	⇔
LaHave	⇔	↑	⇔	⇔	⇔	⇔
Liscomb	⇔	↑	⇔	⇔	⇔	⇔
St. Mary's	↓	⇔	⇔	↑	⇔	⇔
Clam Harbour	⇔	⇔	⇔	⇔	⇔	⇔
N.E. Margaree	⇔	⇔	⇔	⇔	↓	-
Salmon	↓	↑	↑	⇔	⇔	⇔
Churchill	↓	↑	↑	⇔	↓	⇔
Eagle	⇔	⇔	⇔	⇔	⇔	⇔
Torrent	⇔	⇔	⇔	⇔	⇔	⇔
Indian	↓	↑	↑	↑	⇔	⇔
Isle aux Morts	⇔	⇔	⇔	⇔	⇔	⇔
Piper's Hole	↓	↑	⇔	⇔	⇔	⇔
Rocky	⇔	⇔	↑	⇔	⇔	⇔
Mill	⇔	⇔	⇔	⇔	⇔	⇔
Dunk	↓	↑	↑	⇔	⇔	⇔

Results from Atlantic Canada were compared to those of Bouchard and Haemmerli (1990) who, using identical statistical approaches, measured trends in SO_4^{2-} , pH, and ANC from Québec lakes from 1983 to 1988. Their results partly conflict with ours, as they detected increases in SO_4^{2-} and acidity and decreases in ANC in the south-central part of the province, the area closest to Newfoundland. The difference between results may be due to local pollution sources, to differences in weather patterns, or to a combination of the two.

Prior to this study, trends in river chemistry measured by Clair and Whitfield (1983), using Box-Jenkins trend analysis, showed pH decreases in sensitive rivers for the region from 1966-69 to the mid and late 1970's. They also noted inconsistencies in ionic patterns.

Although trends in individual lakes for pH, ANC and SO_4^{2-} do not necessarily confirm each other, neither do they contradict. The inability to link chemical changes is an indication of the complex series of reactions and basin integrative processes which occur in soils, snow and waters, such as seasonal storage and preferential release of ions.

Our data indicate that a general improvement in acidification related water chemistry has occurred in the lakes and rivers of the monitoring network from 1983 to 1989. This improvement has occurred over the breadth of the 1200 km wide sampling area.

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References

- ANONYMOUS. 1984. NAQUADAT dictionary of parameter codes. Water Quality Branch, Environment Canada, Ottawa. 319 p.
- BOUCHARD, A., and J. HAEMMERLI. 1990. Étude des tendances dans le series chronologiques de la qualité de l'eau des lacs du réseau TADPA-Québec. Rapport Environment Canada, Conservation et Protection, Région du Québec, Centre St-Laurent, Longueuil, Québec. 86 p.
- CLAIR, T.A., and P.H. WHITFIELD. 1983. Trends in pH, calcium, and sulfate of rivers of Atlantic Canada. *Limnol. Oceanogr.* 28: 160-165.
- CLAIR, T.A., and S.R. ESTERBY. 1991. Chemical grouping of Environment Canada's lake acidification monitoring sites in Atlantic Canada. Tech. Rep. IWD-AR-WQB-91-168, Environment Canada.
- CLAIR, T.A., G. DEVARENNES, and G. HOWELL. 1992. Chemistry trends in Atlantic Canada lakes. Tech. Rep. IWD-AR-WQB-91-174, Inland Waters Directorate, Environment Canada. 77 p.
- CLUIS, D. 1988. Environmental follow-up: a mixed parametric and non-parametric approach. *Environ. Software* 3: 117-121.
- CLUIS, D., C. LANGLOIS, R. VAN COILLIE, and C. LABERGE. 1988. Development of a software package for trend detection in temporal series: application to water and industrial effluent quality data for the St. Lawrence River. *Env. Monit. Assess.* 12: 429-441.
- HIRSH, R.M., R.B. ALEXANDER, and R.A. SMITH. 1991. Selection of methods for the detection and estimation of trends in water quality. *Wat. Resour. Res.* 27: 803-813.
- HOWELL, G.D. 1986. Water quality characterisation of 78 acid precipitation monitoring lakes in Atlantic Canada. Tech. Rep. IWD-AR-WQB-86-109, Inland Waters Directorate, Environment Canada. 119 p.
- GREEN, P.E.J., B. FREEDMAN, C.A. FIELD, D. HAMILTON, and C. STEWART. 1986. Analysis of water chemistry data for 28 rivers in Atlantic Canada. Tech. Rep. IWD-AR-WQB-86-115, Inland Waters Directorate, Environment Canada.

- KEREKES, J., G. HOWELL, and T. POLLOCK. 1984. Problems associated with the sulfate determination in coloured, humic waters in Kejimikujik National Park, Nova Scotia (Canada). *Verh. Internat. Verein. Limnol.* 22: 1811-1817.
- KILHAM, P. 1982. Acid precipitation: its role in the alkalization of a lake in Michigan. *Limnol. Oceanogr.* 27: 856-867.
- RMCC. 1990a. The 1990 Canadian long-range transport of air pollutants and acid deposition assessment report. Part 2. Emissions and controls. Federal/Provincial Research and Monitoring Committee. Atmospheric Environment Service, Environment Canada, Downsview, Ont.
- RMCC. 1990b. The 1990 Canadian long-range transport of air pollutants and acid deposition assessment report. Part 3. Atmospheric Sciences. Federal/Provincial Research and Monitoring Committee. Atmospheric Environment Service, Environment Canada, Downsview, Ont.
- RYAN, P.M., D.A. SCRUTON, and D.W. STANSBURY. 1990. Acidification monitoring by the Department of Fisheries and Oceans in insular Newfoundland, Canada. *Verh. Internat. Verein. Limnol.* 24: 354-359.
- SIROIS, A., and P.W. SUMMERS. 1989. An estimation of atmospheric deposition input of sulphur and nitrogen oxides to the Kejimikujik watershed: 1979-1987. *Water, Air, Soil Pollut.* 46: 29-43.
- STUMM, W., and J.J. MORGAN. 1981. *Aquatic chemistry*. 2nd Ed. John Wiley and Sons, NY. 720 p.
- TAYLOR, C.H., and J.C. LOFTIS. 1989. Testing for trend in lake and groundwater quality time series. *Wat. Res. Bull.* 25: 715-726.
- UNDERWOOD, J.K., J.G. OGDEN III, and D.H. WALLER. 1989. Precipitation chemistry in Nova Scotia: 1978-1987. *Water, Air, Soil Pollut.* 46: 13-27.
- VET, R.J., W.B. SUKLOFF, M.E. STILL, J.B. MARTIN, W.F. KOBELKA, and A. GAUDENZI. 1988. Canadian air and precipitation monitoring network (CAPMoN) precipitation chemistry data summary 1984. Report: AQRB-88-01, Atmospheric Environment Service, Environment Canada.
- WATT, W.D., D. SCOTT, and S. RAY. 1979. Acidification and other chemical changes in Halifax County lakes after 21 years. *Limnol. Oceanogr.* 24: 1154-1161.

THE EFFECT OF FOREST TYPE ON STREAM ACIDITY AND CONSEQUENTLY, ON BENTHIC FAUNA

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Introduction

The forest area in Denmark is to be doubled within the next 70-100 years. This is the result of a political decision aimed at increasing the tree production and reducing the cultivated farmland, especially on poor soils. It is thought that afforestation will have an overall positive impact on the aquatic environment, primarily through reduced loading of nutrients to lakes and coastal areas. It must, however, be expected that the biological structure of streams affected by this afforestation will change radically due to reduced irradiation (Friberg and Kjeldsen 1994), changes in organic matter input (Cummins et al. 1989), water flow (Borg et al. 1988), and water chemistry. With regards to water chemistry, several investigations (Jenkins et al. 1990; Weatherley et al. 1989) have shown that afforestation with coniferous trees may result in acidification, but the effect of coniferous trees on Danish stream acidity is at present unknown.

This paper presents results of afforestation on water chemistry from six Danish streams running in both coniferous and deciduous forests. Furthermore, the benthic macroinvertebrate density and diversity of two acidic forest streams is compared with the fauna found in neutral forest streams. The possible effects of afforestation on streams currently having a non-forest riparian zone/catchment area are discussed in light of the results.

Material and Methods

Localities

The data presented in this paper were obtained between January and August 1992. The study was carried out in six forest streams situated in the eastern part of Jutland, Denmark. The streams are first and second order streams with 16.3-80.9 0/00 slopes and an average discharge of 1-32 L·s⁻¹.

Four of the streams run in predominately deciduous forests: Gammel Gråbæk, Øvre Tjærbæk, Kjeldbæk and Hørhave Bæk. These localities are dominated by beech (*Fagus sylvatica* L.), ash (*Fraxinus excelsior* L.) and alder (*Alnus glutinosa* L.). Two of the deciduous forest streams, Hørhave Bæk and Gammel Gråbæk, have coarse sand as the dominant soil type in their catchment areas, while Kjeldbæk has loamy sand and Øvre Tjærbæk loamy sand/sandy loam as the dominant soil type.

Two of the streams run in coniferous forests: Rye Nørreskov and Vesterskov. These localities are dominated by sitka spruce (*Picea sitchensis* [Bong.] Carr), but also Douglas spruce (*Pseudotsuga menziesii* [Mirb.] Franco) is abundant. Dominant soil type in the catchment area of both streams is coarse sand.

Tree species composition as well as dominant soil type within the stream catchment areas are shown in Table 1.

Table 1. Mean pH, alkalinity, water colour, catchment soil type and tree species composition in six Danish forest streams. Range (min, max) in parenthesis.

	pH	Alkalinity mmol·L ⁻¹	Water colour mg Pt·L ⁻¹	Dominant soil type in catchment area	Dominant tree species
Rye Nørskov	4.6 (3.7-6.2)	-0.051 (-0.19-0.054)	197 (73-395)	Coarse sand	Sitka spruce Douglas spruce
Vesterskov	5.3 (4.0-6.9)	0.016 (-0.11-0.15)	76 (16-162)	Coarse sand	Sitka spruce
Gammel Gråbæk	8.0 (7.8-8.1)	2.2 (1.9-2.3)	21 (10-70)	Coarse sand	Beech Ash
Øvre Tjærbæk	7.7 (7.6-7.8)	1.7 (1.1-2.2)	14 (7-28)	Loamy sand/ sandy loam	Ash Alder
Kjeldbæk	7.8 (7.3-8.0)	1.4 (0.74-2.0)	25 (12-60)	Loamy sand	Alder Beech
Hørhave Bæk	6.9 (6.7-7.0)	0.26 (0.24-0.30)	17 (7-34)	Coarse sand	Beech

Chemical Analysis

The discharge was measured each time a water sample was taken by means of an Ott current meter.

The chemical analyses were performed as follows: pH was measured in the laboratory with a PHM 85 Precision pH Meter (Radiometer) and a combination glass electrode (GK 2401 C, Radiometer). The alkalinity was measured by Gran titration. When using this method in acid water, alkalinity becomes negative implying a positive, strong acidity. The colour was measured on filtered samples (0.45 μ m membrane filters) using the platinum-cobalt method comparing the absorbance of platinum-cobalt standards with the samples at 410 nm.

Benthic Fauna

Benthos samples were taken in January 1992 within a 100 m stream reach in each of the two streams running in coniferous forest. The water samples for chemical analysis were always collected at the downstream end of the reach. Thirty-five samples were taken in each stream by stratified random sampling (Elliott 1971). Most samples were taken with a 21 cm² core sampler. Stones larger than the core were sampled by lifting them into a hand net (mesh size: 0.20 mm) into which loosened material and invertebrates were swept. All samples were preserved in 4% formalin. The projected area of each stone was estimated by drawing its outline on squared paper. In the laboratory, the samples were sorted and the invertebrates identified.

The benthic faunal data from 1992 were compared with data obtained from three small beech forest streams in December 1987 (Friberg, unpublished data). The methodology was the same as described above except from the fact that about 20 samples were taken in stream reaches approximately 20 m long.

Results

Chemical Variables

Average pH and alkalinity were low and water colour was high in the two coniferous forest streams, Rye Nørreskov, and Vesterskov, compared with the four streams running in deciduous forest (Table 1). All three variables showed a clear seasonal trend in the two coniferous streams: pH and alkalinity increased during the summer period while water colour decreased. No seasonal trend was observed in the four deciduous streams with regard to these three variables.

There was a clear negative relationship between discharge and pH in Rye Nørreskov and Vesterskov during the sampling period (Fig. 1). There was also a clear positive relationship between water colour and discharge in these streams (Fig. 1). A similar relationship between pH or water color, and discharge was not found in the four deciduous streams, even though they exhibited comparable discharge fluctuations during the sampling period.

A clear pH gradient was found in Rye Nørreskov in January (Fig. 2) during a period of high precipitation. Near the spring source, pH fell from about neutral to 3.7 over a stream length of only 1500 m. There were no major tributaries on this stream reach.

Biological Variables

The macroinvertebrate fauna in Rye Nørreskov and Vesterskov was dominated by Plecoptera (Table 2), representing 87% and 63% of the total fauna, respectively. Few other species were present, mainly Diptera and the caseless caddis-larvae *Plectonemia conspersa*. The absence of Amphipoda and Ephemeroptera in Rye Nørreskov and Vesterskov contrast sharply with the macroinvertebrate assemblages found in the three neutral beech forest streams (Table 3); Diptera and Trichoptera were less abundant in the

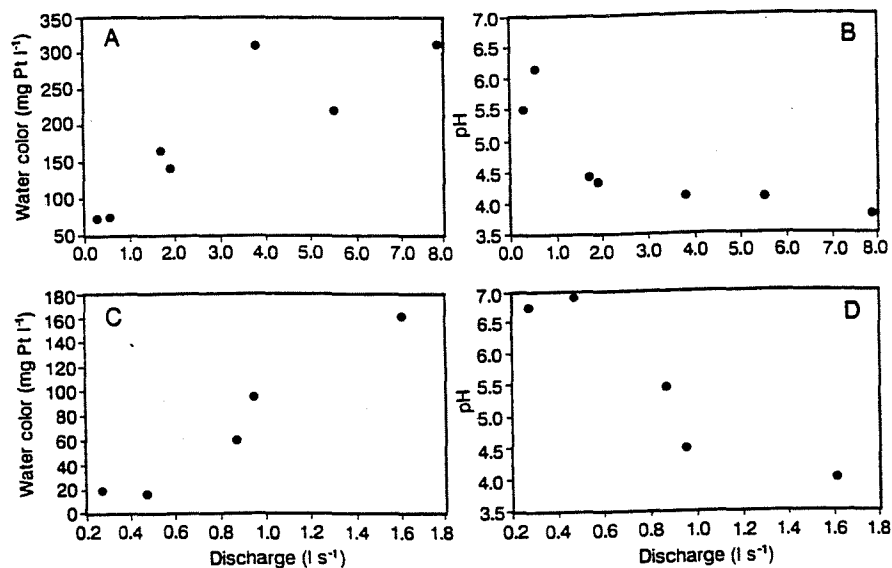


Fig. 1. Relationship between discharge and water colour/pH in stream Rye Norreskov (A/B) and stream Vesterskov (C/D).

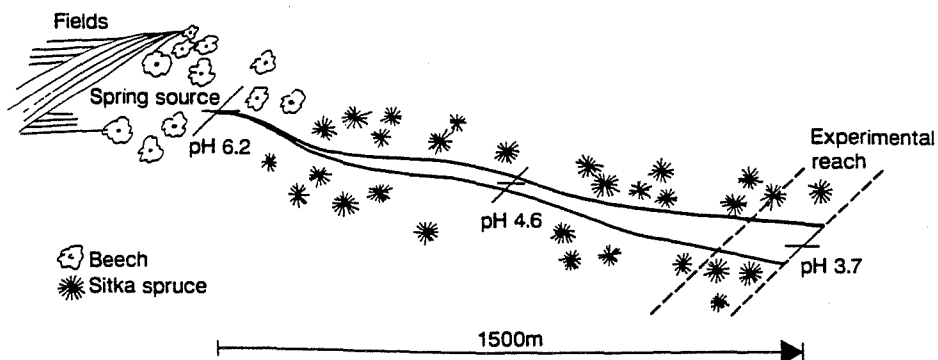


Fig. 2. pH - gradient found in stream Rye Norreskov in January; from spring source to the experimental reach.

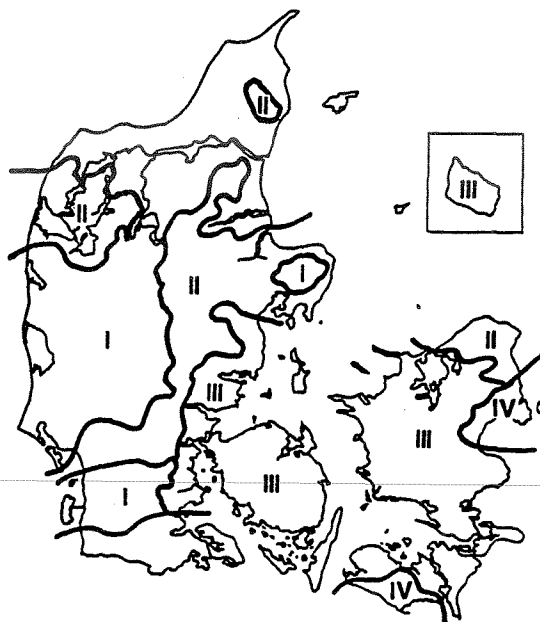


Fig. 3. The dominating soil types in Denmark: (I): coarse to fine sand, (II): loamy sand, (III): sandy loam, (IV): clay.

Table 2. Species composition in two Danish acidic forest streams.

Species	Rye Nørreskov		Vesterskov	
	January 1992 Average number·m ⁻²	+/- 95% C.L.	January 1992 Average number·m ⁻²	+/- 95% C.L.
Plecoptera	86.8%		63.4%	
<i>Leuctra hippopus</i>	13	26		
<i>Leuctra nigra</i>	1513	1525	1129	789
<i>Nemoura flexuosa</i>	78	96	54	87
<i>Nemurella picteti</i>	517	695	585	481
<i>Amphinemura sp.</i>	13	26		
Trichoptera	1.6%		4.4%	
<i>Plectrocnemia conspersa</i>	39	58	109	80
<i>Limnephilidae</i>			14	28
Diptera	7.9%		24.9%	
Chironomidae			14	28
Tanytarsini			27	38
Orthocnemiinae	26	37	381	306
Tanypodinae	13	26	68	70
Simuliidae	26	37	109	119
Eriopterinae	13	26	14	28
Hexatominiae	116	95	82	84
Remaining species	3.7%		7.3%	
Naididae	39	58		
Dysticidae			14	28
Oligochaeta			14	28
Enchytraidae	26	37	177	190
Pisidium sp.	13	26		
Velia sp.	13	26		
Total number of individuals·m ⁻²	2456		2789	

acidic streams compared to the neutral streams. Plecoptera were a relatively unimportant faunal component of the neutral streams, but their absolute number were comparable to what was found in Rye Nørreskov and Vesterskov.

Overall macroinvertebrate abundances in acidic streams were a factor 4 to 10 lower than in the neutral streams (Tables 2 and 3).

There were no fish present in Rye Nørreskov and Vesterskov, while trout (*Salmo trutta*) was present in the three neutral beech forest streams.

Discussion

A clear difference in pH, and consequently, faunal composition, was found between streams running in coniferous forest and streams running in deciduous forest. Alkalinity and soil type alone can not explain the low pH found in coniferous forest streams. Hørhave Bæk, a beech forest stream (Table 1), has low alkalinity and coarse sand as the dominant soil type, like Rye Nørreskov and Vesterskov, but it is not acidic.

The fact that coniferous forests tend to acidify soils, is already known from terrestrial research in Denmark. A pH of 3.7 has been found in the upper soil column under a Danish sitka spruce forest (Nielsen, pers. comm.). Therefore, the close linkage between discharge and pH (or water colour) in Rye Nørreskov and Vesterskov suggests that the streams are acidified as water percolates through the upper soil column to the streams, either as direct runoff or via water-filled ditches, in acidic soils, filled during periods of high precipitation. In summer, stream water is derived from upstream springs with medium alkalinity. Summertime pH reaches neutral values and water colour drops as the amount of leached organic compounds decreases. On the basis of the data presented, it seems reasonable to suggest that the streams in Rye Nørreskov and Vesterskov would remain neutral year round if the forest type had been deciduous instead of coniferous.

The fauna is clearly affected by the low pH. Abundances of the overall community in acidic streams is low compared to what is found in neutral streams. Acidic stream fauna is dominated by Plecoptera. A similar faunal structure in response to acidified water has been found by Rosemond et al. (1992) and Weatherley et al. (1989). The consequences of low invertebrate density to the ecosystem is a reduction in the amount of food available at higher trophic levels and the turnover rate of the detritus pool (Burton et al. 1985). Furthermore, no fish are able to survive in the acidic forest streams, but both Rye Nørreskov and Vesterskov, if neutral, could sustain trout (*Salmo trutta*) spawning and nursery areas.

The dominant soil types in Denmark are shown in Fig. 3. The Weichsel glaciation divided Jutland into two regions: east of the Weichselian terminal moraine, where soils are usually clayey and calcareous (normally high alkalinity), and west of the terminal moraine where soils are sandy and leached (lower alkalinity) (Rebsdorf et al. 1991). Today, most of Jutland is used for agricultural purposes, but as farming is reduced in the near future, it must be expected that the poor sandy soils, being less profitable, will be abandoned first. These former agricultural areas in western Jutland are potential sites for afforestation. The results from this study suggest that extensive coniferous afforestation (using sitka spruce and perhaps other coniferous species as well) in this region of Denmark could result in acidification of streams with medium to low alkalinity. Such an afforestation will have a profound impact on the ecosystem of these streams.

