ACIDIFICATION TRENDS IN SWEDISH LAKES:
AN ASSESSMENT OF PAST WATER CHEMISTRY CONDITIONS
USING LAKE SEDIMENTS

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Abstract
This thesis presents temporal perspectives of lake acidification in Sweden. Sediment records have been used to study timing, trends and causes of acidification, and two different techniques for assessing past lake-water acidity are presented.

A new technique for pH prediction, based on near-infrared (NIR) spectroscopy of surface sediments, is developed. This study shows that there is a pH related fraction of lake sediments that can be recorded by NIR analysis. Relationships between NIR spectra of surface sediment samples and measured lake-water pH values, and between NIR spectra of sediment cores and historical pH values inferred by diatom analysis, are modelled by partial least squares regression. The prediction errors of the models are comparable to those obtained by modelling of diatom and lake-water pH data. By further development NIR spectroscopy can become useful for inferring past pH, as well as several other lake-water parameters, from sediment cores.

Diatom-based predictive models, using multivariate calibration methods, are developed for inferences of lake-water pH, alkalinity and colour. These models are used to provide a regional assessment of recent lake acidification in the provinces of Västerbotten and Norrbotten, northern Sweden. The study shows that a pH decline has occurred in some southeastern lakes, but that most of the lakes have not faced significant changes in lake-water pH, alkalinity and colour. The inferred water chemistry changes are discussed in relation to atmospheric deposition and land-use.

In a study of eight acid-sensitive Swedish boreal-forest lakes a past-analogue approach is used to test whether contemporary expansion of conifers could cause lake acidification. Water chemistry changes associated with the natural pre-historic colonization and expansion of spruce in Sweden (~3000 years B.P.), at times of background atmospheric acidity, are inferred to evaluate the acidification ability of spruce per se on surface waters. This study shows that under natural, unpolluted conditions spruce colonization and expansion did not cause lake acidification.

In a synthesis of palaeolimnological acidification research in Sweden a general model for pH-development for acid clear-water lakes in southern Sweden is presented. The pH-development from the last deglaciation to present time can be divided into four different periods; (i) a natural long-term acidification period (12000 B.P. - 2300 B.P., or later), with a gradual decrease in pH resulting from declining fluxes of base cations from catchment soils; (ii) a human induced alkalization period (2300 B.P. - 1900 A.D.), with a pH increase due to human activities in the catchments; (iii) the recent acidification period (about 1900 A.D. - present), when pH decreased towards 4.5 due to acid deposition and possibly ceased land-use; and (iv) the liming period (1970s - present), when pH often increases to values above 7 following lime treatment to counteract acidification. The implications of these past pH changes for the concept of contemporary lake acidification and for liming policy are outlined.

Key words
palaeolimnology, acidification history, alkalization, acid deposition, land-use, lake sediments, near infrared spectroscopy, diatoms, multivariate calibration, Sweden

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"Brittens kvava stenkolssky
snart sig över landet breder,
svärtar allt det friska, gröna,
slår med giftig anda neder,
stjäl från dagen solens hägn,
strömmar såsom svavelregn
fordom över dömda städer."
Henrik Ibsen, Brand, 1866
(sv. övers.: Sigrid Elmblad)
List of papers

This thesis is based on the following papers which will be referred to by their Roman numerals.


II. Korsman, T. & Birks, H.J.B. Diatom-based water chemistry reconstructions: a comparison of reconstruction techniques. (Manuscript)


V. Korsman, T. Palaeolimnological investigation of lake acidification in northern Sweden. (Manuscript)

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Introduction

Acidification of soils and surface waters is one of the most serious environmental problems in northwestern Europe and eastern North America (Rodhe 1989; Andersson et al. 1992). To understand contemporary changes in lake-water acidity we need long series of data to separate observed variation from natural background. The most widely adopted approach for this is long-term monitoring, usually defined as a systematic measurement of variables and processes over a time scale linked to the environmental problems of interest (Spellerberg 1991). Here, there are two problems.

