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**Palaeolimnological analogues in defining target
assemblages for the recovery of acidified surface waters:
a desk study**

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Palaeolimnological analogues in defining target assemblages for the recovery of acidified surface waters: a desk study

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Executive Summary

- 1) Palaeolimnological techniques have been widely employed to study lake acidification. This approach has been central in testing the cause-effect relationship between acid deposition and lake acidification, and in assessing the magnitude and extent of surface water acidification across the UK.
- 2) Most of these palaeolimnological applications have been based on diatom analysis, and the use of diatom-pH transfer functions to make reconstructions of hydrochemical change in upland lakes associated with acidification.
- 3) Following the signing of the Second Sulphur Protocol, attention is now focusing on emissions reductions and the reversibility of surface waters acidification. There is a clear need for criteria against which to evaluate the recovery process.
- 4) In order to evaluate future recovery, Flower *et al.* (1997) have proposed a palaeolimnological technique for defining targets for the recovery of acidified surface waters. This is based on the technique of analogue matching of lake sediment diatom assemblages. Multivariate statistical methods are used to identify modern analogues for the pre-acidification diatom assemblages of acidified lakes. The chemical and biological status of modern analogue lakes can then potentially provide recovery targets for acidified systems.
- 5) This approach has been successfully applied to several acidified lakes, and modern analogue systems defined for the pre-impact (pre-acidification) status of these impacted sites. An advantage of the approach is that it can provide recovery targets for both chemical and biological status of acidified lakes.

- 6) Modern analogue matching as currently applied makes several key assumptions:
 - a) that analogue matches based on a single biological group (diatoms) effectively represent the hydrochemical and biological variation of low alkalinity systems;
 - b) that the modern data set used to identify modern analogues contains the range of hydrochemical conditions represented by the fossil assemblages;
 - c) that a suitable stable 'baseline' (pre-impact) status can be defined.

- 7) Prior to more comprehensive application of the modern analogue approach to acidified lakes in Britain, these assumptions require evaluation. Three studies are proposed:
 - a) Extension of the current modern lake dataset used for analogue matching by the inclusion of minimally impacted low alkalinity sites from northern Scotland.
 - b) Development of the current technique by including two more fossil groups (chironomids and cladocera) in the modern surface sediment dataset used in the matching procedure. This will allow the assumption that diatoms represent wider ecosystem variation to be tested, and should result in more robust analogue matches.
 - c) A study of hydrochemical and biological variation in the pre-acidification conditions of acidified lakes through high-resolution palaeolimnological study of selected Acid Waters Monitoring Network lakes. This will allow the stability of baseline (pre-acidification) conditions to be evaluated.

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Chapter 1: Introduction

1.1 Acid deposition, surface water acidification and palaeolimnology

The acidification of surface waters via atmospheric contamination by strong acid-forming compounds has been a major environmental issue for the last 20 years or so. Angus Smith (1852 cited in Battarbee *et al.* 1988) first coined the term 'acid rain' when he used it to describe the effect of coal combustion on air and precipitation chemistry at various industrial sites in the UK. The effect this would have on lake ecosystems was first discussed by Gorham (1958). Since then numerous studies have been published which have investigated the problems associated with acid deposition and have confirmed that lake acidification has taken place in large areas across Europe and North America (see for example Battarbee & Charles 1986). In a review of the available data on surface water acidification, Charles *et al.* (1990) draw upon examples from North America, the United Kingdom, Scandinavia and Central Europe (see Table 1). Studies have shown extensive acidification of many lakes within these regions. For example, the Palaeoecological Investigation of Recent Lake Acidification (PIRLA) (Charles & Whitehead, 1986) was a large-scale project that attempted to assess the extent and cause of acidification in North America. In the Adirondack Mountains National Park, New York, all 11 clearwater lakes from the region sampled as part of PIRLA where contemporary pH was below 5.5 have acidified as a result of the deposition of strong acids to their catchments.

In the UK, Battarbee *et al.* (1988) identified a range of lakes impacted by atmospheric contamination resulting in increased acidity levels. Acidification of UK surface waters was identified in central and northern Wales at sites on Lower Palaeozoic Sedimentary and Metamorphic rocks. Acidification was also identified in Cumbria, on Borrowdale volcanic strata, and in Scotland, Southeast of Loch Ness on granitic geology. There are also numerous highly acidic ponds and lakes in the English Pennines. Since this work, further studies have shown that lakes have acidified in areas previously thought to be unaffected by atmospheric deposition, such as lakes on the Cairngorm plateau (Jones *et al.* 1993) and remote sites in the north of Scotland (Allott *et al.* 1995).

Palaeolimnological studies (Charles & Whitehead, 1986; Battarbee & Renberg, 1990) have proved conclusively that acid deposition has resulted in the widespread acidification of surface waters. The timing of acidification in lakes has always occurred after the onset of the industrial revolution and the record of carbonaceous particle deposition of lake sediments clearly demonstrates atmospheric contamination from industrial sources prior to acidification (Rose, 1996; Rose *et al.* 1995). Other studies have provided understanding of the emission, transport, and deposition of pollutants and the processes of atmospheric chemistry (RGAR, 1997).