The data presented in this paper are from an ongoing study. More work will be needed to finally verify the conclusions and to quantify the importance of organic versus inorganic acids in the acidification of streams. In order to assess the relative importance of "natural" and anthropogenic sources of acidification.

Table 3. Group percent composition, estimated abundance·m⁻² and associated 95% confidence limits from Benthic communities in three small neutral beech forest streams (unpublished data from a previous study).

	Station 1	+ 95% C.L.	Station 2	+95% C.L.	Station 3	+95% C.L.
Amphipoda	30%		8%		22%	
<i>Grammarus pulex</i> L.	3010	[2015] [4500]	1270	[840] [1920]	5395	[2795] [10420]
Plecoptera	14%		7%		1%	
<i>Amphinemura</i> sp.	250	[205] [310]	320	[250] [405]	23	[20] [25]
<i>Nemoura flexuosa</i> Aub.	50	[45] [55]	115	[95] [135]	115	[100] [135]
<i>Leuctra hippopus</i> Kempny.	455	[345] [595]	475	[364] [625]	45	[40] [50]
<i>Leuctra nigra</i> Oliv.	635	[435] [915]	180	[150] [225]	45	[40] [50]
Ephemeroptera	8%		7%		3%	
<i>Baëtis rhodani</i> Pict.	810	[545] [1210]	1090	[740] [1605]	630	[455] [870]
Diptera	14%		57%		51%	
	1415	[965] [2080]	9235	[5155] [16500]	12329	[7560] [20100]
Tricoptera	17%		11%		8%	
	1720	[1200] [2470]	1860	[1195] [2900]	1885	[1305] [2725]
Remaining species	17%		10%		15%	
	1670	[1100] [2535]	1590	[1075] [2350]	3535	[2300] [5440]
Total: percent	100%		100%		100%	
number·m ⁻²	10015		16135		24002	

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References

- BORG, H., G.L. STONEMAN, and C.G. WARD. 1988. The effect of logging and regeneration on groundwater, streamflow, and stream salinity in the southern forest of Western Australia. *J. Hydrol.* 99: 253-270.
- BURTON, T.M., R.M. STANDFORD, and J.W. ALLAN. 1985. Acidification effects on stream biota and organic matter processing. *Can. J. Fish. Aquat. Sci.* 42: 664-675.
- CUMMINS, K.W., A. WILZBACH, D.M. GATES, J.B. PERRY, and W.B. TALIAFERRO. 1989. Shredders and riparian vegetation. *BioScience* 39 (1): 24-31.
- ELLIOTT, J.M. 1971. Some methods for the statistical analysis of samples of benthic invertebrates. *Freshwat. Biol. Assoc. Scientif. Publ.* 25.
- FRIBERG, N. and K. KJELDSSEN. (1994). Development of benthic algal biomass in six Danish beech, mixed and coniferous forest streams. *Verh. Internat. Verein. Limnol.* In press.
- JENKINS, A., B.J. COSBY, R.C. FERRIER, T.A.B. WALKER, and J.D. MILLAR. 1990. Modelling stream acidification in afforested catchments: an assessment of the relative effects of acid deposition and afforestation. *J. Hydrol.* 120: 161-181.
- REBSDORF, A., N. THYSSEN, and M. ERLANDSEN. 1991. Regional and temporal variation in pH, alkalinity and carbon dioxide in Danish streams, related to soil type and land use. *Freshwat. Biol.* 25: 419-435.
- ROSEMUND, A.D., S.R. REICE, J.W. ELWOOD, and P.J. MULHOLLAND. 1992. The effect of stream acidity on benthic invertebrate communities in south-eastern United States. *Freshwat. Biol.* 27: 193-209.
- WEATHERLEY, N.S., G.P. RUTT, and S.J. ORMEROD. 1989. Densities of benthic macroinvertebrates in upland Welsh streams of different acidity and land use. *Arch. Hydrobiol.* 115 (3): 417-431.

ACIDIFICATION AND POLLUTION BY HEAVY METALS OF KOLA SURFACE WATERS

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Abstract

Precipitation falling on the northern part of the Kola Peninsula is severely polluted by strong acids and heavy metals, due to emissions from large local industries. Large areas in the affected region are sensitive to acid precipitation, due to local climate and geology. Based on extensive chemical analyses of lake and river water samples collected in the region in 1989-1992, it was demonstrated that extensive surface water acidification has taken place, particularly in the mountains and remote tundra regions. In the industrial centre (within 30 km of the emission sources) there was an observed increase in buffering capacity due to the compensatory deposition of alkaline dust, which contains high concentrations of nickel and copper are observed. Because of the severity of the pollution, the northern Kola Peninsula offers a unique opportunity to study the ecological effects of combined acidification and heavy metal deposition.

Key words: C.I.S., Russia, Kola Peninsula, LRTAP, Acid Rain

Introduction

The Kola Peninsula lies in the north-eastern part of Fennoscandia and is located inside the Arctic Circle. The phenomenon of surface water acidification is well known in Scandinavian countries like Norway, Sweden, and Finland (SFT 1987; Monitor 1986; Forsius et al. 1988). Up to 1989, the Kola North remained an unstudied territory in regards to the evaluation of water acidification. However, previous to this investigation, the area had been exposed to the acidifying influences of emissions from the "Pechenganickel" and "Severonickel" companies, the biggest smelters in the area. Annual emission of sulphurous gases from these two sources is 568 000 tonnes. Along with the acidic impact, Kola North surface waters are being polluted by heavy metals from the same industrial sources. It is well known that the toxicity of heavy metals for the biota is enhanced under acidic conditions.

In 1989-1992 special research on the Kola Peninsula territory was conducted. The goal was to show the geographic extent and intensity of surface water acidification and pollution by heavy metals.

Materials and Methods

The study contained the following components:

1. An examination of small lakes, covering a broad geographic area, for the period September 20 to October 10, 1989-1991 (a total of 284 lakes were investigated). Lakes were selected and sampled according to the Norwegian criteria outlined in SFT (1987).
2. A documentation of acidic episodes on rivers during flood period. Intensive daily observations on water quality variations in the flood period were conducted in a small mountain stream, on the runoff from the lake (3 km²), and on runoff from marsh with a high content of humic acids.
3. A study of buffering capacity in the big lake-river systems in the territorial sections. Data analysis provided by Murmansk regional authority on hydrometeorology and environment control (1953-present) provided the basis for a review of historical trends.
4. A calculation of acidification and its exceedance was done according to the method of A. Henriksen et al. (1990).
5. Heavy metal toxicity and behaviour in acidified waters was investigated.

Water sampling and analysis are performed according to the usual hydrochemical methods (Rond et al. 1975). Samples were analysed at the Institute of North Industrial Ecology Problems (INEP), Norwegian Institute for Water Research (NIVA)[140 samples] and the Water and Environment District Office of Lappland (LAVI)[100 samples]. The following components were determined: pH, major ions (Ca, Mg, K, Na, SO_4 , HCO_3 , Cl), organic matter, turbidity, colour, and metals (Ni, Co, Cu, Zn, Al).

Results and Discussion

Deposition of Sulphur, Nickel and Copper

According to the data of T. Makarova (1989), the present average regional level of anthropogenic sulphur deposition on the Kola Peninsula is approximately $0.7\text{--}1.0 \text{ g S}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$. Deposition values in the industrial regions in the north-west and central parts range from $3\text{--}10 \text{ g S}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$, except near the local sources where values may reach as much as $30 \text{ g S}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$. In the western parts of Finland sulphur deposition is $0.2\text{--}0.6 \text{ g S}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ and in the eastern part, near the border area of the Murmansk region values are $1.0\text{--}1.2 \text{ g S}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ (Kryuchkov and Makarova 1989; Hoem and Joranger 1988).

The average pH of precipitation in northern Fennoscandia is between 4.5 and 4.7. According to the data by T. Makarova (1989) the pH of the precipitation in the eastern part of the Kola Peninsula is 4.7. Around industrial sites where the pH of precipitation has been measured as low as 3.4, periodic episodes of alkaline precipitation up to pH 7.6 have also been observed. High pH values are due to emission of alkaline dust particles.

Dust particle emissions from major industrial sources also contain heavy metals which constitutes a major source of pollution of the Kola North surface waters. The deposition of heavy metals around smelter areas is $29\text{--}310 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$ of nickel and $35\text{--}280 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$ of copper (Makarova and Ratkin 1990). Air pollution by metals is distributed more locally around the smelters than is sulphur deposition.

Sensitivity of Territory

On the basis of regional climate and geology, most waters of the northern Kola Peninsula are sensitive to acidification, especially in the northern tundra where small lakes are typical and the underlying bedrock is primarily composed of granite and basalt with little or no buffering capacity. The centre of the Peninsula is dominated by the Khibiny and Lovozero rock masses which are alkaline in nature (apatite-nepheline syenites). The smelters which contribute the most to sulphur deposition, in the sensitive regions, are located in areas with basic and ultrabasic rocks (Murmansk Region Atlas 1971). Thus, there are no local problems of acidification around the smelters.

Sulfates

Sulphate deposition in the north Kola Peninsula is the principal cause of aquatic acidification. According to the data by A. Henriksen et al. (1990), the natural sulphate content in the lakes on the western Fennoscandia should be $10\text{--}15 \mu\text{eq}\cdot\text{L}^{-1}$. Sulphate content in the north Kola Peninsula lakes is substantially higher than in comparable regions of Norway and Finland. Typically, north Kola lakes values range between $75\text{--}100 \mu\text{eq}\cdot\text{L}^{-1}$ (Fig. 1). Around industrial centres sulphate, the dominant anion in lake waters, exceeds $200 \mu\text{eq}\cdot\text{L}^{-1}$.

Lake Acidification

There is good evidence that many lakes of the north Kola Peninsula have been acidified. Overall 5.6% of lakes had a $\text{pH} < 5$ and 13.5% had $\text{pH} < 6$. More than 30% of the lakes are at risk of becoming acidified because they have $\text{ANC} < 50 \mu\text{eq}\cdot\text{L}^{-1}$ and $\text{HCO}_3^- < \text{SO}_4$. Because Kola Peninsula lakes showed

heterogeneity of water chemistry, the data were divided into 7 regional categories for analysis (Fig. 2). In the industrial regions sulphate buffering capacity of most lakes remains high ($\text{ANC} > 200 \mu\text{eq}\cdot\text{L}^{-1}$) due to basin geology and local emissions of alkaline dust. At the same time, strongly acidified small lakes ($\text{pH} < 5.0$) exist in the mountains within the limits of the regions; Kuorpukas, Chuna, Volchii and Monche. Acidification of water in the northern and eastern tundra may be eminent due to the extreme vulnerability of these territories.

Heavy Metals

Heavy metals are the most hazardous kind of water pollution in the Kola North. Toxic levels of copper and nickel concentrations are observed in the lakes within a 30 km radius of metallurgical enterprises (Fig. 3). In the Pechenga area, nickel concentration in water are often $20\text{-}30 \mu\text{g}\cdot\text{L}^{-1}$. In small acidified lakes on the Kuorpukas mountain (region, ed.)* nickel content exceeds $70 \mu\text{g}\cdot\text{L}^{-1}$. Similar heavy metal concentrations are typical for the Monchegorsk area. Background concentration of Nickel and Copper ($< 1 \mu\text{g}\cdot\text{L}^{-1}$) are found in lakes remote from the industrial centres of eastern and south-western regions: D. Zelentsy, Yenozero and Babye Lake. In industrial centres the heavy metal concentration in local waters was correlated with the sum of base cation $\text{Ca}^{2+} + \text{Mg}^{2+}$ ($r = 0.70$, $n = 45$).

Acidic Episodes

The surface water supply of sub-arctic regions is controlled by precipitation. More than half (40-60%) of the annual river runoff occurs in the spring flood period. Because acids and metals, deposited during the winter and stored in the snow pack, are suddenly released during the spring melt, fauna are exposed to the highest concentrations at this time. Typically, during spring flood, maximum $\text{Ca}^{2+} + \text{Mg}^{2+}$ concentrations in rivers of the industrial centres are $> 200 \mu\text{eq}\cdot\text{L}^{-1}$ in the Pechenga, Monchegorsk and Kovdor regions. However, in the majority of northern and eastern Kola Peninsula, buffering capacity is much less. The most severe conditions occur in areas located within a 30-100 km radius of each smelter. Here alkalinity decreases of 4-6x have been observed. In some cases alkalinity values of zero were recorded. One mountain stream even showed a pH of 4.4-4.7 over a 5-7 day period.

In rivers that have their origin in lakes or wetlands there is no rapid decrease in pH. However, there are still times when alkalinity declines to zero. The input of H^+ and metal ions (Cu, Ni, Co., etc.) in rivers coincides with the early part of the spring melt. The simultaneous occurrence of low pH and high metal concentrations (fig. 4) creates conditions that are particularly hazardous for sub-arctic fauna.

Historic data from several big rivers in the northern Kola Peninsula; Lotta, Ponoy, Varzuga, etc. were obtained from the Hydrometeorological Committee and analysed to determine if there was a long-term trend in alkalinity (Fig. 5). Data were divided among the spring melt and winter low flow periods. In both cases a negative trend of alkalinity was noted over a 30-year period for which the data were considered to be most accurate (1953-1990). It is worth noting that acidification has probably intensified since 1975 when the Severonickel company began to use, high sulphur, Norilsk ore. The observed decline in alkalinity of rivers also indicates a steady erosion of acid neutralising capacity of the watersheds over the same time period.

Critical Loads and Their Exceedance

Based on 3 deposition regimes and an ANC value of $20 \mu\text{eq}\cdot\text{L}^{-1}$, critical loads and their exceedances were calculated with the help of Tor S. Traaen (1991) [Norwegian Institute of Water Research (NIVA), Norway] according to the method of Henriksen et al. (1990). The percentages of lakes in the northern Kola Peninsula, with loadings that exceed or fall short of this critical value, are given in Fig. 6.

*

The editors are assuming that the author is referring to the entire Kuorpukas mountain region, not a single mountain.

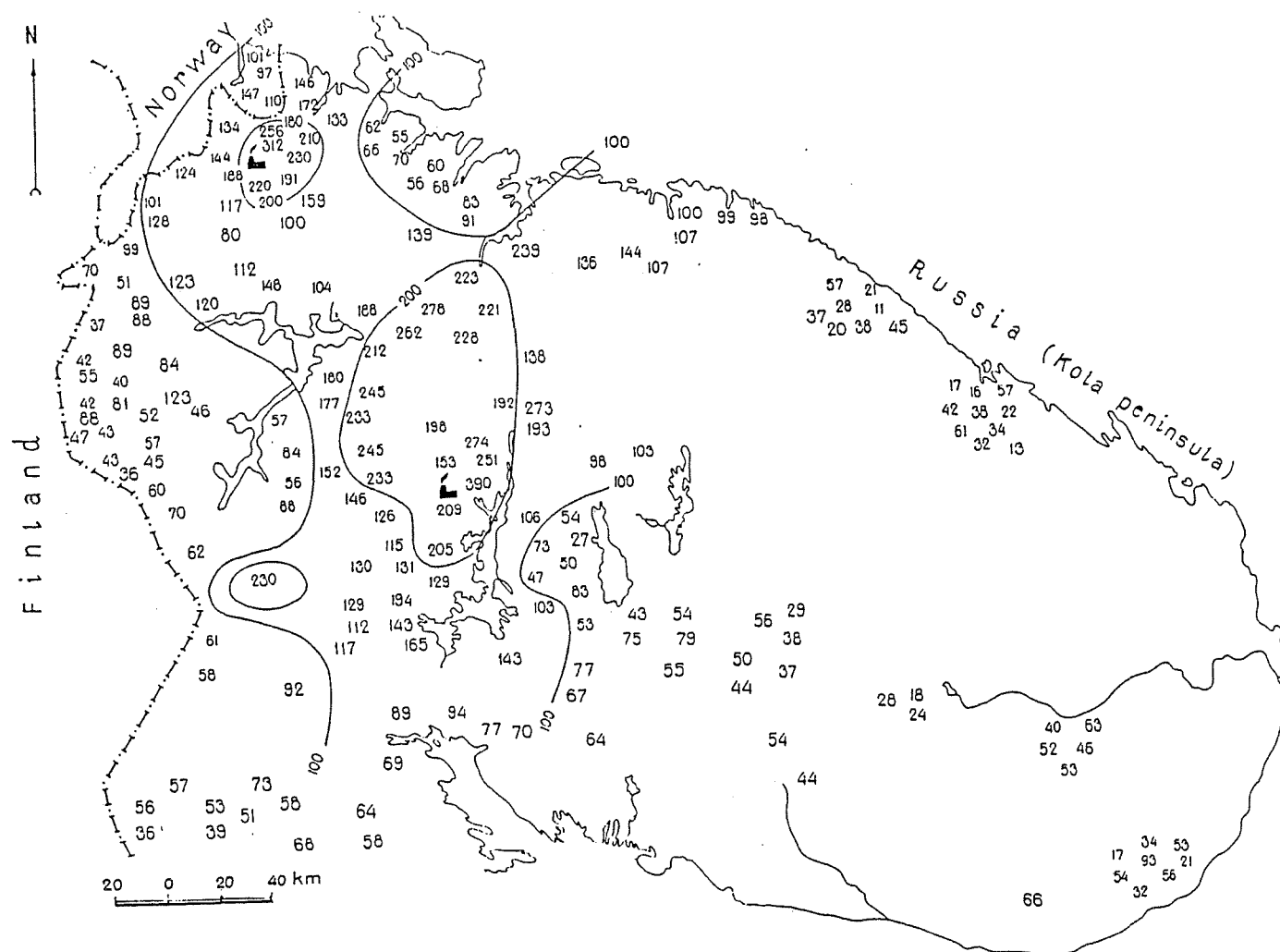


Fig. 1. Average sulphate content ($\mu\text{eq}\cdot\text{L}^{-1}$), after sea salt correction, in fall samples from lakes in the Kola region (1989 - 1992).

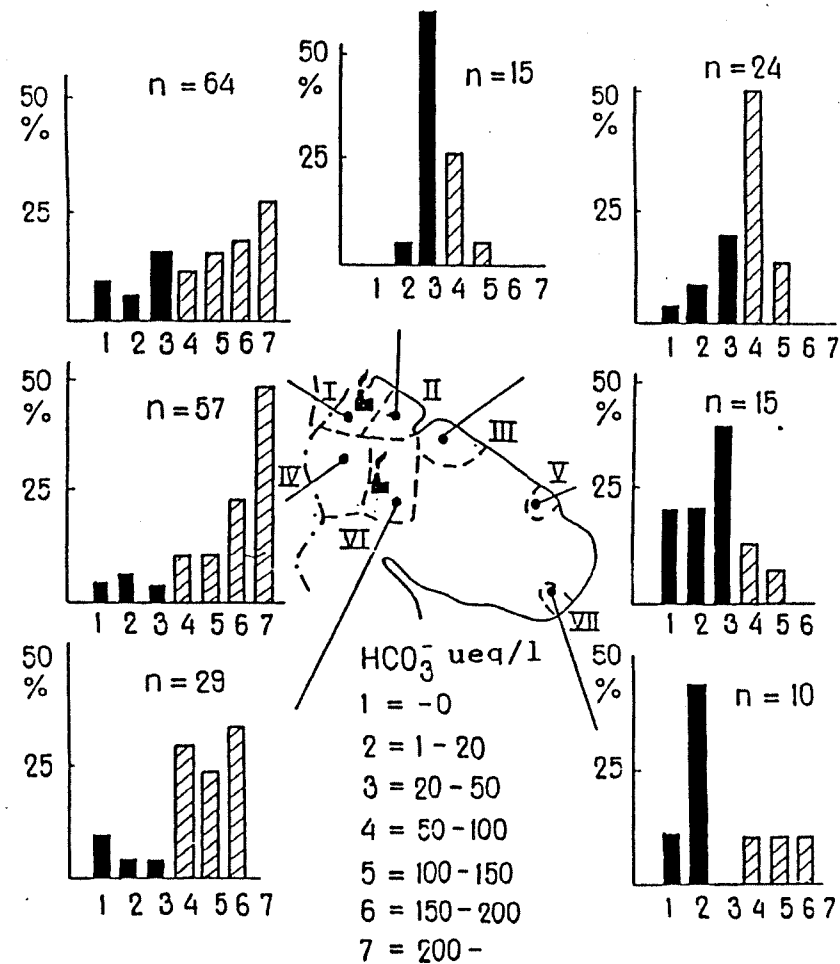
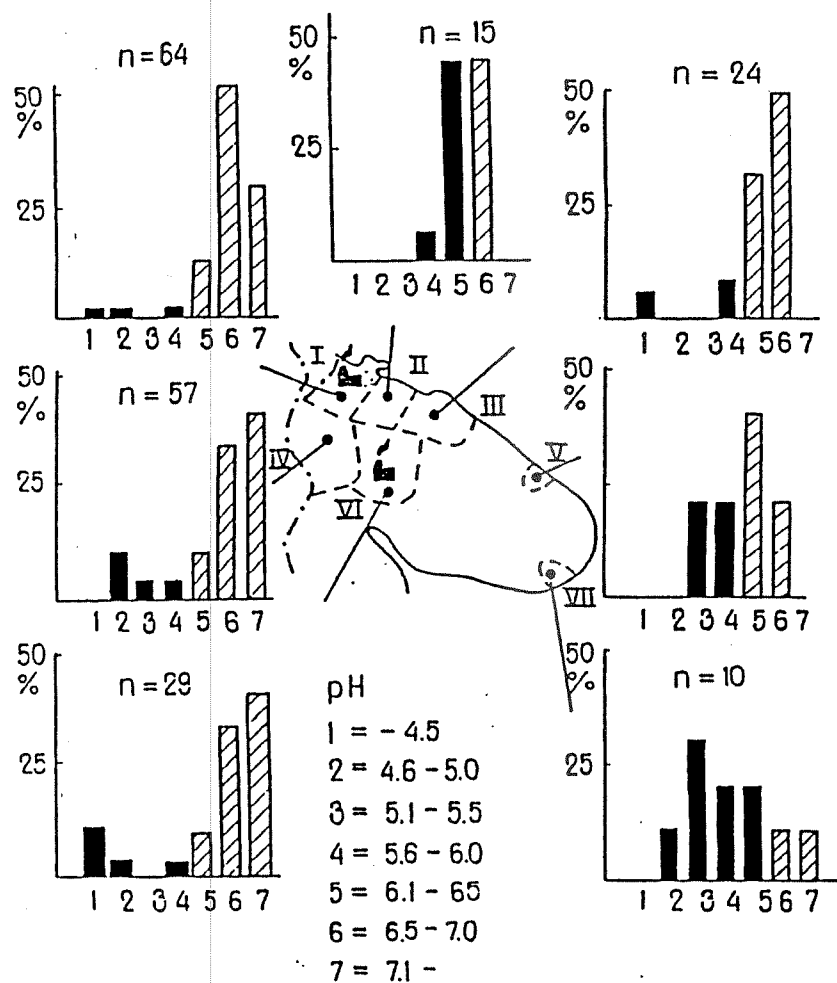


Fig. 2. Autumn pH and alkalinity values, from lakes of different areas, within the Kola region.