Firstly, how can the "natural" background levels of acidity in surface waters be determined? The problem of acid emissions and acidification has long been recognized. In 1852 an English chemist, Robert Angus Smith, noted that coal combustion resulted in sulphuric acid in the rain around the city of Manchester, which he in 1872 termed "acid rain" in his book on chemical climatology (Cowling 1982; Gorham 1989). Gorham and colleagues described the impact of the acid precipitation on aquatic ecosystems in the late 1950s and early 1960s (Gorham 1957; Gorham & Gordon 1960; 1963; Gordon & Gorham 1963). In Sweden, research about acidification started in the 1970s after Odén's alarming article on the long-distance transport of acidity (Odén 1967), though nation-wide monitoring of the impact of acid precipitation on lake ecosystems did not start until the 1980s (Bernes 1991).

Secondly, how can the cause of the observed recent changes in lake acidity be determined? The debate about causes of the recent surface water acidification has been focused on two alternative explanations: acid deposition from fossil fuel combustion, and acidification due to changing land-use and subsequent vegetational and soil changes (Seip 1986; Mason 1990). In most areas suffering from acidification, there is, however, a temporal relationship between changing land-use and increasing acid deposition, which makes it difficult to differentiate between these two alternative processes.

Palaeolimnological research using the sediment records offer an alternative approach that addresses these questions. The general aim of this thesis is to present temporal perspectives of lake acidification in Sweden, specifically;

- to develop predictive models for the assessment of past water chemistry conditions. A new technique based on near-infrared spectroscopy of surface sediments is presented. Diatom-based models for pH, alkalinity, and colour reconstructions using different calibration techniques are evaluated. (Paper I, II)

- to test the hypothesis that land-use and subsequent vegetation and soil changes cause lake acidification. Acidity changes associated with the natural pre-historic colonization and expansion of spruce in Sweden, at times of background atmospheric acidity, are inferred to evaluate the acidification ability of spruce afforestation per se on surface waters. (Paper III)

- to present general acidification trends for Swedish lakes. The pH-development from the last deglaciation to present time for acid clear-water lakes in southern Sweden is discussed in relation to natural development, land-use and atmospheric deposition. (Paper IV)

- to provide a regional assessment of recent lake acidification in northern Sweden. The inferred water chemistry changes are discussed in relation to atmospheric deposition and land-use. (Paper V)
Palaeolimnological methods for water chemistry inferences

Lake sediments provide the temporal record required to assess trends and to identify causes of lake acidification. Sediment material, mainly derived from the catchment and the lake itself, is continuously deposited at the sediment surface and thereby builds up an historical "archive" of the lake and its catchment. A variety of sediment components have been used by palaeolimnologists to provide information on lake-water acidity and related parameters. Analyses of biological remains, e.g. diatoms, chrysophytes, chironomids and cladocerans, provide information on changes in lake-water acidity (Charles et al. 1990a; Battarbee et al. 1990). Historical trends in atmospheric pollution from fossil fuel combustion can be assessed by analysis of spheroidal carbonaceous particles (soot), polycyclic aromatic hydrocarbons (PAH), sulphur and lead (Berge et al. 1990; Charles et al. 1990b; Jones et al. 1990; Renberg et al. 1990; Sweets et al. 1990).

In paper (I) a new approach for sediment-based analysis of lake-water acidity, near-infrared (NIR) spectroscopy, is presented. NIR analysis has previously been used in industry for analysing e.g. the moisture, protein, fat, and carbohydrate content in food products (Osborne & Fearn 1986). It is hypothesized that the acidification of lakes, resulting in major changes of flora and fauna, should be reflected in the sediment composition and thereby in the NIR-spectra of the sediments. This study shows that there is a pH related fraction that can be recorded by NIR-analysis and modelled by partial least squares (PLS) regression (Paper (I), Fig. 2). The prediction error of the calibration model using surface sediments is comparable to those obtained by modelling of diatom and lake-water pH. Possible solutions for developing predictive models for inference of past pH are outlined.

NIR spectroscopy has the potential to become a fast and simple method for inferring pH as well as several other lake-water chemistry parameters. Preliminary data screening, using the calibration dataset described in paper (I), indicates that several other parameters might be predicted with rather good accuracy. For nitrogen (Org-N, NO3-N, NH4-N, Tot-N), total phosphorus, total organic carbon, aluminium, magnesium, silicon and potassium, the prediction errors in relation to the measured range are comparable to that of pH. Information loss or distortion due to diagenetic processes might, however, complicate past reconstruction using sediment cores.