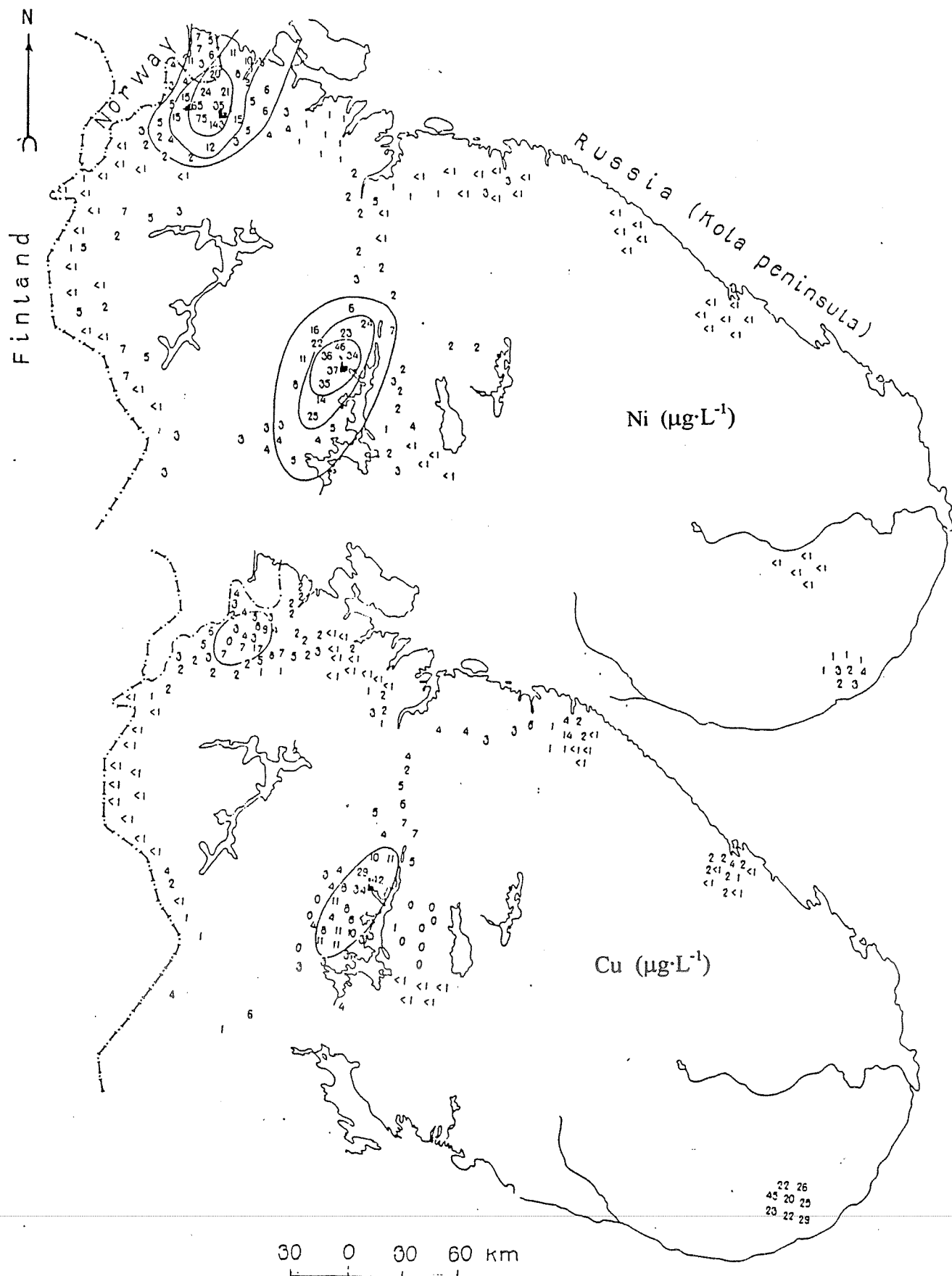


Fig. 3. Autumn Nickel and Copper content ($\mu\text{g}\cdot\text{L}^{-1}$) in lake-waters from the Kola region (1990 -1993).

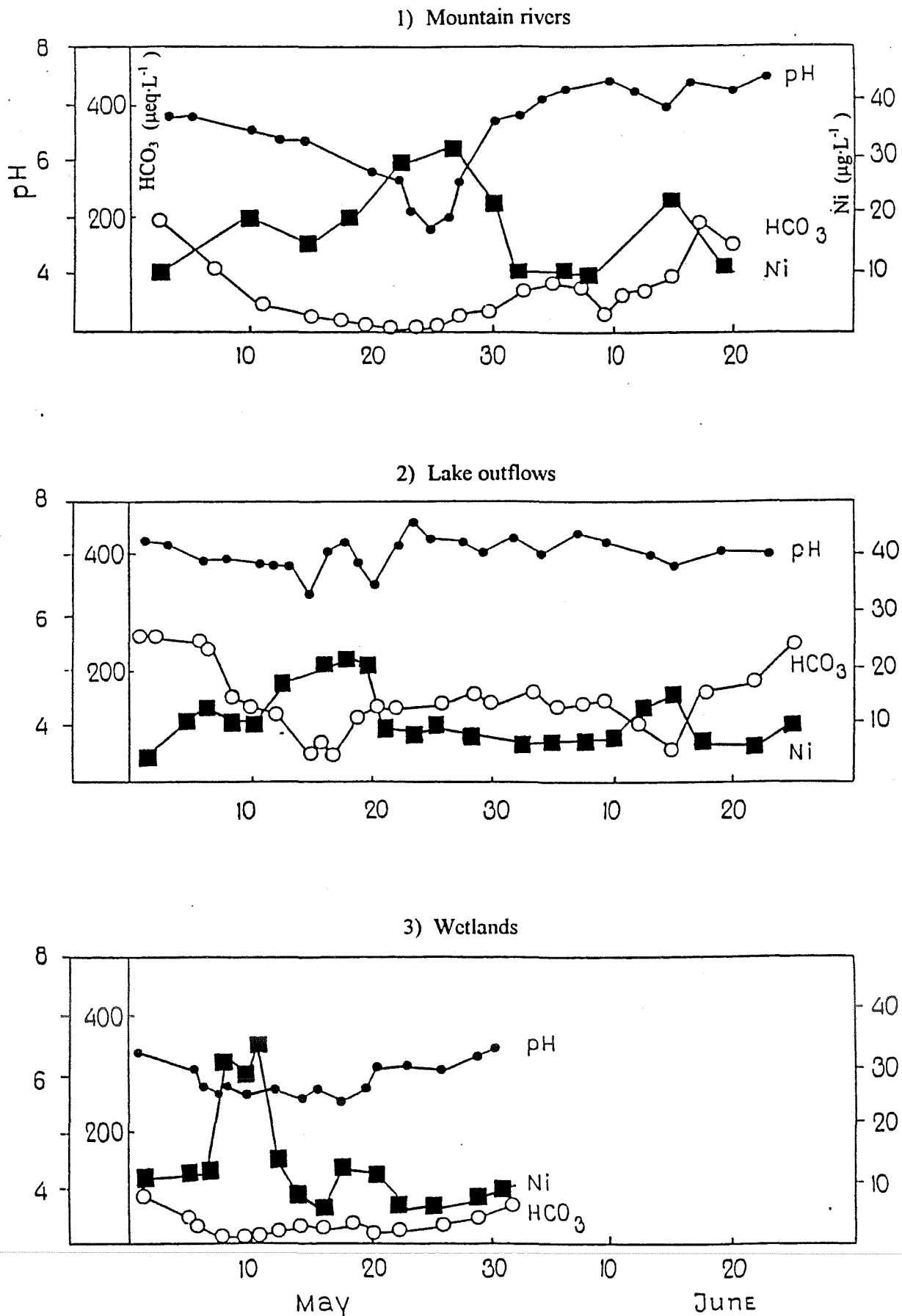


Fig. 4. Episodic changes in pH, HCO_3 and Ni concentrations during the flood time in the Chunozero water catchment (approximately 30 km south of the smelter "Severonickel"): 1-Mountain rivers, 2-Lakes outflow, 3-Wetlands

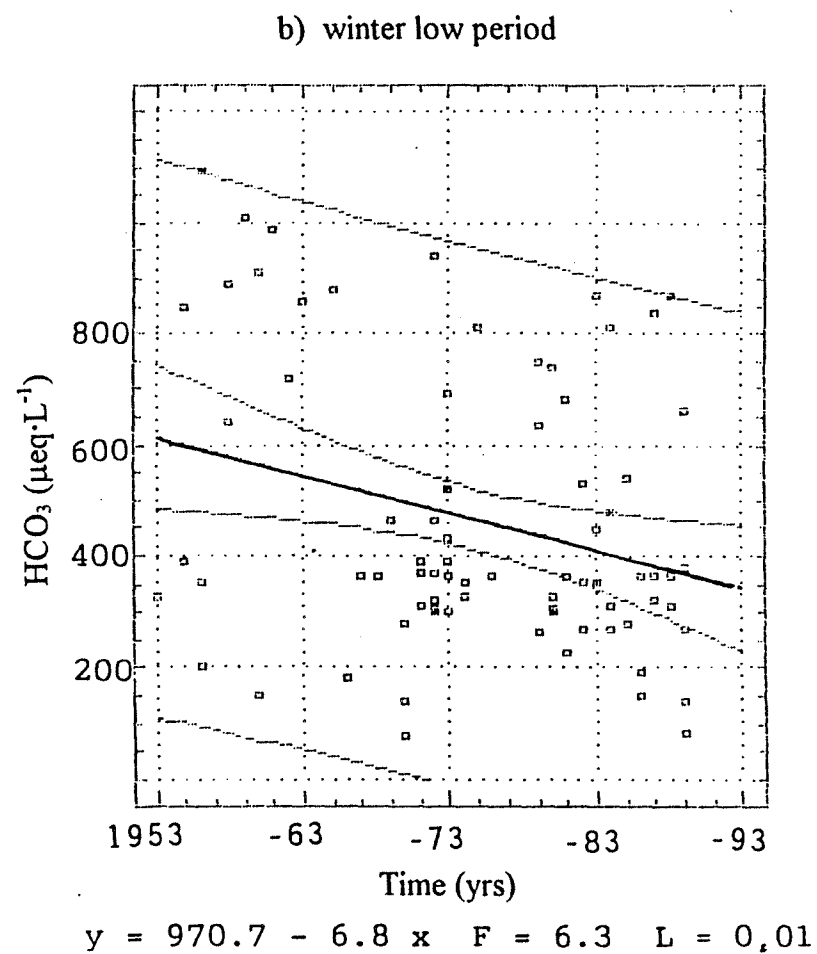
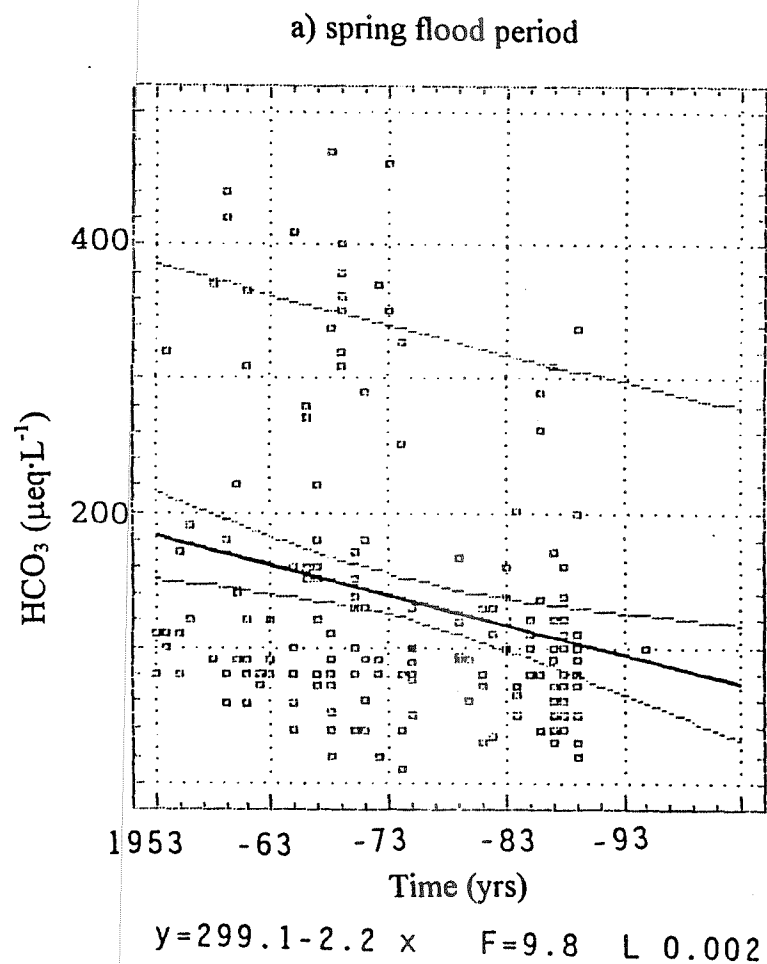


Fig. 5. Long-term trend of alkalinity decreases in the Kola North big rivers (Lotta, Ponoy, Ura, and Teriberka): a) spring flood period, b) winter low period

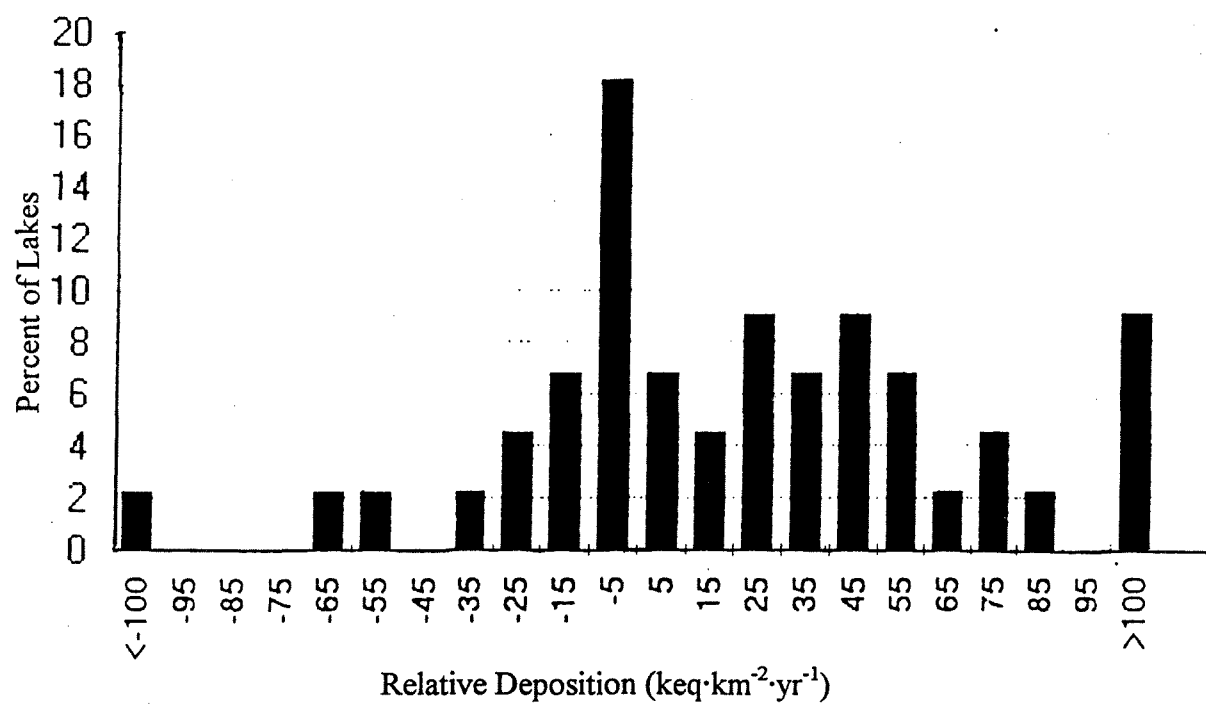


Fig. 6. Percent distribution of lakes in the Kola Peninsula relative to the critical load required to maintain $\text{ANC} > 20 \mu\text{eq}\cdot\text{L}^{-1}$, exceedance indicated by negative values.

Conclusion

Most surface waters in the northern Kola Peninsula are sensitive to acidification. Currently, acidic deposition in the region is much higher than for comparable and adjacent areas of the arctic and sub-arctic due largely to the presents of two major ore smelters. Sulphate content (after correction for the sea salt) of northern Kola Peninsula lakes is typically 75-150 $\mu\text{eq}\cdot\text{L}^{-1}$.

Of the 284 lakes surveyed, 13.5% were $< \text{pH } 6$ and 5.6% $< \text{pH } 5$; about 30% were in danger of becoming acidified (alkalinity $< 50 \mu\text{eq}\cdot\text{L}^{-1}$). Acidified lakes occurred mainly in the mountains (Kourpuukas, Chuna, Volchij, Moncha-tundra) and in remote regions where the inherent sensitivity of the territory to acid loading was high. In the industrial centres where high sulphate ($> 200 \mu\text{eq}\cdot\text{L}^{-1}$) and heavy metal ($\text{Ni} > 20 \mu\text{g}\cdot\text{L}^{-1}$; $\text{Cu} > 10 \mu\text{g}\cdot\text{L}^{-1}$) concentrations were observed, ANC was also high ($> 100 \mu\text{eq}\cdot\text{L}^{-1}$) due to the ameliorative effect of alkaline dust emissions from local enterprises.

During spring melt a critical situation is generated in rivers situated 30-100 km from the smelter complexes. Considerable erosion of alkalinity is observed ($\text{HCO}_3^- < \text{SO}_4^{2-}$) and acidic episodes have been recorded for a number of mountain rivers. The period of low pH in rivers (4.4-4.7) only occurs for 5-7 days, but may be particularly hazardous to fauna since it occurs in conjunction with high metal concentrations.

Long-term data sets (1953 -1990) showed a tendency for buffering capacity to decrease in several big rivers (Lotta, Ura, Varzuga and Ponoy). This indicates that major transformations of whole catchment areas may be occurring due to the influence of acid precipitation.

Measurements of sulphur critical loads in surface waters show that loading exceeds critical values in a number of regions, including remote eastern parts of the northern Kola Peninsula.

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References

- Forsius, M., K. Kinnunen, P. Kortelainen, I. Kaemaeri, I. Mannio, and M. Verta. 1988. Jarvien happamoituminen etenee. (Summary in English: The acidification state of Finnish lakes). Suomen Luonto 47(4): 46-51 (in Finnish).
- Henriksen, A., I. Kaemaeri, M. Posh, G. Loevblad, M. Forsius, and A. Wilander. 1990. Critical loads to surface waters in Fennoscandia. Nord. 124: 32 p. + App.
- Hoem, K. and E. Jorlander. 1988. Precipitation and air chemistry at the Norwegian background stations. NILU TZ; 1/1988.
- Keller, W., and I.R. Pitblado. 1986. Water quality changes in Sudbury area lakes: a comparison of synoptic surveys in 1974-1976 and 1981-1983. Water, Air and Soil Pollution, 29: 285-296.
- Kryuchkov, V. and T. Makarova. 1989. Aerotechnogenic impact on the Kola North ecosystem. Apatity, Edition of Kola Sciences Centre USSR Academy of Sciences. 96 p. (in Russian).
- Makarova, T. and N. Ratkin. 1990. Complex ecological-geographical forecast of Kola Peninsula environment condition. Report. Fonds of Kola Sciences Centre, Apatity. 40 p. (in Russian).

- Monitor. 1986. Sura och foersurade vatten. Naturvardsverket informerar, 180 p. In C. Bernes (ed.). Schmidts boktryckeri AB, Helsingborg. (in Swedish).
- Rond, M.C., A.E. Greenberg, and M.I. Taras (ed.). 1975. Standard method for examination for water and waste water. Fourteenth edition. USA. 1195 p.
- SFT. 1987. The Norwegian State Pollution Control Authority. 1000 lake survey Norway 1986. Report 283/87. Oslo, Norway. 33 p. + App.
- Traaen, T., T. Moiseenko, T. Dauvalter, S. Rognerud, A. Henriksen, and L. Kudravserva. 1991. Acidification of surface waters, nickel and copper in water and lake sediments in the Russian-Norwegian border areas. Progress reports for 1989-1990. Working Group for Water Environmental Protection Commission. Oslo (NIVA) and Apatity (INEP), 20 p.

WATER QUALITY TRENDS IN QUÉBEC LAKES

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Abstract

In the Laflamme Lake Watershed (LLW), results of trends analysis in precipitation and water chemistry as they relate to the effects of acidic deposition are reported for the 1981 to 1991 period. In precipitation, decreases in SO_4^{2-} concentration and increases in NO_3^- concentration were observed. In lake water, while concentrations of ANC and Ca^{2+} decreased gradually, SO_4^{2-} and NO_3^- patterns reversed around 1986, resulting in no monotonic trend for SO_4^{2-} concentrations over the combined 11 year period and a decreasing trend for NO_3^- concentrations. In some of the four nearby lakes, decreasing ANC and $\text{Ca}^{2+} + \text{Mg}^{2+}$ trends were also observed but no overall trends were detected for SO_4^{2-} , NO_3^- or pH.

The analysis of temporal trends in water chemistry was extended to 42 lakes in southern Québec over the December 1984 to December 1991 period. The predominant trend observed was a decrease in lake SO_4^{2-} concentrations. For 16 of the 31 lakes, decreasing trends in SO_4^{2-} were accompanied by decreasing $\text{Ca}^{2+} + \text{Mg}^{2+}$ (10 lakes), decreasing $\text{Ca}^{2+} + \text{Mg}^{2+}$ and ANC (2 lakes: dilution) or increasing ANC (4 lakes: recovery). One lake exhibited a significant increase in SO_4^{2-} concentrations and a significant decrease in ANC (acidification).

Key words: Laflamme Lake Watershed (LLW), SO_4^{2-} , NO_3^- , ANC, acidification

Introduction

In the province of Québec, there are about 900 000 waterbodies many of which are subject to acid deposition. The effects of acid precipitation on aquatic ecosystems are monitored through various networks. A provincial agency operates a spatial lake acidity monitoring network and a biological monitoring network which covers the area north of the St. Lawrence River, south of the 51°N parallel, and between the 62°W meridian and the Ottawa River to the east. Federal agencies operate a temporal lake acidity network (LQN), a calibrated watershed (Laflamme Lake Watershed - LLW), and a biomonitoring project. The combined information from these projects produces a comprehensive view of acidification in the province.

It is estimated that 52% of average-sized lakes (10-2 000 ha, $N = 31\ 808$) in the area being monitored have pH values at or below 6 and that 19.3% of lakes have pH values at or below 5.5 (Dupont 1992). Most lakes on the Canadian Shield, except for a small area north of Ottawa, the Abitibi lowlands, and the Saguenay, have very low acid neutralizing capacities (below $10\ \text{mg}\cdot\text{L}^{-1}$ or $200\ \text{meq}\cdot\text{L}^{-1}$). During the 1980's, lakes received wet SO_4^{2-} loadings of $10\text{-}40\ \text{kg}\ \text{SO}_4^{2-}\cdot\text{ha}^{-1}$. Deposition of SO_4^{2-} declines along a north-easterly gradient as distance from the major sources of SO_2 , located to the south and west, increases. In 1991, lake water SO_4^{2-} concentrations in the LQN network ranged from $1.3\text{-}10\ \text{mg}\cdot\text{L}^{-1}$ showing the same gradient that was found in wet SO_4^{2-} loading. This article looks at temporal trends in water quality at some specific sites and regionally across southern Québec.

Material and Methods

The LQN network was established to document trends in surface water chemistry. It is made up of 64 headwater lakes that lie on the Canadian Shield. Time series data are only available for the 41 lakes west of the Saguenay River. Lakes in the study area generally follow a pattern of decreasing mineralization and

increasing sensitivity to acid precipitation from west to east. Most of the territory is associated with shallow glacial tills overlying silicate bedrock (amphibolite, granite, or gneiss). The soils in the watersheds are shallow, have a very low carbonate content, and are generally podzolic in nature. The vegetation is either made up of species typical of the boreal forest (balsam fir, spruce, and white birch) or species typical of the southeastern mixed forest (pine, hemlock, and yellow birch). Lake area ranges from 5-200 ha. The Laflamme Lake Watershed area is located north of Québec City at about the midway point of the LQN network. The chemistry of the four LQN network lakes near LLW (Bonneville, Veilleux, Macleod and Josselin lakes) is dominated by Ca^{2+} and SO_4^{2-} with a total ionic concentration of about $200 \text{ meq}\cdot\text{L}^{-1}$ and pH values below 6.2. LLW is a Ca^{2+} and HCO_3^- dominated system with an ionic concentration near $400 \text{ meq}\cdot\text{L}^{-1}$ and a pH near 6.4 (Papineau and Haemmerli 1992).

Water sampling began in 1981 at LLW on a weekly basis. Helicopter surveys of the LQN network lakes were started in May 1983. Temporal sites were sampled every two months while spatial sites were sampled once or twice a year. The sampling protocol and database for the LQN network are documented in Dubois et al. (1992). Chemical analyses were conducted using standard Environment Canada procedures (Anonymous 1984). Samples were analysed for major cations (Ca^{2+} , Mg^{2+} , Na^+ , K^+), major anions (SO_4^{2-} , Cl^- , NO_2^- , NO_3^-), conductivity, pH, alkalinity (Gran and total), color, silicates, ammonium ion, dissolved organic and inorganic carbon, and heavy metals (Al, Mn, Fe). Charge balance and theoretical conductivity as well as the relationship to historical limits, were all considered prior to acceptance of analytical results.

Daily precipitation data came from the Montmorency station of the CAPMoN (Canadian Air and Precipitation Network) network located adjacent to LLW. Mean concentrations of major ions in precipitation were volume weighted. Annual loading was obtained by summing individual events (product of daily concentration and daily precipitation amount) and corrected for the unanalysed amounts of precipitation on a monthly basis.

Trend analysis was carried out using the "Detect" software package (Cluis et al. 1988). Nonparametric tests in the software package include Kendall and seasonal Kendall tests (Hirsch et al. 1982; Hirsch and Slack 1984), the Hirsch and Slack test (Hirsch and Slack 1984), and the Lettenmaier-Spearman test (Lettenmaier 1976). The reasons for using these particular tests are given in Berryman et al. (1988). A trend was considered significant when its magnitude was large compared to the variance of the process. In the "Detect" software package, a trend is considered significant at $p \leq 0.05$.

Results

Trends in Deposition and Precipitation Over LLW

In the LLW area, precipitation is abundant and temperatures are low. Between 1981 and 1991, annual precipitation averaged 132 mm of which 44 mm arrived as snow. Volume-weighted mean pH of precipitation was 4.4. An 11 year average annual wet deposition of $19.7 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1} \text{ SO}_4^{2-}$ (range 14.4-24.6), $3.5 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1} \text{ NO}_3^-$ -N (range 2.8-4.7), and $0.47 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1} \text{ H}^+$ (range 0.36-0.62). Dry deposition was estimated to be less than 20% of total deposition at this site (Barrie and Sirois 1986). Several variables showed seasonal or long-term variations (Table I). Trend analysis was performed on monthly data, at times compiled into quarterly form if seasonality was found. From 1981 to 1985, both wet atmospheric H^+ and NO_3^- loading showed a significant increasing trend, while SO_4^{2-} and NH_4^+ concentrations showed little change. Despite a marked increase from atmospheric loading in 1988, the trend over the 1981-1991 period showed a decrease in both the concentration and loading of SO_4^{2-} and Cl^- (Fig. 1). However, NO_3^- concentrations did increase over the last decade. No significant trend was seen for pH, NH_4^+ , Ca^{2+} , or the amount of precipitation over the 11 year period.

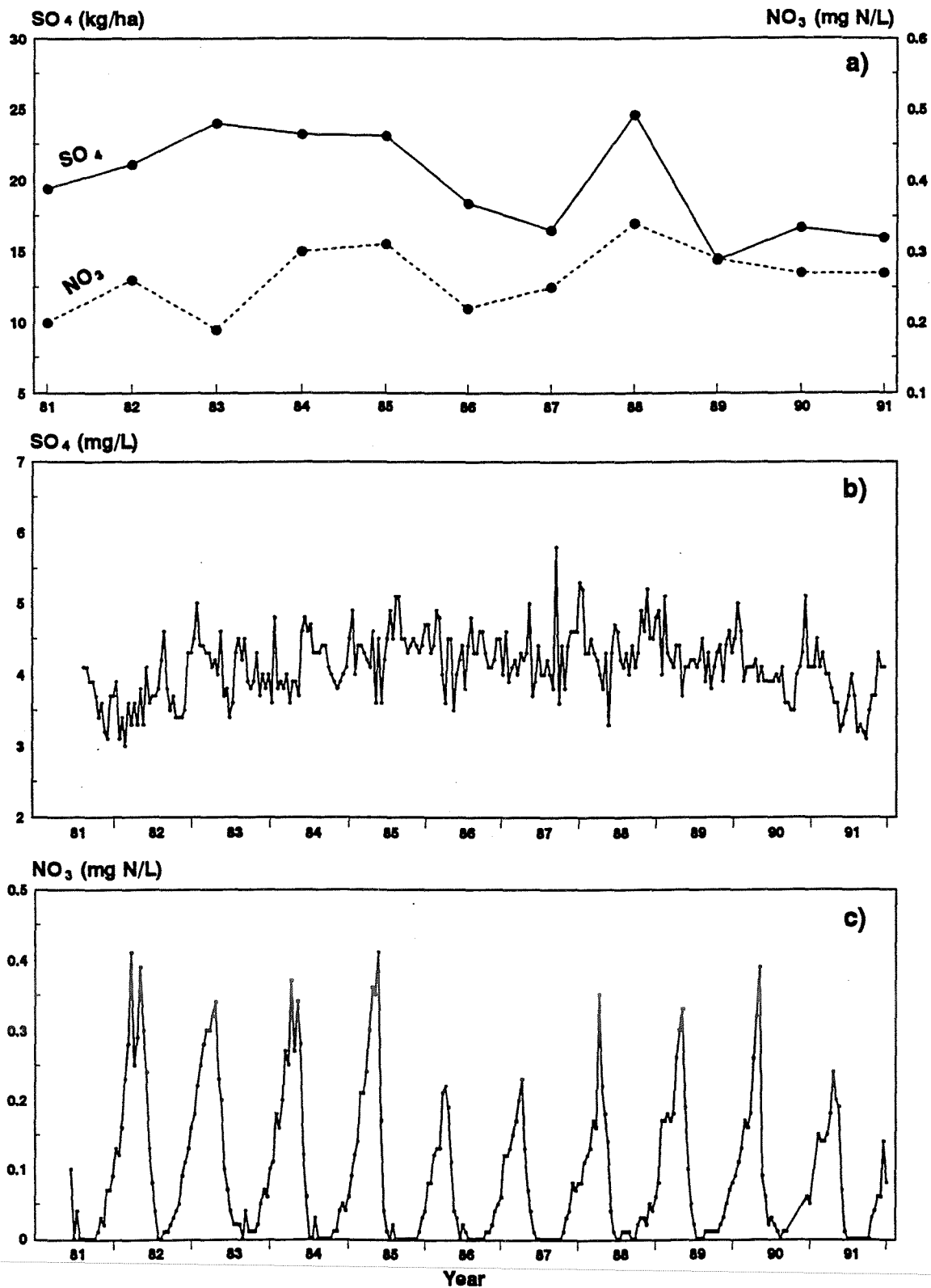


Fig. 1. Trends in sulphates and nitrates in precipitation (a) and lake water (b,c) for the Laflamme Lake watershed

Surface Water Trends For Lakes in the LLW Area

Theoretically, when acidic pollutants containing H_2SO_4 and HNO_3 enter dilute waters, the acids are first neutralised by a loss of bicarbonate ions and to some extent by the weathering of base cations. The evidence of such acidification in surface waters is detected by a loss of ANC and an increase of SO_4^{2-} concentrations, followed by an eventual decline in pH. This reaction is considered to be reversible when SO_4^{2-} inputs decrease although the pathways of recovery may not be the same as when acidification occurred.

Temporal trends in water chemistry identified at Laflamme Lake are given in Table I. For the 11 year period (June 1981 to May 1992) several decreases were observed: acid neutralising capacity (ANC) decreased by $0.89 \text{ mg}\cdot\text{L}^{-1}$ (from 5.71 to 4.82), Ca^{2+} decreased by $0.06 \text{ mg}\cdot\text{L}^{-1}$ (from 2.42 to 2.35) and NO_3^- decreased by $0.014 \text{ mg N}\cdot\text{L}^{-1}$ (from 0.083 to 0.069). Decreases in these three variables were first detected between 1986 and 1988. Mean ANC and Ca^{2+} levels were lower over the June 1986 to May 1992 period ($4.98 \text{ mg}\cdot\text{L}^{-1}$ ANC; $2.34 \text{ mg}\cdot\text{L}^{-1}$ Ca^{2+}) than in the earlier June 1981 to May 1986 period ($5.61 \text{ mg}\cdot\text{L}^{-1}$ ANC; $2.43 \text{ mg}\cdot\text{L}^{-1}$ Ca^{2+}) (Fig. 2). In the case of both SO_4^{2-} and NO_3^- , patterns reversed around 1986 (Fig. 1). After a substantial drop in NO_3^- between 1981 and 1986, values increased slowly after 1986 and an upward trend was identified for the 1986-1992 period. On the other hand, lake SO_4^{2-} concentrations exhibited a net increasing trend for many years up until 1990. From June 1981 to May 1986, this increase was from 3.62 to $4.5 \text{ mg}\cdot\text{L}^{-1}$ while from December 1984 to December 1991, concentrations decreased from 4.5 to $3.9 \text{ mg}\cdot\text{L}^{-1}$. These results show that there was no monotonic trend for SO_4^{2-} concentrations in the combined 11 year series. The highest mean annual SO_4^{2-} concentration ($4.5 \text{ mg}\cdot\text{L}^{-1}$) in lake water was measured in 1985, while the lowest mean annual NO_3^- concentration was seen in 1986 ($0.06 \text{ mg N}\cdot\text{L}^{-1}$).

For the other four lakes (Bonneville, Josselin, Macleod, and Veilleux), time series are shorter. Decreasing ANC trends were identified for Veilleux and Macleod between 1983 and 1991 (Fig. 2), but no significant trends were found for Bonneville and Josselin. A decreasing trend in $\text{Ca}^{2+}+\text{Mg}^{2+}$ was also observed at Macleod Lake over this period. No significant changes in pH, SO_4^{2-} or NO_3^- were observed over the 8.5 year period of record. Similar to LLW, the SO_4^{2-} pattern reversed in the four lakes although on different dates. At Bonneville Lake, SO_4^{2-} values increased from June 1983 ($3 \text{ mg}\cdot\text{L}^{-1}$) to April 1986 ($5.7 \text{ mg}\cdot\text{L}^{-1}$), then dropped to a level of about $4 \text{ mg}\cdot\text{L}^{-1}$ resulting in two significant trends of opposite direction. The other three lakes were sampled infrequently to permit trend analysis of data from the early years when SO_4^{2-} levels were seen to increase in Bonneville lake. Over the period December 1984 to December 1991 two important drops in SO_4^{2-} concentrations occurred (Table 1). The first of these was seen in November 1988 at Veilleux and Josselin lakes. The second in May 1990 at Macleod Lake. For the same 1984 to 1991 period, four of the five lakes in the LLW area exhibited significant decreasing trends in SO_4^{2-} concentrations of between 0.6 and $0.8 \text{ mg}\cdot\text{L}^{-1}$ (13-19%) or approximately $14 \text{ meq}\cdot\text{L}^{-1}$.

Surface Water Trends at LQN Network Lakes

The temporal trend analysis of water quality variables was extended to the 42 lakes of the LQN network located west of the Saguenay River. The December 1984 to December 1991 period was chosen for analysis for two reasons. First, over this time period all data came from the same laboratory. Second, there were indications that, after 1985, wet SO_4^{2-} loading was decreasing in both the western and eastern parts of the network. We chose to analyse the data as a separate unit to limit the number of pattern changes for a given variable. Results from the trend analysis carried out on the first five years of LQN data are reported in Bouchard and Haemmerli (1992).

Over the period (1984-1991), most lakes (33 out of 42) showed a decrease in SO_4^{2-} concentrations and only one increased. Decreases in SO_4^{2-} concentration ranged from 0.3 to $2.2 \text{ mg}\cdot\text{L}^{-1}$ or 10.1 to 32.8% (Table 1). The largest decreases are seen in the area just north of Ottawa. The lowest decreases ($< 1 \text{ mg}\cdot\text{L}^{-1}$) were seen around the LLW area and in the Pontiac (Fig. 3). In the Ottawa area, SO_4^{2-}

Table 1. Time series analysis results for LQN network.

Location	Variable	Period	N	Test	Trend	Change
Montmorency	SO ₄ ²⁻ conc	1981-91	44	S.K.	Dec	1.71 to 1.27
CAPMoN	SO ₄ ²⁻ load	1981-91	44	S.K.	Dec	1.89 to 1.39
	NO ₃ ⁻ conc	1981-91	44	S.K.	Inc	0.26 to 0.31
	pH	1981-85	60	Ken.	Dec	4.55 to 4.37
	H load	1981-85	60	Ken.	Inc	0.032 to 0.052
	NO ₃ ⁻ load	1981-85	60	Ken.	Inc	0.23 to 0.35
Laflamme	G-alk	06/81-05/92	44	S.K.	Dec	5.71 to 4.82
	SO ₄ ²⁻	07/81-05/86	59	S/L	Inc	3.62 to 4.50
	NO ₃ ⁻	06/81-05/92	44	S.K.	Dec	0.083 to 0.069
	NO ₃ ⁻	06/81-05/86	20	S.K.	Dec	0.098 to 0.075
	NO ₃ ⁻	06/86-05/92	24	S.K.	Inc	0.034 to 0.100
	Ca ²⁺	06/81-05/92	44	S.K.	Dec	2.42 to 2.35
	Ca+Mg	12/84-12/91	43	S.K.	Dec	171 to 151
	SO ₄ ²⁻	12/84-12/91	43	S/L	Dec	4.5 to 3.9
Bonneville	SO ₄ ²⁻	12/84-12/91	43	H&S	Dec	4.6 to 3.8
Veilleux	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	3.6 to 2.9
	Ca+Mg	12/84-12/91	15	Ken.	Dec	82 to 73
	G-alk	12/84-12/91	15	S.K.	Dec	1.47 to .74
Macleod	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	4.3 to 3.5
	Ca+Mg	12/84-12/91	15	Ken.	Dec	77 to 67
	Ca+Mg	06/83-12/91	18	H.S	Dec	76.6 to 68.5
	G-alk	06/83-12/91	18	Ken.	Dec	1.21 to 0.18
Josselin	Ca+Mg	12/84-12/91	15	Ken.	Inc	76 to 77
Lagou	Ca+Mg	12/84-12/91	43	S/L	Dec	86 to 74
	SO ₄ ²⁻	12/84-12/91	43	Ken.	Dec	4.5 to 4.0
Eclair	SO ₄ ²⁻	12/84-12/91	43	Ken.	Dec	4.3 to 3.6
	ANC	12/84-12/91	43	S.K.	Inc	1.7 to 2.1
Lemaine	SO ₄ ²⁻	12/84-12/91	43	S/L	Dec	5.8 to 4.6
	ANC	12/84-12/91	43	S.K.	Inc	1.0 to 1.3
Truite-Rouge	SO ₄ ²⁻	12/84-12/91	43	H&S	Dec	6.4 to 5.4
Chomeur	pH	12/84-12/91	43	S.K.	Inc	6.6 to 6.7
	Ca+Mg	12/84-12/91	43	S.K.	Inc	172 to 173
	SO ₄ ²⁻	12/84-12/91	43	S/L	Dec	4.9 to 3.7
	ANC	12/84-12/91	43	S.K.	Inc	4.9 to 5.6
Thomas	Ca+Mg	12/84-12/91	43	S.K.	Inc	230 to 235
	SO ₄ ²⁻	12/84-12/91	43	Ken.	Dec	3.6 to 3.2
Nolette	pH	12/84-12/91	43	S.K.	Dec	6.5 to 6.4
	Ca+Mg	12/84-12/91	43	S.K.	Dec	187 to 170
	ANC	12/84-12/91	43	S/L	Dec	4.9 to 3.5
Chevreuil	Ca+Mg	12/84-12/91	43	S.K.	Dec	258 to 249
	SO ₄ ²⁻	12/84-12/91	43	H&S	Dec	5.9 to 4.7
Kidney	SO ₄ ²⁻	12/84-12/91	43	H&S	Dec	7.8 to 6.3
Blais	SO ₄ ²⁻	12/84-12/91	43	S/L	Dec	8.3 to 7.2
	ANC	12/84-12/91	43	S.K.	Inc	13.5 to 14.0
David	SO ₄ ²⁻	12/84-12/91	43	S/L	Dec	7.8 to 6.4
Lake 6827	SO ₄ ²⁻	06/86-12/91	34	Ken.	Dec	5.4 to 4.7
Lake 88188	ANC	06/86-12/91	34	Ken.	Inc	1.0 to 1.2
Blériot	SO ₄ ²⁻	06/86-12/91	34	Ken.	Dec	6.3 to 6.0

Table 1. Cont'd.