At present the main sediment constituents used for reconstruction of the acidification history are remains of diatoms and chrysophytes, which are acid sensitive siliceous algae. Diatoms have long been known as good pH-indicators. The first attempt to relate diatoms to the acidity of the lake-water was made in the 1930s by Hustedt, who classified diatoms into five classes according to their pH-tolerances (Hustedt 1939). This diatom-pH classification was used by Nygaard (1956), who proposed a calibration method for quantitative reconstruction of past pH. His method for pH-reconstruction based on acidity/alkalinity indices was further developed by e.g. Merilainen (1967) and Renberg & Hellberg (1982). The history of diatom-based pH-reconstructions is outlined by Battarbee (1984), Battarbee et al. (1986) and Renberg (1986).

Instead of grouping diatoms into pH categories, later improvements of diatom-based reconstruction models have made full use of the contemporary diatom data through multivariate techniques (ter Braak & van Dam 1989, Birks et al. 1990a). Today the approach most adopted for pH-reconstructions is weighted averaging (WA) regression and calibration (ter Braak 1987; Birks et al. 1990a). Using these techniques, rather low prediction errors (0.3-0.4 pH-units) can be achieved (ter Braak & van Dam 1989; Birks et al. 1990a; Dixit et al. 1991; Stevenson et al. 1991; Cumming et al. 1992a; paper II). The diatom-based reconstruction models have also been extended to other chemistry parameters of interest in acidification research. Predictive models have been developed for reconstruction of e.g. lake-water aluminium, acid neutralizing capacity (ANC), and dissolved organic carbon (DOC) (Birks et al. 1990b; Kingston & Birks 1990; Dixit et al. 1991). In paper (II) diatom-based predictive models for
alkalinity and colour, two parameters closely related to ANC and DOC, are presented. The weighted average approach has also been used for development of chrysophyte-based inference models (Smol et al. 1984; Cumming, et al. 1991; Cumming, et al. 1992a; 1992b), which might add valuable information to diatom-based inferences.

Recent improvements of palaeolimnological reconstruction models have combined the advantages of WA with those of partial least squares (PLS) regression. WA assumes unimodal relationships between the parameter to be predicted and the species data, which is a generally sound ecological assumption (ter Braak & Pretice 1988), but ignores residual correlation among the species (ter Braak & Juggins 1993; ter Braak et al. 1993). PLS, commonly used in chemometrics (Geladi & Kowalski 1986; Martens & Næs 1989), is a linear method for multivariate calibration that utilizes the residual structure in the species data. The new technique, weighted averaging partial least squares (WA-PLS), performs better than simple WA in data sets where the species responses to be used in calibration are influenced by a "nuisance" variable (i.e. an environmental variable other than the one to be predicted) (ter Braak & Juggins 1993; ter Braak et al. 1993).

In paper (II) diatom-based inference models, using different calibration techniques, are compared. These models, developed for assessment of water chemistry changes in northern Swedish lakes (Paper V), represent the first diatom-based inference models for this region. Large calibration data-sets, preferably specific to the region of application, are needed to obtain accurate predictions of past water chemistry conditions (Davis & Smol 1986). This is particularly important for palaeolimnological reconstructions in northern Sweden where many lakes are naturally acidic (Bernes 1991). The rarity of pristine, natural acid lakes in the modern calibration data-set developed for the Surface Waters Acidification Programme (Stevenson et al. 1991), was considered one of the problems resulting in no-analogue situations for pre-1850 samples from a Scottish lake (Birks et al. 1990a). In the palaeolimnological assessment of past water chemistry conditions in northern Swedish lakes none of the pre-industrial diatom assemblages appear to lack close modern analogues (Paper V).

The best model for the inference of lake-water pH is obtained by using simple WA and weighted averaging with tolerance downweighting (WAtol). For prediction of alkalinity there is valuable information in the residual structure after extracting the first weighted average component. This residual information results in a slight improvement of prediction errors by using a two component WA-PLS model (Paper II, Table 3). The diatom response to colour shows a linear relationship, which was best modelled by PLS.

The diatom-based models now developed for reconstruction of past pH have, presumably, reached their limit in terms of low prediction error. Two future challenges for diatom-based acidity reconstructions can be envisaged; (i) the development of simple routine methods for assessing past acidity status, and (ii) the development of methods for assessing acid episodes.