Location	Variable	Period	N	Test	Trend	Change
Najoua	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	6.4 to 5.0
	Ca+Mg	12/84-12/91	15	Ken.	Dec	93 to 79
	G-alk	12/84-12/91	15	Ken.	Dec	0.07 to -0.64
Adanys	pH	12/84-12/91	15	Ken.	Dec	5.3 to 5.0
	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	5.1 to 4.3
	Ca+Mg	12/84-12/91	15	Ken.	Dec	85 to 76
	G-alk	12/84-12/91	15	Ken.	Dec	0.37 to -0.37
Fauvette	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	5.5 to 4.6
	Ca+Mg	12/84-12/91	15	Ken.	Dec	139 to 132
Boisvert	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	6.5 to 5.5
	Ca+Mg	12/84-12/91	15	Ken.	Dec	160 to 152
Thibert	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	5.9 to 4.8
Daniel	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	5.5 to 4.4
	Ca+Mg	12/84-12/91	15	Ken.	Dec	129 to 113
Belle-Truite	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	4.45 to 4.0
Pothier	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	5.1 to 4.0
Laurent	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	6.8 to 5.8
Lake 75869	pH	06/86-12/91	11	Ken.	Dec	6.4 to 5.5
	ANC	06/86-12/91	11	Ken.	Dec	1.6 to 0.1
	SO ₄ ²⁻	06/86-12/91	11	Ken.	Inc	6.6 to 7.6
Murex	pH	12/86-12/91	10	Ken.	Dec	5.3 to 5.1
Papillons	ANC	12/84-12/91	15	Ken.	Dec	4.6 to 3.4
Général-White	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	7.4 to 6.0
Grégoire	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	5.8 to 4.7
Scelier	ANC	12/84-12/91	15	Ken.	Dec	34.6 to 31.1
	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	11.1 to 9.3
Boheme	ANC	12/84-12/91	15	Ken.	Dec	27.9 to 20.4
	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	6.7 to 4.5
	Ca+Mg	12/84-12/91	15	Ken.	Dec	673 to 533
Duck	ANC	12/84-12/91	15	Ken.	Dec	21.6 to 18.6
	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	8.7 to 7.1
	Ca+Mg	12/84-12/91	15	Ken.	Dec	586 to 541
Sheridan	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	10.0 to 8.0
Clair	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	8.8 to 7.8
Graham	SO ₄ ²⁻	12/84-12/91	15	Ken.	Dec	7.8 to 5.9

- N = number of observations used in test
 S.K. = seasonal Kendall test
 Ken. = Kendall test
 S/L = Spearman/Lettenmaier test
 H&S = Hirsch & Slack test
 Inc = increase
 Dec = decrease
 Change = initial value and final value of the regression line estimated by least squares. ANC (G-alk), SO₄²⁻ and Ca²⁺ values are in mg·L⁻¹; pH in units; Ca+Mg in meq·L⁻¹; NO₃⁻ concentrations (conc) in mgN·L⁻¹; H⁺ load in kg·ha⁻²·mo⁻¹; SO₄²⁻ loads in kgSO₄²⁻·ha⁻²·mo⁻¹; NO₃⁻ loads in kgN·ha⁻²·mo⁻¹.

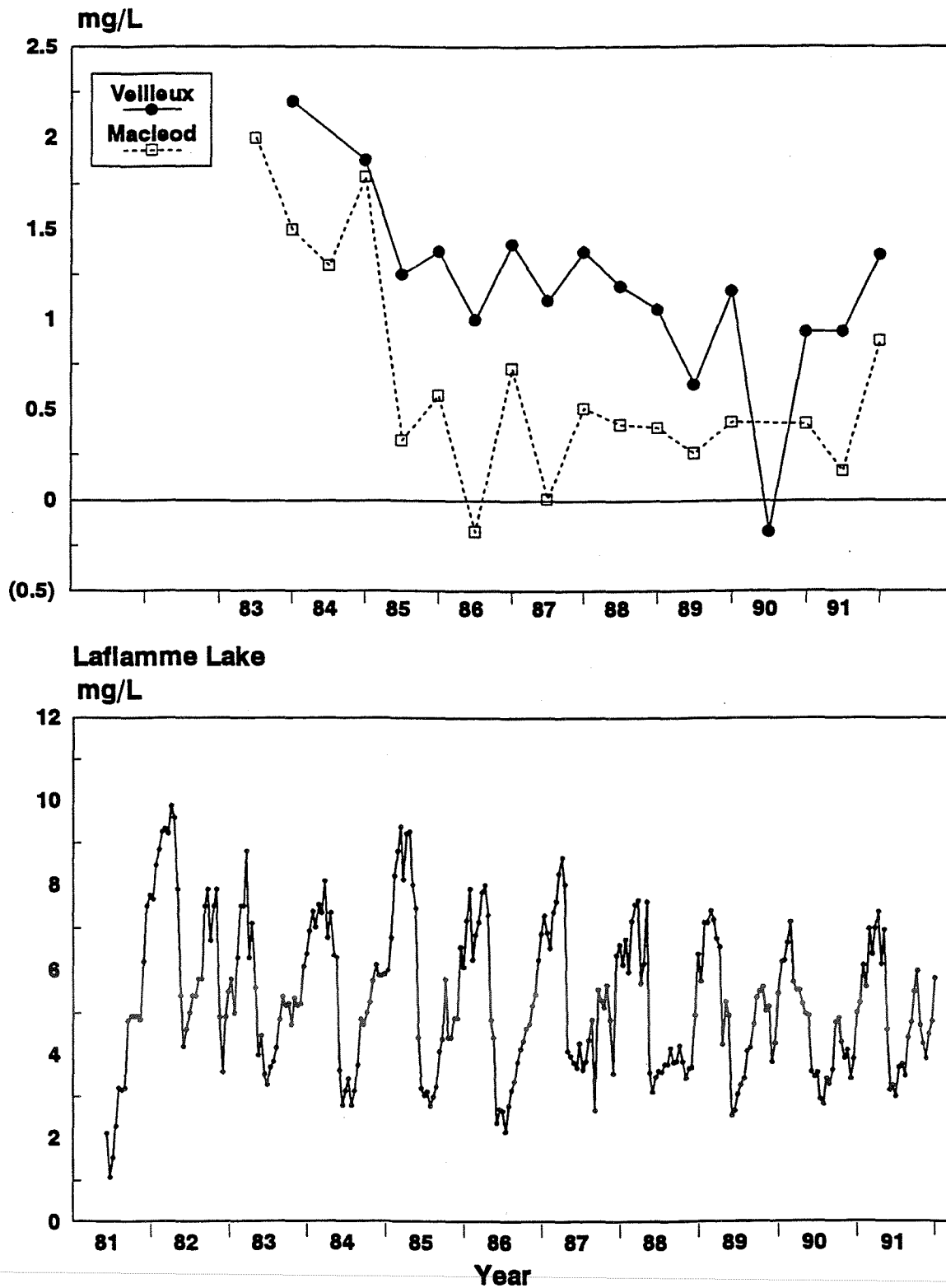


Fig. 2. Acid neutralising capacity decreases in the Laflamme Lake area.

concentrations have been decreasing since 1983 and do not show the initial increase that was reported in the LLW area.

By comparison to SO_4^{2-} fewer trends were observed for ANC (5 increases, 6 decreases), $\text{Ca}^{2+} + \text{Mg}^{2+}$ (13 decreases, 3 increases) and pH (1 increase and 4 decreases). The most common combinations of chemical trends were: a decrease in SO_4^{2-} with no change in pH, ANC or $\text{Ca}^{2+} + \text{Mg}^{2+}$ (15 lakes), a decrease in SO_4^{2-} accompanied by decreasing $\text{Ca}^{2+} + \text{Mg}^{2+}$ (10 lakes), decreasing SO_4^{2-} and increasing ANC (4 lakes: recovery) and decreasing SO_4^{2-} , $\text{Ca}^{2+} + \text{Mg}^{2+}$ and ANC (2 lakes: dilution). In the lakes where SO_4^{2-} concentrations increased a significant decrease in ANC occurred, indicating acidification. There does not seem to be a geographic gradient of recovery from the effects of acid deposition in terms of ANC trends. The few significant decreases in ANC or pH occur in lakes located near the northern limits of the area (Fig. 3) suggesting recent acidification.

Discussion

The lakes that are being monitored are headwater lakes usually very sensitive to acid deposition. This type of lake should be the first to respond to changes in the atmospheric deposition of acidifying pollutants. For lakes in southern Québec, Dupont (1992b) determined with the use of modelling (SIGMA/SLAM) that acid deposition is responsible for pH decreases of 0.14 to 0.75 units since the onset of industrial activity. Dupont and Grimard (1986) have also shown that there is a good spatial relationship between SO_4^{2-} content in lakes and precipitation in southern Québec. However, they were not able to find a similar relationship for NO_3^- .

There are very few sites in Québec where both precipitation chemistry and water chemistry are monitored simultaneously. This makes it difficult to study and establish, on a finer scale, the relationship between trends in precipitation variables and trends in water quality variables. At the LLW, trends for SO_4^{2-} and NO_3^- concentrations in precipitation and lake water over an 11 year period are not the same. While a 25% decrease in SO_4^{2-} concentrations in precipitation is reported, there is no overall trend for SO_4^{2-} in lake water. Even the correlation between annual means for SO_4^{2-} concentrations in precipitation and lake water is low ($r = 0.04$). While NO_3^- concentrations in precipitation have increased by 19%, the overall decrease in NO_3^- in lake water over the same period is 17%.

Over a shorter period, there are signs that lake SO_4^{2-} concentrations are responding to decreased SO_4^{2-} loading. From December 1984 to December 1991, several decreasing trends in lake SO_4^{2-} concentrations were found in the LLW area as well as over a broader area. There is a spatial pattern to this decrease. The largest decrease occurred in the south-west section of the area. This section receives the largest SO_4^{2-} deposition and is close to SO_2 sources in Ontario and the United States. There also seems to be a larger decrease, than the average, for the lakes in the north-east section of the network. This may be because this section is more affected by an important local source, Rouyn Noranda, than the section along the St. Lawrence. Important decreases in SO_2 emissions occurred at Rouyn Noranda in the late 1980's. Because of these decreases in SO_2 emissions, it is surprising that decreases in SO_4^{2-} concentrations in the section just south of Rouyn Noranda (north-west corner of LQN) are small or do not exist at all. This section seems to contain lakes that behave somewhat like lakes in the Sudbury area. Keller et al. (1992) have reported slight increases in concentration of SO_4^{2-} and decreases in pH and ANC in Sudbury area lakes in the late 1980's. This small reversal in recovery may be linked to climatic factors such as precipitation patterns. The abundant precipitation of 1988 may have led to a large export of acid and sulphate stored in the watershed during the preceding two dry years.

As for NO_3^- , although there does not seem to be any relationship between NO_3^- concentration trends in precipitation and lake water in the LLW area, there are indications that NO_3^- levels are increasing in other LQN lakes. N-induced surface water acidification has received very little attention so far in Québec but

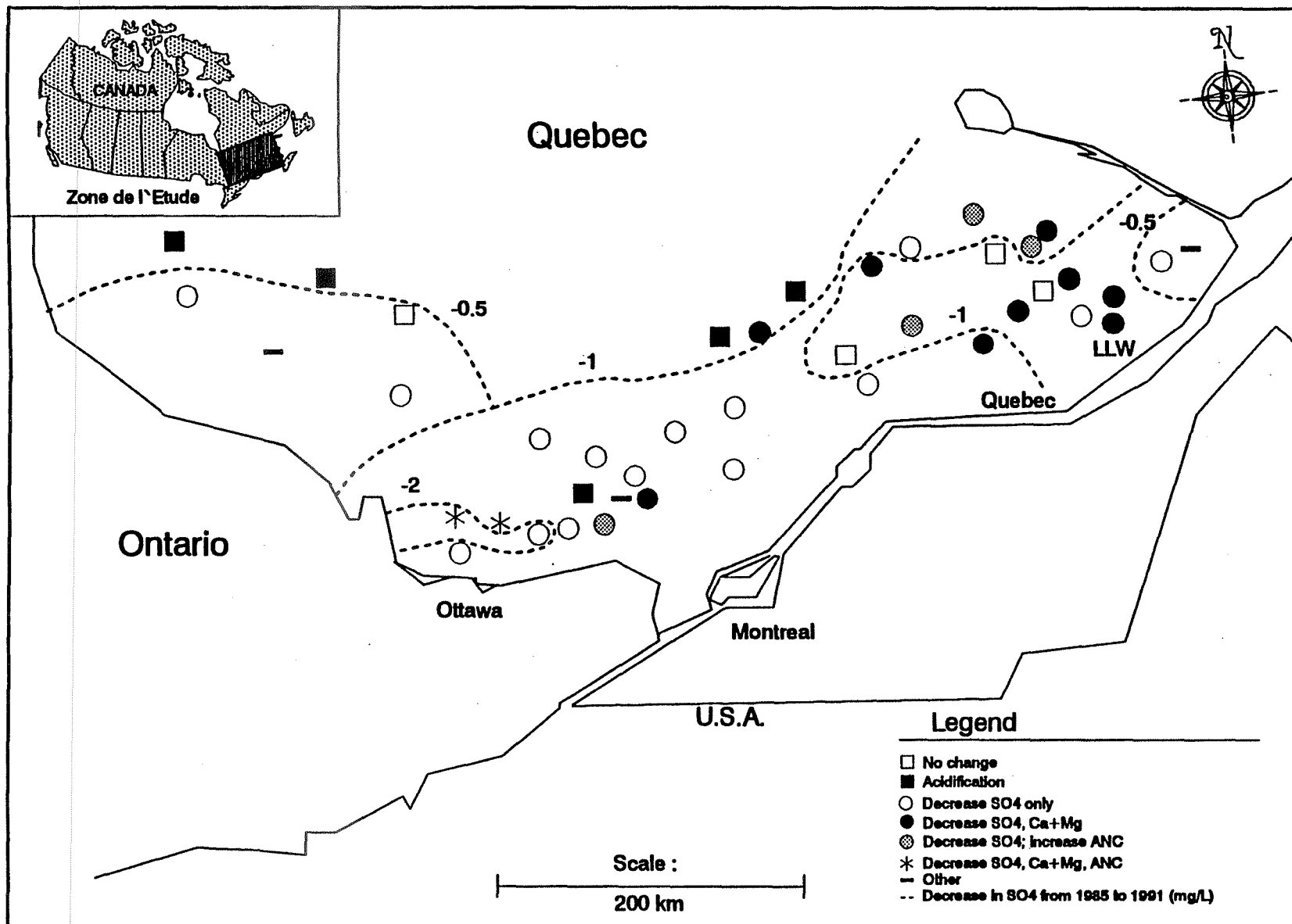


Fig. 3. Combination of chemical trends in water quality in southern Québec.

information from trend analysis is becoming available and may shed some light on the situation. The important seasonality of this variable may complicate the interpretation of trend analysis results.

For lakes on the Canadian Shield, both acidification (Dillon et al. 1987) and recovery (Dillon et al. 1986; Kelso and Jeffries 1988) have been documented in Ontario. In the LLW area, trends in decreasing ANC (3 out of 5 lakes) were reported. Four other very sensitive lakes (Fauvette, des Papillons, Belle-Truite, Chevreuil) also exhibit decreasing ANC trends. There are four additional lakes in the LQN network (Najoua, Adanys, Nolette, 75869) which showed both a decreasing pH and ANC. These trends can be considered to document acidification in Québec. Recovery of lakes, as measured by an increase in ANC, seems to lag the decrease in lake SO_4^{2-} concentrations. Few increasing trends in ANC have been identified and even those were confined to the December 1984 to December 1991 period. Several decreasing trends in $\text{Ca}^{2+} + \text{Mg}^{2+}$ were reported during this same period, indicating that the watersheds may respond to a decrease in SO_4^{2-} by decreasing cation export and increasing ANC outputs. Kelso and Jeffries (1988) reported that both mechanisms seem to have occurred in Ontario lakes, the response to decreases in SO_4^{2-} being in terms of increased ANC from 1979 to 1982 and in terms of decreases in $\text{Ca}^{2+} + \text{Mg}^{2+}$ from 1982 to 1985. Around Rouyn Noranda, Dupont (1992c) reports that decreases in lake SO_4^{2-} concentrations between 1982 and 1991 were matched by decreases in base cations with little change in ANC.

The rate of recovery of surface waters may increase further, because additional decreases in wet SO_4^{2-} deposition are expected in the upcoming years, as the Canadian acid rain reduction programme (1994) and the 1995 deadline for the first set of emission reductions in the United States (1995) comes into effect. Modelling predicts that the acidity of lakes will decrease in a large portion of southern Québec once the goals of the emission reduction programmes are met. However, even then, special attention will have to be given to the rate of recovery of very sensitive lakes since their critical loads ($10\text{-}15 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$) may still be exceeded by atmospheric deposition, a situation which may not allow the lakes to maintain ecosystem structure and function.

References

- ANONYMOUS. 1984. *NAQUADAT, dictionary of parameters codes*. Water Quality Branch, Environment Canada, Ottawa. 319 p.
- BARRIE, L.A., and A. SIROIS. 1986. Wet and dry deposition of sulphates and nitrates in eastern Canada: 1979-1982. *Water, Air and Soil Pollut.* 30: 303-310.
- BERRYMAN, D., B. BOBÉE, D. CLUIS, and J. HAEMMERLI. 1988. Nonparametric tests for trend detection on water quality time series. *Wat. Res. Bull.* 24 (3): 545-556.
- BOUCHARD, A. and J. HAEMMERLI. 1992. Trend detection in water quality time series of LRTAP-Québec network lakes. *Water, Air and Soil Pollut.* 62 (1-2): 89-110.
- CLUIS, D., C. LABERGE, and C. HOULE. 1988. *Détection des tendances et dépassement de normes en qualité de l'eau*. Institut National de la recherche Scientifique (INRS-Eau), St. Foy, Quebec. 127 p.
- DILLON, P.J., R.A. REID, and D. DEGROSOIS. 1987. The rate of acidification of aquatic ecosystems in Ontario, Canada. *Nature* 329 (6134): 45-48.
- DILLON, P.J., R.A. REID, and R. GIRARD. 1986. Changes in the chemistry of lakes near Sudbury, Ontario following reductions of SO_2 emissions. *Water, Air and Soil Pollut.* 31 (1-2): 59-65.
- DUBOIS, L., A. BOUCHARD, M. PAPINEAU, and D. LABONTÉ. 1992. *Temporal evolution of water quality in southern Quebec lakes affected by acid precipitation*. Environment Canada, St. Foy, Quebec. 170 p.
- DUPONT, J. 1992a. Quebec lake survey: Statistical assessment of surface water quality. *Water, Air and Soil Pollut.* 61 (1-2): 107-124.
- DUPONT, J. 1992b. Quebec lake survey: Origin and extent of acidification. *Water, Air and Soil Pollut.* 61 (1-2): 125-137.
- DUPONT, J. 1992c. *Effets des réductions des émissions de SO_2 sur la qualité de l'eau des lacs de la région de Rouyn-Noranda*. Ministère de l'Environnement du Québec, Québec. 74 p.

- DUPONT, J. and Y. GRIMARD. 1986. Systematic study of lake water acidity in Québec. *Water, Air and Soil Pollut.* 31 (1-2): 223-230.
- HIRSCH, R.M. and J.R. SLACK. 1984. A nonparametric trend test for seasonal data with serial dependence. *Wat. Resour. Res.* 20: 727-732.
- HIRSCH, R.M., J.R. SLACK and R.A. Smith. 1982. Techniques of trend analysis for monthly water quality data. *Wat. Resour. Res.* 18 (1): 107-121.
- KELLER, W., J.R. PITBLADO, and J. CARBONE. 1992. Chemical responses of acidic lakes in the Sudbury, Ontario, area to reduced smelter emissions, 1981-1989. *Can. J. Fish. Aquat. Sci.* 49 (Suppl. 1): 25-32.
- KELSO, J.R.M. and D.S. JEFFRIES. 1988. Response of headwater lakes to varying atmospheric deposition in north-central Ontario. *Can. J. Fish. Aquat. Sci.* 45 (11): 1905-1911.
- LETTENMAIER, D.P. 1976. Detection of trends in water quality data from records with dependent observations. *Wat. Resour. Res.* 12 (5): 1037-1046.
- PAPINEAU, M. and J. HAEMMERLI. 1992. Changes in water quality in the Laflamme lake watershed area, Canada. *Water, Air and Soil Pollut.* 61 (1-2): 95-105.

ACIDIFICATION STATUS OF DANISH LAKES AND STREAMS RELATED TO SOIL TYPE AND LAND USE

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Abstract

An outline of the acidification status of Danish lakes and streams is given with reference to soil type and land use. About 10% of Danish lakes and streams have pH values below 6.5. A compilation of chemical and biological data shows that possibly 50% of Danish low-alkalinity seepage lakes and pools have become more acidic, while few have increased in alkalinity during the latest 30-50 years. Four streams in a sandy region and two streams in a calcareous region have been monitored for a 12 year period. The four streams in sandy areas have shown a significant acidification trend, while the two other streams have shown no signs of acidification. The main cause for the acidification of the seepage lakes is probably atmospheric deposition. Stream acidification may be attributed to both land use practice and atmospheric deposition.

Key words: Acidification, status, Denmark, regional, lakes, streams, trends, land use, soil type

Introduction

The general water chemistry of Danish lakes and streams reflects the catchment soil type and land use patterns. There are two major soil types in Denmark, one is dominated by sandy soils poor in lime, located in the leached sandy Riss landscape, west of the Weichselian terminal moraine (Region 1, Fig. 1). The other is generally rich in lime, consisting of clayey, calcareous moraine of the Weichselian landscape, in the eastern parts of Denmark (Region 2, Fig. 1). Due to intensive agriculture in Denmark (65% of the total area), forestry, and a rather dense population, most Danish lakes and streams are affected by human activities. Effects such as: eutrophication, alkalisation through liming, acidification, and ochre formation, through pyrite oxidation and dissolution in catchments where land reclamation has been conducted in the past, confound the interpretation of acidification derived from the atmospheric deposition. Only in nature reserves and other uncultivated areas such as heathlands, dune landscapes and naturally forested areas is it possible to detect acidification caused solely from atmospheric deposition.

The present outline of the acidification status of Danish lakes and streams takes into account the above considerations. It is not simply confined to the impact of atmospherically derived acidification, but considers the sum of the acidifying and alkalising processes involved.

Lakes

Geographic Distribution

In Denmark about 7200 lakes have been registered and mapped with a total area of about 470 km² or 1.1% of the total area of Denmark (Miljøministeriet 1990). Most of them are small (less than 2 ha.). At the Danish National Environmental Research Institute (NERI) a lake database of chemical and biological data has been established which presently contains data from about 500 lakes, representing different geographic areas, soil types, and land uses. Only about 10% of these lakes have pH values below 6.5 (Table 1).

A compilation of available chemical and biological information on acidity or sensitivity to acidification was gathered from literature references and other reported or unpublished material, in order to characterise all known acidic and acid sensitive lakes and pools in Denmark. Information was found for approximately 110 lakes and pools (Rebsdorf and Nygaard 1991). Only lakes and pools with mean alkalinities less than 0.4 mmol·L⁻¹ or, if no alkalinity values were available, lakes with pH less than 7.0 were included. The

majority of the lakes are situated in sandy soils (Region 1, Fig. 1) and concentrated in three areas, one in the north-western part of Jutland near the coast (mostly dune lakes), another in the south-western part of Jutland (mostly heath pools), and a third in central Jutland, where some well studied clearwater *Lobelia* lakes are situated, such as the Grane Langsø and Kalgaard Sø. In the north-eastern part of the largest Danish island (Zealand), the soil is relatively sandy, and there are a few acidic and low-alkaline small lakes and pools, such as Lake Store Gribso. 75% of lakes are humic with water colour above 50 mg·L⁻¹ Pt (Fig. 2), and pH distribution is bimodal (Fig. 3). All registered localities are seepage lakes or pools with no permanent inflows or outlets, which implies that in-lake processes may have a considerable influence on the acid-base status.

Table 1. Distribution of mean pH in 477 Danish lakes (NERI, unpublished).

Percentage of lakes	pH interval	Mean pH
5.5	<5.3	4.7
2.5	5.3-6.0	5.6
2.5	6.0-6.5	6.3
84.3	6.5-9.0	8.0
5.2	>9.0	9.3

Trends

From two lakes, Store Gribso and Grane Langsø, there is substantial evidence that rapid acidification has occurred during the last 30-50 years (Wium-Andersen 1987; Rebsdorf 1983; Rebsdorf and Nygaard 1991). In addition, Rebsdorf and Nygaard (1991) showed that 18 out of 43 lakes and pools showed a tendency towards acidification, three became more alkaline, and 22 showed no tendency. Based upon diatom inferred pH change, studied by van Dam (in press) and partly published in Charles et al. (1990), 10 out of 17 localities had become more acidic, four remained constant, and three had become less acidic during the period between the 1940-50's and 1982.

Streams

Geographic Distribution

In contrast to the acidic or acid sensitive seepage lakes described above, almost all Danish streams are influenced both by point and non-point sources which affect their chemical status. Alkaline waters are primarily found in eastern Denmark (Region 2, Fig. 1), and the less alkaline and acidic waters belong primarily to Region 1. About 290 streams are monitored in a national monitoring programme and are included in a NERI database. The pH distribution (Fig. 4) of 33 streams in Region 1 and 259 streams in Region 2 shows that Region 1 streams have pH values around 7 ± 1 , while Region 2 streams have pH values nearly one unit higher. A few exceptions to these acidic pH values are streams, located in the sandy areas, near the border between Regions 1 and 2, and the Skærbæk. These streams have been incorporated into the International Co-operative Programme (ICP) on Assessment and Monitoring of Acidification of Rivers and Lakes. The regional variation of both pH and alkalinity, studied by Rebsdorf et al. (1991), showed a pH distribution similar to the national monitoring programme, however, values were slightly

lower (Fig. 5 and Table 2). The reason for this difference is probably Rebsdorf et al. (1991) contained a greater proportion of pH values from small headwater streams than did the NERI stream data base. The alkalinity values differ considerably between the two regions (Fig. 5 and Table 2).

Table 2. Regional mean pH and alkalinity of Danish streams.

Region	pH	Alkalinity $\text{mmol}\cdot\text{L}^{-1}$	References
1	7.1		NERI database
	6.9	0.59	Rebsdorf et al. (1991)
2	7.7		NERI database
	7.6	2.24	Rebsdorf et al. (1991)

Trends

In six Danish streams the pH and alkalinity has been measured for a 12 year period. Three of them belong to the sandy Region 1 and about half of their catchment areas is intensively cultivated farmland. One, stream situated just at the border between Regions 1 and 2 (Skærbæk), is the Danish site for the ICP on Assessment and Monitoring of Acidification of Rivers and Lakes. The soil is dominated by quartz sand, and the catchment area is almost completely covered by heath vegetation. The remaining two streams belong to the calcareous Region 2, and their catchments are intensively cultivated. A regional trend analysis showed that the three streams from the sandy areas in Region 1 and the Skærbæk showed a significant acidification trend for both pH ($-0.027 \text{ units}\cdot\text{yr}^{-1}$, $P=0.0001$) and alkalinity ($-0.0047 \text{ mmol}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$, $P=0.0215$). An example showing the changes in pH and alkalinity for one of the streams in Region 1 from 1977 to 1988 is illustrated in Fig. 6. The Skærbæk has low alkalinity compared to most other Danish streams (mean alkalinity $\approx 0.05 \text{ mmol}\cdot\text{L}^{-1}$), while the other three streams, in Region 1, have medium alkalinities (0.37 , 0.51 , and $0.79 \text{ mmol}\cdot\text{L}^{-1}$) (Rebsdorf et al. 1991). The two streams in the calcareous Region 2 (alkalinity 1.6 and $2.8 \text{ mmol}\cdot\text{L}^{-1}$, respectively) showed no trends.

Discussion and Conclusion

Because the Danish acidic and low-alkaline lakes and pools generally are seepage lakes and the lake water is more or less of ombrogenic nature, atmospheric deposition, is undoubtedly a major acidifying factor, as evidenced by the lake Store Gribso in north-east Zealand (Wium-Andersen 1987; Rebsdorf and Nygaard 1991), Grane Langsø (Rebsdorf 1983; van Dam, in press; Charles et al. 1990; Rebsdorf and

Nygaard 1991; Jacobsen 1992), and several other lakes and pools for which comparisons of new and old chemistry and diatom samples were made (Rebsdorf and Nygaard 1991). Many of the brown-water lakes and pools contain natural organic acids. The humic lake Store Gribso, has experienced a pH decrease from about 5.0 in 1949-1950 to about 4.2 in 1985 (Wium-Andersen 1987), suggesting that naturally acidic lakes and pools have been further acidified.

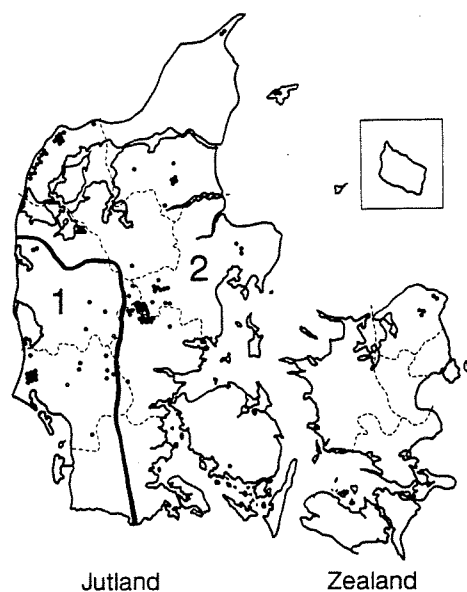


Fig.1. Map of Denmark, divided into two main regions: Region 1 is sandy Riss landscape and Region 2 is clayey, calcareous Weichselian landscape. The location of 109 known acid or low-alkaline ($\text{alk.} < 0.4 \text{ mmol l}^{-1}$) lakes and pools are indicated (dots).

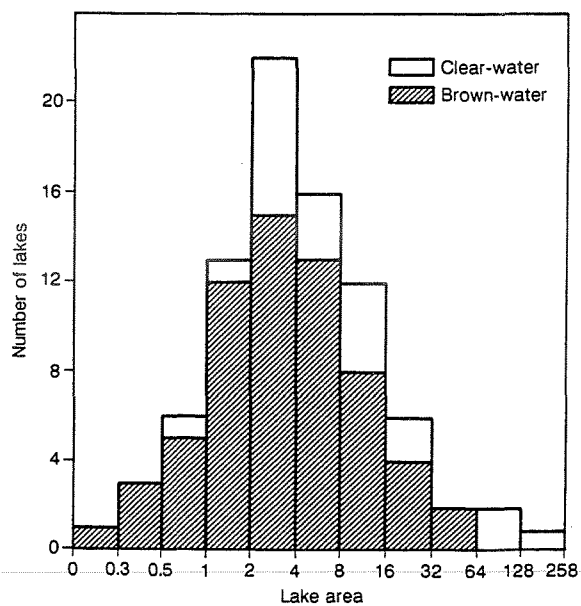


Fig. 2. Distribution of brown-water (colour $50 \text{ mg l}^{-1} \text{ Pt}$) and clear-water lakes with alkalinities, 0.4 mmol l^{-1} .

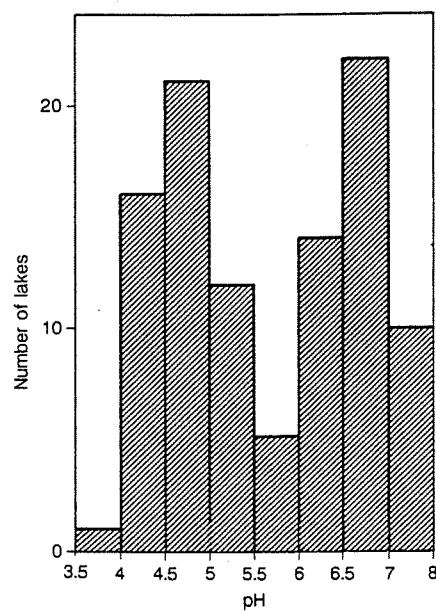


Fig.3 . Distribution of the mean pH values for 101 acid and low-alkaline (alkalinity $< 0.4 \text{ mmol l}^{-1}$) Danish lakes and pools.

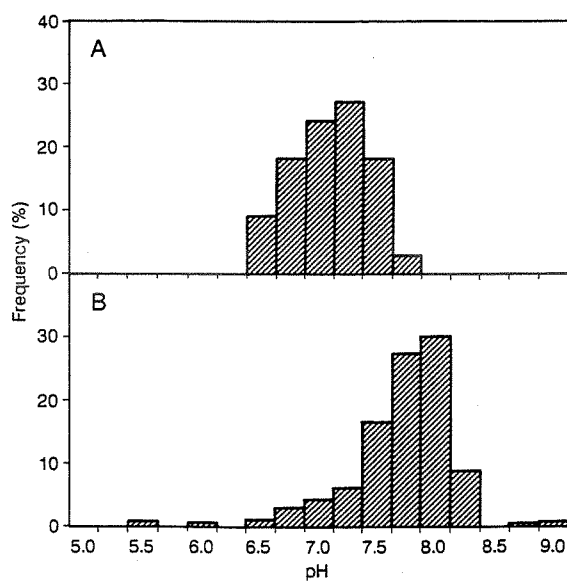


Fig. 4. Frequency distribution of the mean pH values of (A) 33 national monitoring streams in region 1; (B) 259 national monitoring streams in region 2 (Fig. 1)

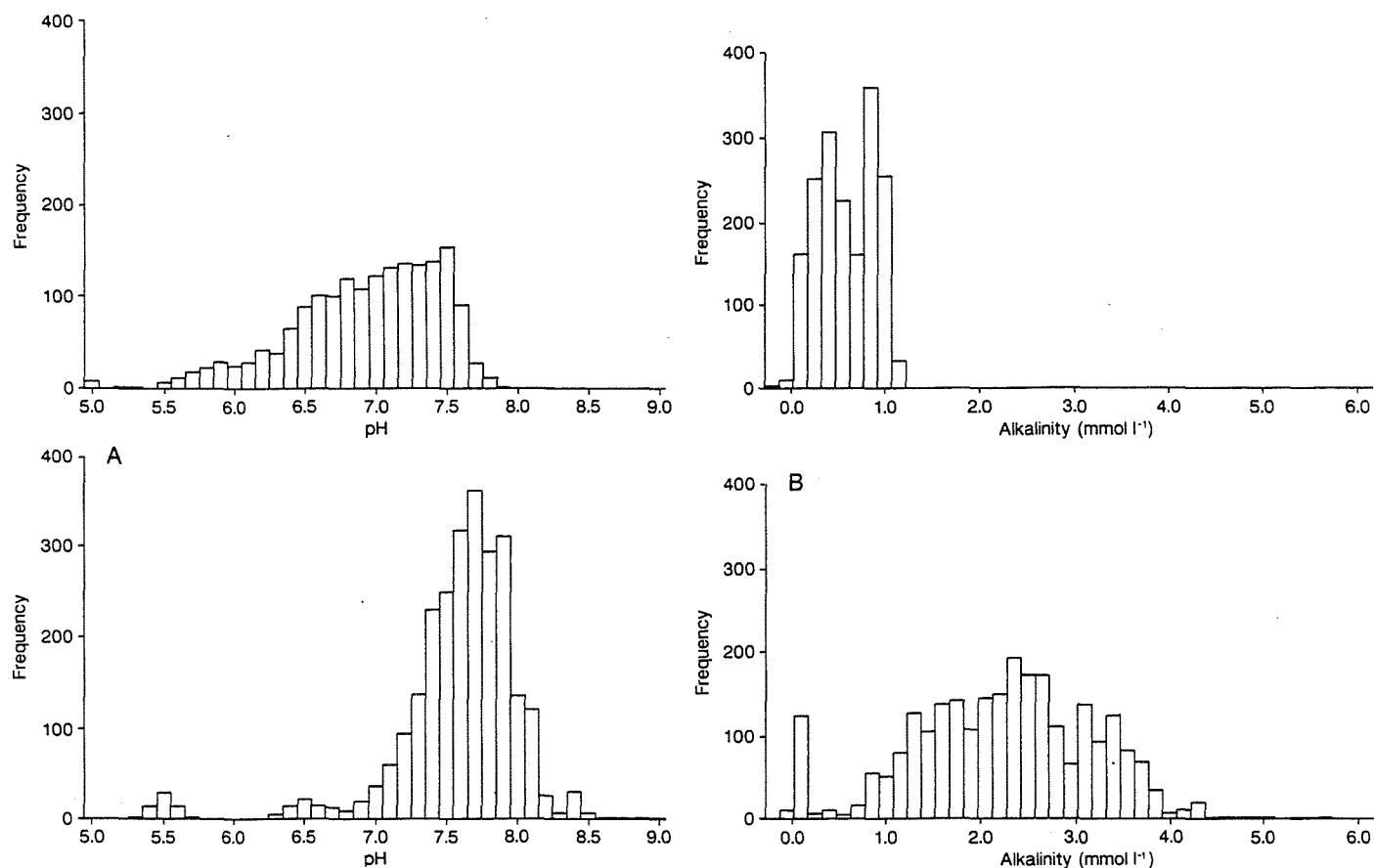


Fig. 5. Frequency distribution of mean pH values of pH (A) and alkalinity (B) in 49 streams in region 1 (top) and 110 streams in region 2 (bottom) (Rebsdorf et al. 1991).

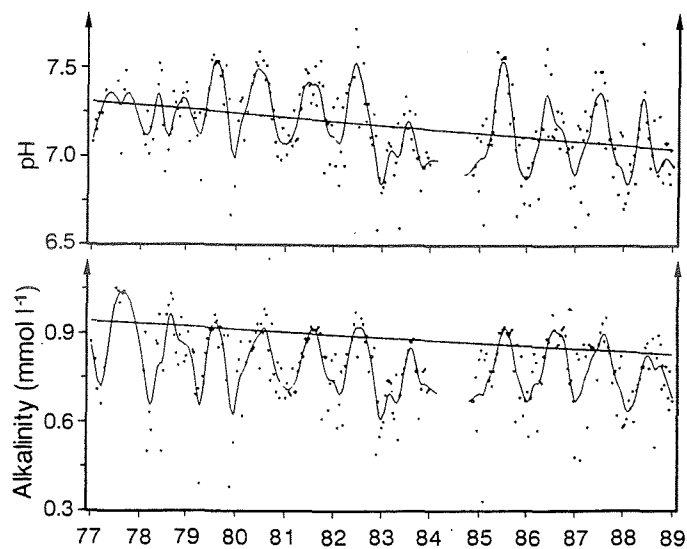


Fig. 6. Long-term trends of pH and alkalinity in Gryda Å at Sognstrup. Values between 15 May and 15 September are included in the analysis of trend for alkalinity whereas all values are included for pH. Further information in text. Rebsdorf et al. (1991).

Almost all Danish streams are running through cultivated catchment areas, therefore, they are influenced by anthropogenic activities. One small stream, the Skærbæk in central Jutland, drains an uncultivated heath area. The observed decrease in its pH (5.8 to 5.5) from 1977 to 1988 is undoubtedly due to atmospheric deposition. Observed decreases in pH and alkalinity for the three streams in the sandy areas of Region 1 was probably caused by agricultural activity in their catchments. The main acidifying factor is believed to be nitrification of ammonia-containing manures applied to the fields. This is in agreement with increases in the nitrate concentrations in the streamwater. Analysis of groundwater in parts of western Jutland, which have been influenced by manure show decreases in pH and alkalinity and simultaneous increases in nitrate concentration (Pedersen 1985; Rebsdorf et al. 1991). Two Danish streams in areas planted with coniferous trees in sandy soils have recently been shown to exhibit acidifying tendencies depending on the amount of precipitation. High amounts of precipitation leach humic material from the forest soil to the streamwater, which in turn becomes very brown ($395 \text{ mg} \cdot \text{L}^{-1} \text{ Pt}$) and acidic (pH 3.7) (Friberg and Rebsdorf 1997, see this volume).

References

- CHARLES, D.F., R.W. BATTARBEE, I. RENBERG, H. VAN DAM, and J.P. SMOL. 1990. Paleocological analysis of lake acidification trends in North America and Europe using diatoms and Chrysophytes. *In* Soils, aquatic processes, and lake acidification, p. 207-276. S.A. Norton, S.E. Lindberg, and A.L. Page (ed.) Acidic precipitation. Vol. 4. Advances in Environmental Science. Springer, New York, Berlin, Heidelberg.
- FRIBERG, N., and A. REBSDORF. 1997. The effect of forest type on stream acidity and consequently, on benthic fauna. Can. Tech. Rep. Fish. Aquat. Sci. Report no. # p.
- JACOBSEN, H. 1992. Bundplanter svinder i Grane Langsø (Bottom plants disappear in lake Grane Langsø). Vand og Miljø 9: 95-97. (in Danish).
- MIJØMINISTERIET. 1990. Tal om Natur og Miljø 1990 (Figures on nature and environment). Miljøministeriet (Ministry of the Environment), Copenhagen. 200 p. (in Danish).
- PEDERSEN, J.B. 1985. Forsuring af grundvandet i Vestjylland (Acidification of groundwater in western Jutland). Vandteknik, 53: 109-115. (in Danish).
- REBSDORF, A. 1983. Are Danish lakes threatened by acid rain?. *In* Ecological effects of Acid deposition. Nat. Swed. Environ. Project. Bd. Rep. PM 1636, p. 287-297.
- REBSDORF, A., and E. NYGAARD. 1991. Danske sure og forsuringstruede søer (Danish acid and acid vulnerable lakes). Miljøprojekt nr. 184. Miljøministeriet (Ministry of the Environment), Copenhagen. 108 p. (in Danish).
- REBSDORF, A., N. THYSEN, and M. ERLANDSEN. 1991. Regional and temporal variation in pH, alkalinity and carbon dioxide in Danish streams, related to soil type and land use. Freshw. Biol. 25: 419-435.
- VAN DAM, H. In press. Impact of acidification on diatoms and chemistry of softwater lakes and pools in Denmark, The Netherlands, Belgium and West Germany.
- WIUM-ANDERSEN, S. 1987. The development of the acid lake Grib sø in Denmark after 1950. *In* Reversibility of acidification, H. Barth (ed.). Elsevier, New York. p. 76-84.