The diatom-based reconstruction techniques used today require skilful taxonomic expertise. Diatom identification to species level or below is required, which makes the counting very time-consuming. For certain problems addressed by lake managers, e.g. whether or not lime should be applied to a lake, it is perhaps sufficient to achieve a rough estimate of the acidity status of the lakes. It would therefore be desirable to develop simpler and more rapid methods for reconstructing past lake-water pH. Such methods could involve diatom data based on larger taxonomic groups (genera or functional groups), but would also require development of powerful calibration methods to extract information from data with lower "resolution".

Diatom-inferred pH represents an average pH-value for some years depending on the time resolution of the analysed sample, which is mainly influenced by the sampling strategy used and the degree of sediment bioturbation. Assessment of acid episodes from sediment data is a real challenge for palaeolimnologists. To obtain estimates of past acid episodes, either qualitative or quantitative, the diatom-based inferences must most likely be complemented with estimates derived by other inference methods. Cumming et al. (1992a) outline one future direction in their discussion on chrysophyte response to pH decreases associated with snowmelt.
Causes of lake acidification

The main hypotheses proposed to explain recent lake acidification are increased acid precipitation and changes in land-use. The former, defended by the majority of the scientific community, states that deposition of strong mineral acids, primarily sulphuric acid derived from emissions of SO$_2$ from fossil fuel combustion, is the main cause of surface water acidification. The land-use hypothesis states that natural soil forming processes (accumulation of humus) following e.g. abandonment of agriculture, is the major cause of the recent acidification of low-alkalinity surface waters. This hypothesis has been defended by Rosenqvist (1978) and Krug & Frink (1983). The increase of acid deposition has, however, occurred simultaneously with major changes in land-use in most areas, which makes it difficult to differentiate between effects of land-use changes and atmospheric deposition. Palaeolimnological methods may be the best way to test alternative acidification hypotheses (Seip 1986).

Considerable effort has been put into testing the influence of land-use changes on surface water acidification (Jones et al., 1986; Battarbee et al., 1985; Battarbee et al., 1990; Birks et al., 1990c; Paper III). All these reject land-use as the major cause of recent acidification. There are, however, some palaeolimnological studies showing clear impacts of catchment vegetation and soil changes on lake-water chemistry (Davis et al. 1983; Renberg et al. 1993; Paper III), but none of these studies provide evidence for land-use changes as the primary cause of acidification, as stated by Rosenqvist (1978) and Krug & Frink (1983).

In Paper (III) a past-analogue approach (time-for-space substitution) is used to test whether contemporary expansion of conifers can be involved in the acidification of lakes. For instance, in southern Sweden, where a large number of lakes are severely acidified, a considerable expansion of Norway spruce (Picea abies (L.) Karst.) has occurred during the last hundred years. Water chemistry changes associated with the immigration of spruce into Sweden (~3000 years B.P.) are inferred in eight lakes. This study shows that under natural, unpolluted conditions spruce colonization and expansion did not cause lake acidification. However, a slight increase in diatom-inferred DOC is recorded in three lakes. With the present levels of air pollution spruce afforestation on acid-sensitive soils may, however, contribute to surface water acidification due to the filtering of aerosols in the canopies.

Acidification trends

There are two general concepts of long-term pH-development in low-alkalinity lakes prior to recent acidification in the 20th century (Charles et al. 1990a); (i) pH has been rather stable over thousands of years in pre-industrial times and (ii) lakes have undergone a gradual long-term Holocene acidification. Neither of these two models seem correct for acidified southern Swedish clear-water lakes (Renberg 1990; Renberg et al. 1993; Paper IV).

Renberg (1990) recorded an abrupt increase in inferred pH in two southwestern Swedish lakes about 2000 years ago, and these higher pH conditions (~1 pH-unit above "natural" levels) lasted until the onset of recent acidification. This human induced alkalization period was recorded in ten additional lakes in a study aimed at assessing whether the observed trend is general for a larger area of southern Sweden (Renberg et al. 1993; Paper IV).