GERMAN NETWORK OF MONITORING SITES IS COMPLETED BY SITES IN THE FORMER GDR

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So far several regions within the former Federal Republic of Germany have been included in the International Co-operative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes. At the beginning of 1992 the 55 sampling sites were reduced to 30 and the monitoring network was completed by the addition of nine sites in the former GDR, most of them running waters. As in the western part of Germany, wooded highlands and sandy areas are susceptible to acidification.

Regions affected by high deposition rates of acidifying air pollutants were chosen for the monitoring programme (Fig. 1): Erzgebirge, Elbsandsteingebirge, and the Sächsische Tieflandsbucht (Saxon lowlands). The latter region near the industrialised Leipzig/Halle area had the highest monitored sulphate load of the former GDR ($6 \text{ g S} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$, mean value 1985-1989). In the Erzgebirge and the Elbsandsteingebirge the values ranged from 3 to $4 \text{ g S} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$. In comparison, the sulphate load of the study areas in the former Federal Republic of Germany ranged from 1 to $2 \text{ g S} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$. The highest loading of nitrate, the other acidifying anion, occurred in the Erzgebirge ($0.6\text{-}0.8 \text{ g N} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$) due to relatively high precipitation in the highlands. In the Leipzig area values ranged from 0.4 to $0.6 \text{ g N} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$, as they did in most other regions of the former GDR. In general, nitrate deposition is lower than in the study areas in the former Federal Republic ($0.5\text{-}1.0 \text{ g N} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$). All values were reported as wet deposition data in the ambient air (Immissions-Jahresbericht 1989).

In the Erzgebirge, four streams and two reservoirs are monitored. The stream sampling sites are all situated on the mountain ridges adjacent to the border with CSFR. The stream Wolfsbach in Vogtland, the most western part of the Erzgebirge, is located in a primary rock zone, formed by phyllite, connecting the Erzgebirge with the Fichtelgebirge. Above the phyllite, brown earth-like soils have developed. A third of the catchment is wooded, predominantly with spruce, while the rest is used for agriculture. The low acidification of the stream allows the occurrence of *Margaritifera margaritifera*.

In the granite masses of the western part of the Erzgebirge, the stream Große Pyra and the reservoir Sosa are monitored. The soils, here, also consist of brown earth. At both sampling sites the catchment is entirely wooded, mainly with spruce. The Große Pyra and one of its tributaries originates in a moorland. During snow melt this stream is periodically acidic with pH values as low as 4 (biotic acidification score 0.25, according to the system of Raddum et al. [1988]).

In the centre of the Erzgebirge the stream Rote Pockau and the reservoir Neunzehnhain were selected for monitoring sites. In the catchment of the Rote Pockau, the geological subsoil consists of 90% orthogneiss and 10% granite with brown earth on top. The catchment, characterised by extensive peat areas, is dominated by spruce monocultures without settlements. In this area 51-70% of the trees are damaged by emissions. The periodic acidic nature of the stream is indicated by its biological score of 0.25 (Raddum et al. 1988).

The investigated stream in the eastern part of the Erzgebirge is the Wilde Weißeritz. The sampling site is situated near the station Zinnwald where the highest NO_3^- deposition rate of the former GDR was measured. The geological subsoil is composed of 80% phyllite and 20% granite and gneiss. Three quarters of the vegetation of the catchment consists of spruce forest, one quarter is grazing land. The trees

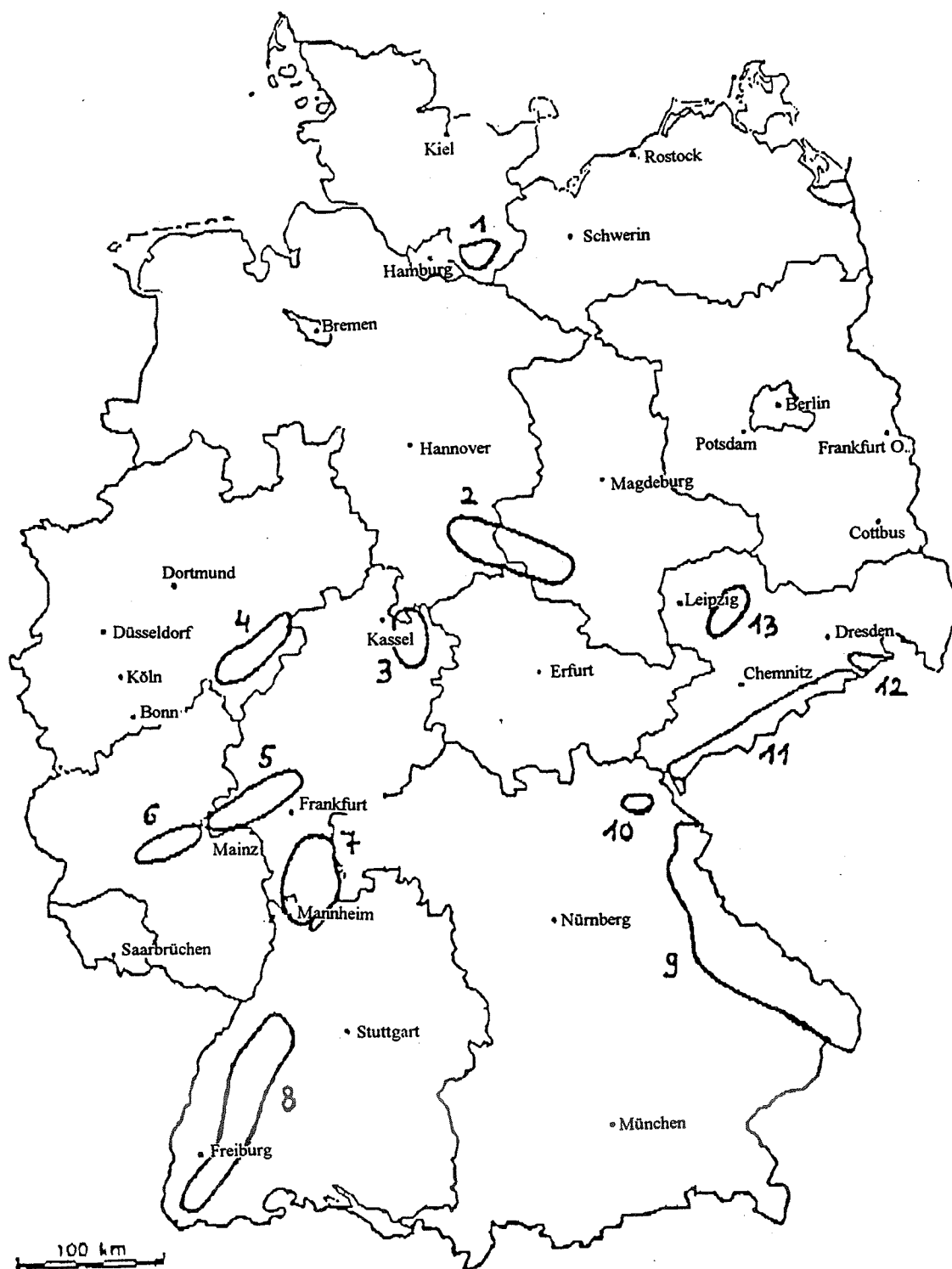


Fig. 1. Regions of Germany included in the ICP on Assessment and Monitoring of Acidification of Rivers and Lakes (1: Lauenburger Seenplatte, 2: Harz, 3: Kaufunger Wald, 4: Rothaargebirge, 5: Taunus, 6: Hunsrück, 7: Odenwald, 8: Schwarzwald, 9: Ostbayerisches Grundgebirge, 10: Fichtelgebirge, 11: Erzgebirge, 12: Elbsandsteingebirge, 13: Sächsische Tieflandsbucht).

are highly damaged by emissions. Biological investigations classify the Wilde Weißeritz as periodically acidic (score 0.25).

The Elbsandsteingebirge adjoins the Erzgebirge to the east. Its geological sub-soil consists of sandstone. The substrate at the Taubenbach stream site is 80% sandstone and 20% quartzite. The catchment and its coniferous forest have been partially damaged by emissions and logging. So far, this site is not as acidified as the Erzgebirge. Biological results indicate moderately acid water (score 0.5).

The Sächsische Tieflandsbucht (Saxon Lowlands) is a glacially formed plain covered by clay soils, with only a few wooded hilltops, formed by porphyry. Two forested areas were selected for monitoring: 1) the Colditzer Forest (sampling site at the stream Ettelsbach) with sub-soil's consisting of quartzporphyr, gravel and sand. Here the mixed forest is composed of 55% deciduous trees (mainly oak, but also beech and birch) and 45% coniferous forest (pine, spruce and some larch); 2) the Dahleener Heide (sampling site at the stream Heidelberg) is mostly forested by pine. The catchment of the Heidelberg is covered with 85% pine and 15% deciduous woods (oak, alder and copper beech).

Both Ettelsbach and Heidelberg can be characterised as highly acidic waters (score 0.0). The pH-values stay below 5, year round, often reaching values below pH 4. In both streams very high sulphate concentrations are measured, corresponding to the high rate of sulphur deposition in this area.

References

- IMMISSIONS-JAHRESBERICHT. 1989. Meteorologischer Dienst der DDR. Meteorologisches Observatorium Wahnsdorf, Juli 1990.
- RADDUM G.G., A. FJELLHEIM, and T. HESTHAGEN. 1988. Monitoring of acidification through the use of aquatic organisms. Verh. Internat. Verein. Limnol. 23: 2291-2297.

Ciliata and Testacea as Indicators

Dr. G. Burkl

Munchen, Sept. 10, 1992

In the framework of our acidification monitoring programme in Germany we have included two small extra projects that may be of general interest. To establish the degree and extent of acidification of surface waters, chemical and biological methods are used. As biological indicators, benthic invertebrates and diatoms are normally used. We investigated the possibility of using other indicator-species in our river, and began two small pilot projects with Ciliata and Testacea. This work was carried out by Prof. Dr. Foissner, University of Salzburg, Austria.

Ciliata are highly specialised unicellular organisms, which serve well as bioindicators. However, a great deal of experience and work is required to identify them. Testaceae are a form of unicellular amoebae, living in a small shell. They are also unicellular.

Samples were taken in the upper course (near the source) of the following small streams, in the Fichtelgebirge region, one of the most acidified areas of Bavaria:

Stream	pH	Classification
Eger	6.0	periodically acidified
Röslau	5.2	periodically acidified
Zinnbach	4.1	permanently highly acidified

First results show:

- 1). Ciliatae are good indicators, not only of oligotrophy, but also of acidification. Peats contain ciliates of an entirely different species composition, so it seems possible to distinguish between the influence of humic acids and the minerals acid in rain. In the three sampling sites 20 - 23 different species were found.
- 2). Testacea found at the three sampling sites, resemble those of forest soils. They also seem to be good indicators, as the species composition differs among the three samples. Abundances increase with decreasing pH 6,800,000,000 up to 16,800,000,000 ind. \cdot m⁻². They seem to play an important role in the energy budget of acidified streams by replacing the missing invertebrate biomass.

HYDROBIOLOGICAL INVESTIGATION OF THREE ACIDIFIED RESERVOIRS IN THE JIZERA MOUNTAINS, THE CZECH REPUBLIC, DURING THE SUMMER STRATIFICATION

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Abstract

Three drinking water reservoirs in the Jizera Mountains (northern Bohemia, Czech Republic) were studied in the summer of 1992. These reservoirs are situated on a geologically sensitive area strongly affected by acid deposition. Stratification of temperature, conductivity, oxygen, pH, alkalinity, concentration of chlorophyll-a, and biovolume of the main species of phytoplankton were studied in vertical profiles. Species composition of phytoplankton and zooplankton were also determined. Acidification was recognized as a factor which significantly influenced both chemistry and plankton communities of all three reservoirs. Analysis of historical data showed that acidification probably began in the 1950's, and was first mistaken for dystrophy.

Key words: Mountain reservoirs, acidification, phytoplankton, zooplankton

Introduction

The acidity of water in the Jizera Mountain reservoirs was recognized in the 1920's, shortly after their construction, and was attributed to the dystrophic character of the basins (Gessner 1925, 1929). During investigations conducted in the 1950's and 1960's, the presence of extensive moors in the watersheds and the occurrence of *Sphagnum* in the littoral zone of the reservoirs were considered to be the reason for a continuing decrease in the pH of water (Sládeček 1955; Zubčenko 1955; Jirásek et al. 1959; Lhotský 1963a,b; Perman and Lhotský 1963; Chalupa 1965). In comparison with the data of Gessner (1925, 1929), more recent studies have shown reduced numbers of zooplankton species (Sládeček 1955) and the disappearance of fish from the reservoirs (Jirásek et al. 1959; Lhotský 1963a,b).

Investigations conducted in the 1980's were aimed at studying the effect of deforestation on the hydrology and chemistry of surface waters of the area. The results of these studies are summarized in reports by Bednářová et al. (1988), Bednářová (1990). Extremely low values of surface water pH (3.1-4.5) during the spring melting period and a mass die-back of coniferous forests in the whole region of the Jizera Mountains were characteristic of the 1800's (Bednářová 1988). Acid deposition was implicated as the cause of these changes.

Liming of waters in the Jizera Mountains has probably been conducted since the beginning of the century in order to maintain fish populations (Jirásek et al. 1959; Lhotský 1963b), and reached its highest intensity in the 1980's when, not only reservoirs, but also their watersheds and the entire mountain region were limed.

The aim of our study was to investigate the reservoirs Souš, Bedřichov, and Josefův Důl during the summer stratification, to assess the present state of their acidification.

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Study Sites and Methods

The Jizera Mountains are situated in northern Bohemia, Czech Republic (temperate zone, 50°45'-50°48'N, 15°8'-15°17'E). Their highest peak reaches 1124 m a.s.l. The geological substrate is granite. Mean annual precipitation is about 1400 mm, with a regular maximum in July. Mean annual temperature is 5°C. Wet deposition of sulphate (15 t·km⁻²·yr⁻¹), and nitrate (7 t·km⁻²·yr⁻¹) is about twice that of other mountain regions of Bohemia (Křeček 1991; Moldan 1991). Originally, covered by coniferous forests (mostly Norway spruce, *Picea abies*) and numerous peat bogs, this region has subsequently been deforested and overgrown by extensive grass stands (*Calamagrostis villosa* dominating).

Flooding at the end of the 1800's provided impetus for building reservoirs for protection. Six reservoirs were built between 1902 and 1915. The Souš and Bedřichov reservoirs are in this group. The Josefův Důl reservoir dates from the early 1980's. At present, all three reservoirs serve as sources of drinking water. Their basic morphometric and hydrologic characteristics are listed in Table 1.

Table 1. Characteristics of the reservoirs.

Parameter	Reservoir		
	Souš	Bedřichov	Josefův Důl
River	Černá Desná	Černá Nisa	Kamenice
Year impounded	1915	1905	1982
Altitude (m a.s.l.)	769	775	733
Surface area (km ²)	0.86	0.37	1.45
Maximum depth (m)	19.3	13.5	38.2
Volume (10 ⁶ m ³)	7.6	2.1	23.3
Watershed area (km ²)	13.2	4.1	19.8
Water residence time (days)	179	41	464

At the beginning of the century, streams in the watersheds of Bedřichov and Souš contained brown trout (*Salmo trutta*). Later (in 1910 and 1916, respectively), Arctic char (*Salvelinus alpinus*) was introduced to these reservoirs (Gessner 1929). In 1959, however, only Arctic char survived in streams located in the watershed of Souš (Jirásek et al. 1959; Lhotský 1963b). Presently all reservoirs are fishless, despite attempts to reintroduce Arctic char.

The reservoirs were sampled on June 17-18 and on July 22-23, 1992 from a boat anchored close to the deepest point (in Souš and Bedřichov), or from the intake tower on the Josefův Důl reservoir.

Temperature, pH, and the concentration of dissolved oxygen were measured by using a WTW pH-meter and a Y.S.I. oxygen meter *in situ*, for profiling the reservoirs. Alkalinity was determined by Gran titration of samples taken from different depths by a Van Dorn sampler; conductivity (corrected to 20°C) was measured on these samples with a Radelkis conductometer. A Secchi disk was used to measure transparency.

Phytoplankton samples were taken from different strata within the water column with a Van Dorn sampler. Subsamples for identification, counting, and measurement were preserved in Lugol's solution. These

samples were examined using a microscope with an immersion objective. Biomass of major groups of algae was expressed as biovolume. Concentration of chlorophyll-a was determined on unpreserved samples with a SLM fluorometer after hot extraction in an acetone:methanol mixture (Pechar 1987). Zooplankton samples were taken by vertical hauls from the bottom to the lake surface using plankton nets of 40 and 200 μm mesh size. Samples were preserved in formalin and used to determine zooplankton species composition.

Results and Discussion

Physical Parameters

The reservoirs under study are dimictic lakes with typical summer temperature stratification (Fig. 1). In both the Souš and Josefův Důl reservoirs a stable, homogeneous epilimnion is sharply separated from the hypolimnion during the summer period, due to their depth and the high winds in the deforested territory of the Jizera Mountains. The thermocline is normally situated at a depth of 6-8 m. In comparison the Bedřichov reservoir which is also exposed to strong winds showed only weak stratification during the summer season due to its shallower depth. In Souš and Bedřichov, a properly developed thermal stratification was not found before deforestation (Zubčenko 1955; Jirásek et al. 1959).

The vertical distribution of oxygen was of a clinograde type, showing a hypolimnetic deficit (Fig. 1). Conductivity ranged between 40-70 $\mu\text{S}\cdot\text{cm}^{-1}$ (Table 2). The transparencies of Souš and Bedřichov were 2-3 m. The Secchi disk values reached 4-5 m in Josefův Důl. Low values of transparency as well as the decrease in concentration of dissolved oxygen in the hypolimnion, are indicative of the relatively high primary productivity of these reservoirs.

Chemistry

The range of pH in the surface layer of Josefův Důl and Bedřichov was 5.0-5.5. The Souš reservoir was at pH 5.5 in June but rose to pH 6.2 in July due to liming of the reservoir (Fig. 1). Alkalinity in the surface samples from all three reservoirs ranged between -7.5 and 7.3 $\text{meq}\cdot\text{L}^{-1}$ in June, but in July the values reached 20 $\text{meq}\cdot\text{L}^{-1}$ in the Souš reservoir, and 34 $\text{meq}\cdot\text{L}^{-1}$ in Bedřichov (Table 2). The summer stratification of pH in Souš and Josefův Důl should be noted. Higher values of pH in the epilimnion are evidence of alkalization of the reservoirs by tributaries from the watersheds, and/or as a direct effect of liming (Fig. 2). The relatively "alkaline" tributaries, after liming, only influence the sharply separated epilimnion. We expect pH values to return close to the values of the hypolimnion during the autumnal circulation.

Values of pH and alkalinity from the 1920's (Gessner 1929), from the years 1947-1950 (Zubčenko 1955), 1959 (Jirásek et al. 1959), 1960-1962 (Chalupa 1965), and from the 1980's (Bednářová 1988), suggest that pH dropped below 4.5 and alkalinity close to zero in Souš and Bedřichov as early as the beginning of the 1950's. Seven years of chemical investigation of Souš conducted in the 1980's did not show any trend in pH (Bednářová et al. 1988). Our recent values of pH, however, are significantly higher than those reported by Bednářová. The interpretation of older data is complicated by the fact that different methods and sampling sites were used, and in particular, because the reservoirs and their watersheds have been limed. The first documented liming was carried out on the inflows of Bedřichov in 1961 (Lhotský 1963b). Since the early 1980's, the whole territory has been limed aerially (Bednářová et al. 1988). In addition, acid deposition has probably decreased as a result of deforestation, because the absence of forests in a mountain area reduces horizontal (dry, ed.) deposition, a major component of the total acid load to the watershed.

The Jizera Mountains are situated at the epicentrum of acidification in Europe as can be seen from the distribution of isolines of annual weighted mean pH of precipitation in Europe (Wright 1983), and from patterns of sulphate and nitrate deposition (Eliassen et al. 1988). Acidification of the Šumava Mountains and the High Tatra Mountains, which are more remote from the centres of emissions, began to take place

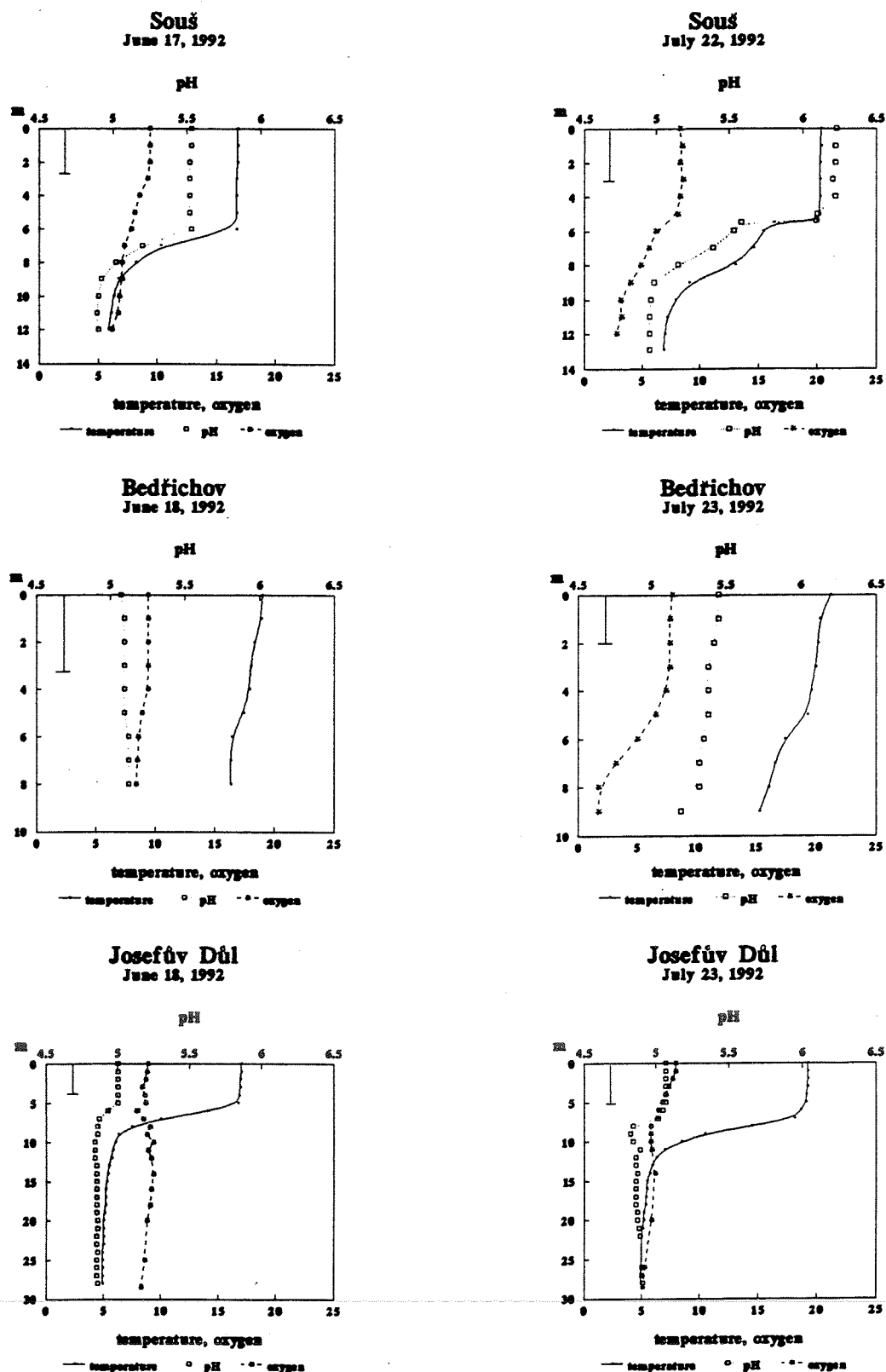


Fig. 1 Vertical temperature, oxygen and pH profiles of the Souš, Bedřichov and Josefův Důl reservoirs (1992).

Table 2. Vertical distribution of conductivity (K_{20} , $\mu\text{S}\cdot\text{cm}^{-1}$), alkalinity (Alk, $\mu\text{eq}\cdot\text{L}^{-1}$), and concentration of chlorophyll-a (Chl-a, $\mu\text{g}\cdot\text{L}^{-1}$) in the reservoirs.

	June 1992				July 1992			
	Depth	K_{20}	Alk	Chl-a	Depth	K_{20}	Alk	Chl-a
Souš	0	45	7.3	3.6	0	43	19.6	12.0
	1	45	5.2	3.5	1	43	17.8	11.9
	5	45	4.6	6.6	5	43	16.6	11.4
	12	45	-5.6	1.4	12	43	2.4	4.4
Bedřichov	0	59	0.8	5.4	0	53	7.9	16.4
	1	58	3.6	5.6	1	52	9.7	16.9
	3	58	2.2	6.8	5	51	10.3	8.4
	7	58	4.9	4.4	10	51	33.8	4.1
Josefův Důl	0	69	-7.1	2.1	0	65	-4.0	2.5
	1	70	-2.0	2.1	1	66	-5.0	2.4
	5	68	-2.8	1.2	5	66	-4.4	2.6
	10	71	-6.5	2.0	10	67	-5.5	4.0
	20	70	-6.0	0.5	20	64	-5.4	2.3
	27	66	-7.5	0.4	28	68	-5.6	2.6

in the 1960's and in the 1970's, respectively (Stuchlík et al. 1985; Fott et al. 1987; Fott et al. 1992), consequently, we can assume that the exposed area of the Jizera Mountains was affected by acidification 20 or more years earlier.

Phytoplankton

Algal species composition was similar in all three reservoirs. Dinophyceae dominated, forming 63-99% of the total biomass of algae, (most common genera: *Peridinium* and *Gymnodinium*), while Chrysophyceae (*Chrysococcus*, *Ochromonas*) contributed 2-16%, and Cryptophyceae (*Cryptomonas*) 2-15% (Fig. 3). Chlorophyceae (*Chlamydomonas*) and Bacillariophyceae were observed in some strata, but were an insignificant part of total algal abundance. Blue-green algae (*Cyanophyceae*) also composed a negligible part of the phytoplankton. Overall, the number of taxa was low, a feature consistent with the acidified state of the reservoirs.

Comparing our data to those of Perman and Lhotský (1963) shows that phytoplankton species composition has not changed significantly during the last 30 years. A bloom of *Peridinium umbonatum* (= *P. inconspicuum*) was first observed in 1959 (Lhotský 1963b); in the 1980's, dense populations of this alga caused a problem with drinking water treatment (Ladislav Havel, pers. comm.). The dominance of

inophyceae (namely *Peridinium*) is typical for the phytoplankton of acidified lakes and may be considered indicative of an advanced stage of acidification (Yan 1979; Fott et al., in press).

Generally the biomass of the phytoplankton was rather low (chlorophyll-a values of $0.4\text{--}6.8\text{ mg}\cdot\text{L}^{-1}$; biomass expressed as a biovolume $0.04\text{--}1.2\text{ mm}^3\cdot\text{L}^{-1}$). However, July values found in depths of 0–5 m in the Souš and Bedřichov reservoirs showed chlorophyll-a concentrations of $8\text{--}17\text{ mg}\cdot\text{L}^{-1}$ and biomasses of $1.0\text{--}5.3\text{ mm}^3\cdot\text{L}^{-1}$. Most of the biomass of phytoplankton was concentrated in the upper 5 m layer in each reservoir (Fig. 3 and Table 2). Relatively high concentrations of chlorophyll-a in the hypolimnion of Josefův Důl suggest the presents of algae adaptated to lower light conditions. July values of both chlorophyll-a and phytoplankton biovolume were significantly higher in all reservoirs.

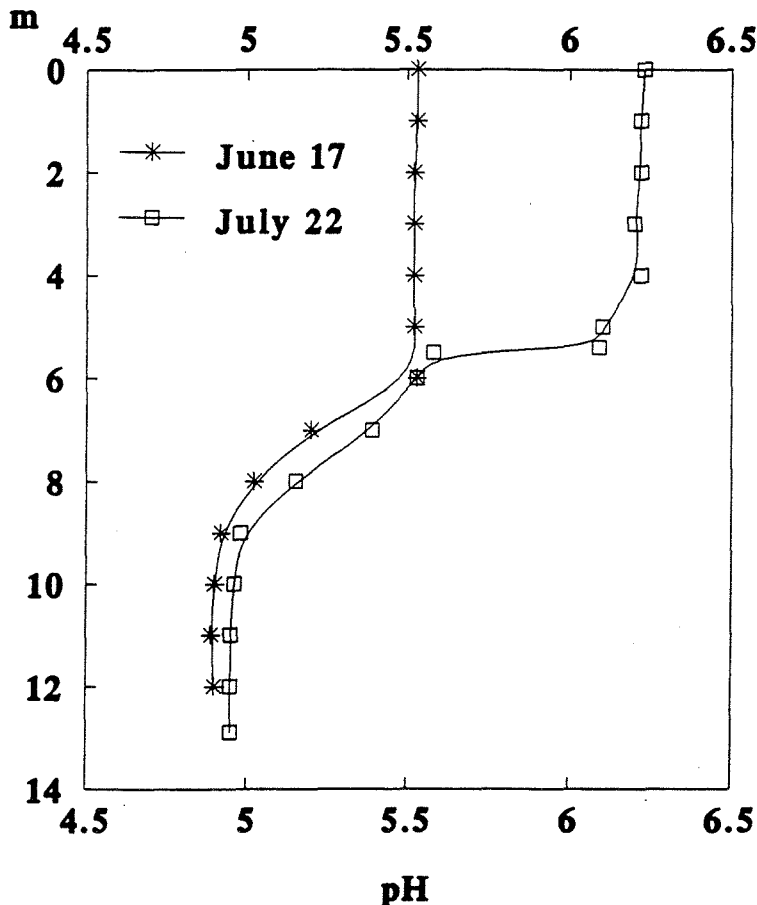


Fig. 2. Vertical temperature, oxygen and pH of the Souš reservoir.

Zooplankton

The major species of zooplankton in the reservoirs were *Brachionus sericus*, *Keratella valga*, *Microcodon clavus* (Rotatoria), *Ceriodaphnia quadrangula* (Crustacea: Cladocera), and *Acanthocyclops vernalis* (Crustacea: Copepoda). A complete species list is given in Table 3.

Microcodon clavus and *Keratella serrulata* dominant the zooplankton of strongly acidified lakes in the Šumava Mountains, south-western Bohemia (Fott et al., in press). Previous records of zooplankton in Souš (Gessner 1925, 1929; Sládeček 1955; Jirásek et al. 1959) show that *Keratella cochlearis* completely disappeared, while the acidophilous species *Keratella serrulata* first appeared between 1924 and the 1950's. Further changes in species composition of the zooplankton such as the extinction of *Bosmina longirostris* and *Eudiaptomus gracilis* (presents last reported by Jirásek et al. [1959]), were caused by increasing acidity rather than dystrophy in the reservoirs.

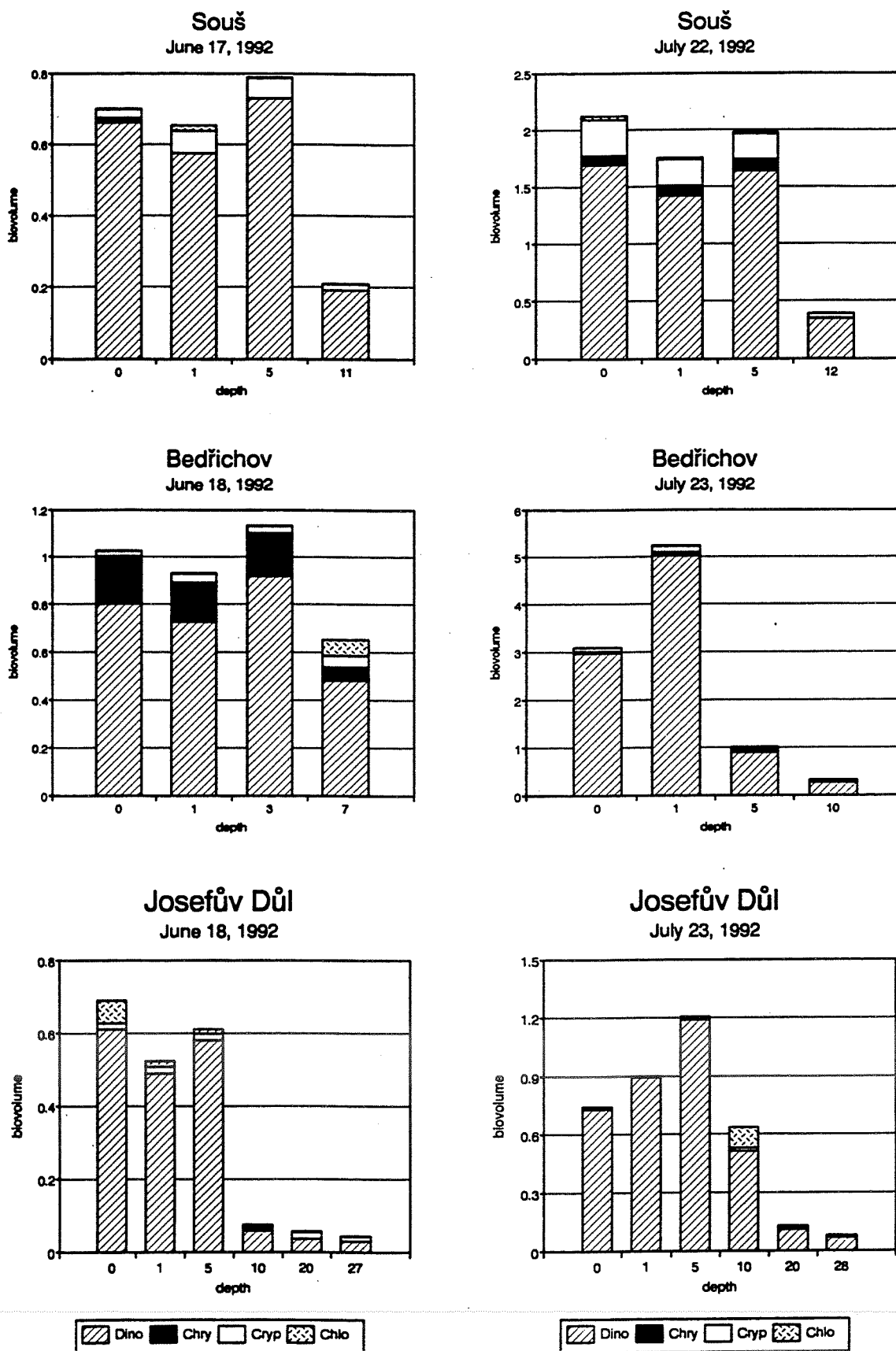


Fig. 3. Phytoplankton biomass in vertical profiles ($\text{mm}^3 \cdot \text{L}^{-1}$) from the Souš, Bedřichov and Josefův Důl reservoirs (1992).

The species *Brachionus sericus*, *Keratella valga*, *Ceriodaphnia quadrangula*, *Chydorus sphaericus*, and *Acanthocyclops vernalis* seem to exhibit a wide range of pH tolerance, as they have been present in the Souš reservoir since the first investigation in the 1920's (Gessner 1925). It seems likely that *Keratella valga* and *Ceriodaphnia quadrangula* were previously misidentified as *Keratella quadrata* and *Ceriodaphnia pulchella* and that no species replacements of these species occurred.

Conclusions

1. The reservoirs investigated are situated in a geologically sensitive area, heavily impacted by acid deposition. The natural acidity of water in the Souš and Bedřichov reservoirs, caused by humic acids, was altered by acid deposition, in the early 1950's. Anthropogenic acidification has caused the deforestation of the surrounding territory, decreased the pH of surface waters, and degraded life in streams and reservoirs.
2. Intensive liming of both watersheds and reservoirs has been conducted for many years, but has not eliminated the acidification problem. However, values of pH higher than 5 are occasionally found in reservoir epilimnion, perhaps due to liming or reduced deposition following deforestation of watersheds. A temporary alkalization of the epilimnion of Souš during the summer stratification, accompanied by a conspicuous increase in biomass of the phytoplankton, is, however, the only significant effect of the trends mentioned.
3. The seasonal instability of water chemistry together with the biological changes which result (such as short term peaks of phytoplankton), make drinking water treatment of these waters difficult.

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References

- BEDNÁŘOVÁ, M. 1988. Sledování kvalitativních ukazatelů povrchových vod v pramenných oblastech Jizerských hor. Sborník konference Vodohospodářské důsledky imisní kalamity v Jizerských horách, Liberec, Czechoslovakia, November 8-10, 1988: 188-194. Czechoslovak Society for Science and Technology (ČSVTS), Ústí n. Labem. [Investigation of qualitative parameters of surface waters in headwater areas of the Jizera Mountains. In Czech.]
- BEDNÁŘOVÁ, M. 1990. Jakost povrchových vod ve vybraných pramenných oblastech, In J. Urban, et al. Změny hydrologického režimu v pramenných oblastech. Závěrečná zpráva [Changes of the hydrological regime in headwater areas. Final report.]: 90-121. Water Research Institute of T.G. Masaryk, Praha. [Quality of surface waters in selected headwater areas. In Czech.]
- BEDNÁŘOVÁ, M., L. KAŠPÁREK, and L. VOTRUBA. 1988. Změny odtokového režimu v extrémně postižených oblastech. Závěrečná zpráva. Water Research Institute, Praha. 81 p. [Changes of the run-off regime in extremely affected areas. Final report. In Czech.]
- CHALUPA, J. 1965. Chemické složení vod československých údolních nádrží a rybníků. Acta hygienica, Praha. 33 p. [Chemistry of reservoirs and fish ponds in Czechoslovakia. In Czech.]
- ELIASSEN, A., O. HOV, T. IVERSEN, J. SALT BONES, and D. SIMPSON. 1988. Estimates of airborne transboundary transport of sulphur and nitrogen over Europe. EMEP/MSC W Rep. 1/88, Oslo.

- FOTT, J., E. STUHLÍK, and Z. STUHLÍKOVÁ. 1987. Acidification of lakes in Czechoslovakia, p. 77-79. In B. Moldan and T. Paces (ed.). Extended abstracts of International Workshop on Geochemistry and Monitoring in Representative Basins (GEOMON), Prague, Czechoslovakia, April 27-May 1, 1987. Geological Survey, Prague.
- FOTT, J., E. STUHLÍK, Z. STUHLÍKOVÁ, V. STRÁŠKRABOVÁ, J. KOPÁČEK, and K. ŠIMEK. 1992. Acidification of lakes in Tatra Mountains (Czechoslovakia) and its ecological consequences, p. 69-81. In R. Mosello, B.M. Wathne, and G. Giussani (ed.). Limnology on groups of remote lakes: Ongoing and planned activities. Documenta Ist. ital. Idrobiol. 32.
- FOTT, J., M. PRAŽÁKOVÁ, E. STUHLÍK, and Z. STUHLÍKOVÁ. In press. Acidification of lakes in Šumava (Bohemia) and in the High Tatra Mountains (Slovakia). Hydrobiologia.
- GESSNER, F. 1925. Das Plankton der Isergebirgstalsperren. Jahrbuch d. Naturfreunde in Reichenberg 48: 51-69.
- GESSNER, F. 1929. Die Biologie der Moorseen. Untersucht an den Moortalsperren des Isergebirges. Arch. Hydrobiol. 20: 1-64.
- JIRÁSEK, J., L. HOCHMAN, and B. LOSOS. 1959. Výzkum dystrofních vod na Liberecku. Závěrečná zpráva. Department of Fisheries and Hydrobiology, Agricultural University of Brno. 19 p. [Investigation of dystrophic waters in the Liberec region. Final report. In Czech.]
- KŘEČEK, J. 1991. Forests of Bohemia. Annual field report, EARTHWATCH, Boston. 21 p.
- LHOTSKÝ, O. 1963a. Vodní režim a vodohospodářský význam Jizerských hor. Severočeské museum, Liberec, 6: 1-30. [Water regime of the Jizera Mountains, and their water management significance. In Czech, summary in German.]
- LHOTSKÝ, O. 1963b. Dystrofní údolní nádrže Jizerských hor. Vodní hospodářství 13 (5): 166-168. [Dystrophic reservoirs in the Jizera Mountains. In Czech.]
- MOLDAN, B. 1991. Atmospheric deposition: a biogeochemical process. Rozpravy ČSAV, řada MPV, Academia, Praha. 101 (1): 1-108.
- PECHAR, L. 1987. Use of an acetone:methanol mixture for the extraction and spectrophotometric determination of chlorophyll-a in phytoplankton. Arch. Hydrobiol. Suppl. 78: 99-117.
- PERMAN, J. and O. LHOTSKÝ. 1963. Über das Vorkommen von Wasserblüten in einigen Wasserbecken Nordböhmens. Sborník Vysoké školy chemicko-technologické v Praze. Technologie vody 7 (2): 305-327.
- SLÁDEČEK, V. 1955. Zooplankton soušské nádrže s příspěvkem k metodice jeho kvantitativního stanovení. Časopis Národního musea odd. přírodověd. 124 (2): 151-160. [The zooplankton of the reservoir Souš with a contribution to the method of its quantitative determination. In Czech, summary in Russian and English.]
- STUHLÍK, E., Z. STUHLÍKOVÁ, J. FOTT, L. RŮŽIČKA, and J. VRBA. 1985. Vliv kyselých srážek na vody na území Tatranského národního parku. Zborník prác o Tatranskom národnom parku 26: 173-211. [Effect of acid precipitation on waters of the TANAP territory. In Czech, summary in Russian, German and English.]
- WRIGHT, R.F. 1983. Acidification of freshwaters in Europe. Wat. Qual. Bull. 8: 137-142.
- YAN, N.D. 1979. Phytoplankton community of an acidified, heavy metal-contaminated lake near Sudbury, Ontario: 1973-1977. Water, Air Soil Pollut. 11: 43-55.
- ZUBČENKO, D. 1955. Problém údolních nádrží s rašelinným povodím. Vodní hospodářství 5 (1-2): 38-48. [Reservoirs with dystrophic watersheds. In Czech.]

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APPENDIX B. Agenda

1. Opening of meeting.
2. Information from the Secretariat in Geneva on latest developments of the ICP's and report from working on effects.
3. Presentation of Intercalibration Report 92 06.
4. Presentation of draft Report 1991: Data.
5. Presentation of report on long term trends by Avis Newell.
6. Presentation of draft information brochure from the ICP on water effects.
7. Discussion of further development of the ICP on Waters based on the evaluation work.
8. Announcement of an international workshop on "Chemical and biological methods and importance of intercalibration" hosted by Russia June 7th-12th, 1993 at Baykalsktown (Baikal Lake).
9. National presentations of new findings relevant for the ICP on water effects.
10. Next meeting.
11. Other business.
12. Closure of the meeting.