The general trend for pH-development presented in Paper (IV) distinguishes four different periods. In the late-glacial period (12000 B.P.) the lakes had high pH, which gradually decreased due to gradual soil acidification and declined fluxes of base cations from the catchment soils (Paper IV, Fig. 5). This natural acidification, with most rapid pH-decrease in the early post-glacial period, is consistent with other palaeolimnological studies on long-term acidification (Charles et al. 1990a). Whitehead et al. (1986) recorded a pH decrease from 7.5 to 5 in two North American lakes. In our study the maximum pre-human pH-decrease recorded was ~1.5, from 7 to 5.5.
The alkalization period (2300 B.P. or later - 1900 A.D.) is attributed to increased base saturation of soils and increased flux of base cations and nutrients to the lakes, due to human activities in the catchments. The start of this period coincides with the Iron Age agricultural expansion, which began in central settlement areas about 2000 B.P. spreading thereafter into marginal areas (Berglund 1991). Strong correlation between pH and pollen from plants associated with agricultural activities, and between pH and charcoal indicating fires, support the hypothesis of human impact on water chemistry of these lakes (Renberg et al. 1993). The processes resulting in increased base cation and nutrient transport from soils to surface waters are discussed by Renberg et al. (1993; Paper IV). Davis et al. (1983) found a similar alkalization due to anthropogenic disturbance in the catchments of three North American lakes.

At the beginning of the 20th century pH started to decrease in many lakes (Paper IV). This recent acidification period has been confirmed by several palaeolimnological studies in southern Sweden (see references in Paper IV) and also in other European countries and North America (Charles et al. 1990a). The pH did not, however, decrease below the "natural" pre-alkalization level until the 1960s in the lakes studied by Renberg et al. (1993; Paper IV, Fig. 8). Although the study by Renberg et al. (1993; Paper IV) showed that land-use is important in influencing lake-water chemistry, it is not suggested that the severe recent acidification with pH values of about 4.5 and fish kills in southern Swedish lakes is caused by land-use changes. Such low pH values never existed in these lakes before, and this major pH decline is only found in areas with high acid deposition. There are overwhelming evidence from a series of critically designed studies that acid deposition is the major cause of recent surface-water acidification (Jones et al., 1986; Battarbee et al., 1985; Battarbee et al., 1990; Birks et al., 1990c; Charles et al. 1990a; Sullivan 1990).

**Palaeolimnological environmental impact assessment**

Recent methodological advances have made palaeolimnology a powerful tool for environmental impact assessment and lake management (Smol 1992; Anderson 1993). These advances include e.g. development of quantitative models for environmental reconstructions and improved sediment sampling techniques.

In most regions of the world background data are not available to provide information on the lake status needed for management decisions, and this is particularly obvious in the case of acidification. The recent acidification started in the 19th century (Charles et al. 1990a) but the pH scale to describe the acidity of aqueous solutions was not developed until 1909 (Sorenson 1909). With the new tools palaeolimnologists can, however, answer questions such as: Has the lake acidified? When did the pH decline start? and What is the cause of the observed changes?

These questions have to be rigorously evaluated in order to obtain successful effects of acidification countermeasures. In Sweden more than 6000 lakes have been limed (Fleisher et al. 1993) to counteract acidification. The aim of this liming program is to improve water conditions so that the naturally occurring flora and fauna can persist or re-colonize the limed water (Nyberg and Thormelof 1988), which in chemical terms has been defined as raising the pH to >6 and the alkalinity to >0.1 meq l⁻¹. There is, however, increasing concern that some northern Swedish lakes being limed are naturally acidic (Bishop 1991; Kullberg et al. 1993). Furthermore, palaeolimnological studies have showed that liming does not necessarily restore the natural, pre-acidification flora of the lakes (Flower et al. 1990, Renberg & Hultberg 1992).

Regional assessment of acidification status using palaeolimnological techniques have been made in the Adirondack region, North America, (Sullivan et al. 1990; Cumming et al. 1992a) and in northern Sweden (Paper V). In the Adirondack studies, the palaeolimnological record showed recent acidification had occurred in almost all lakes with a contemporary pH <6. It was, however, also shown that the magnitude of acidification was slightly lower than that obtained from computer-based model estimates. They suggested that the role of catchment and in-lake neutralization of acidic inputs had been more important than previously assumed and that the pre-industrial conditions were more acidic than estimated from other models.
The palaeolimnological assessment of lake-water chemistry changes in northern Sweden (Paper V) showed that most of the lakes had not faced significant changes in pH, alkalinity or colour since pre-industrial time. The pH decline recorded in some southeastern lakes is discussed in the context of acid deposition and changed land-use. The importance of DOC on the acidity status of northern Swedish surface waters shown by Bishop (1991) and Kullberg et al. (1993) is supported by the relationship between diatom-inferred changes in colour and pH/alkalinity. The study highlights the importance of background data to understand contemporary changes in lake-water conditions.

References


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