Table 1: Selected regions showing the extent of surface water acidification throughout North America and Northern Europe. (Main data source: Charles et al., 1990)

Geographic Region	Local Area	Lakes Acidified ?	Comments
United States of America	Adirondacks	Yes	11 clearwater lakes with pH <5.5. Lake with pH >6.0 have not acidified or have done slightly
	New England	Yes - recently	Trends are unclear
	Northern Great Lakes	Little or none Post 1900 pH decline in 4 Wisconsin lakes	Lakes are generally seepage lakes with low alkalinity (-38 to 80 $\mu\text{eq l}^{-1}$) and function differently to those of the Adirondacks
	North Florida	Little or none	12% of regions lakes have pH <5.0. 4 lakes studied were naturally acidic due to low cation exchange capacity and base saturation of catchment soils
Canada	Eastern Canada	Yes	Studies biased towards lakes close to point source emissions, e.g. Sudbury, Ontario
UK	Central & Northern Wales	Yes	Strong acidification (1-1.5 pH units in some lakes)
	Cumbria	Yes	An area of high acid deposition, though acidified lakes restricted to geologically sensitive areas, e.g. Borrowdale volcanic strata
	Southeast Scotland	Yes	On granite bedrock
	Northern Scotland	Yes	Sites that were thought to be relatively unaffected by acid deposition have been found to have acidified recently (e.g. Allott <i>et al.</i> 1995)
Rest of Europe	Scandinavia	Yes	Large scale loss of fish populations and damage to forests- many acidified lakes
	The Netherlands	Yes	
	Denmark	Yes	
	Germany	Yes	

1.2 *International Emissions Reductions*

The acceptance of the cause-effect relationship between acid deposition and acidification has led to international efforts to reduce acid emissions. In 1979, the Convention on Long-Range Transboundary Air Pollution (LRTAP) was adopted to implement measures to reduce the levels of sulphate emissions from industrial sources. Currently there are 40 signatory countries to LRTAP, which has identified principles for international co-operation and for the abatement of the emissions of pollutants.

Under the LRTAP convention, a number of protocols have been developed that commit member states to certain abatement actions. The early protocols limited emissions of sulphur dioxide (the 1985 Helsinki Protocol) and oxides of nitrogen (the 1988 Sofia Protocol). All countries were required to cap their emissions or reduce them in relation to a given reference year (see Table 2). In 1994, the Second Protocol on the Further Reduction of Sulphur Emissions was adopted. This, the Oslo Protocol, introduced a new method of setting reduction targets by incorporating critical loads, an effects based approach to deposition (Bull, 1995). Critical loads maps represent the sensitivity of ecosystems to given levels of modelled acid deposition. Using modelled deposition across Europe for given abatement strategies, critical load exceedence maps indicate the effect on ecosystems for that level of abatement. Setting abatement targets for international protocols now involves the benefits to ecosystems that a given reduction will involve, and through Integrated Assessment Modelling cost effective abatement strategies have been developed.

Currently a further Protocol is being developed for emissions of nitrogen oxides. This protocol will be 'multi-pollutant/multi-effect', as sources of nitrogen oxides include acid deposition, ammonia and ozone, and cause a range of environmental disturbances (e.g. acidification, eutrophication, and direct effects to human and vegetation health (RGAR, 1997).

The UK, whilst not signing up to the Helsinki Protocol, has met with the protocol's requirements by reducing emissions of sulphur dioxide by 35% (RGAR, 1997). A national strategy has also been developed to outline the ways in which the UK will meet its obligations to the Oslo protocol. In the light of these international reductions in air pollutants, the emphasis is now shifting towards the recovery of acidified surface waters. There is now a need to investigate the process of recovery in acidified surface waters to evaluate and model the response of surface waters to reduced acid deposition. Given that the protocols arranged under the LRTAP convention are having success in reducing the level of acid deposition to catchments across Europe and North America the role of recovery is now central to the acid deposition debate.

The aim of this report is to evaluate the modern analogue approach to setting targets for the recovery of acidified surface waters. The approach uses palaeolimnological and multivariate techniques to compare one aspect of the flora or fauna of an acidified lake with the flora and fauna of other lakes. To identify potential targets for recovery, the technique allows the selection of a baseline or pre-impact condition for an acidified lake and the comparison of this to the modern flora and fauna of other lakes. Lakes identified using the technique are known as modern analogues. Identification of modern analogue sites potentially allows chemical and biological targets to be set for recovery.

Table 2: Current protocols developed as part of the LRTAP convention to curb the emissions of sulphur dioxide and oxides of nitrogen implicated in the acidification of surface waters across Europe and North America

Protocol	Year Adopted	Year in Force	Requirements of the Protocol
EMEP	1984	1988	The Protocol on Long-Term Financing of the Co-operative Programme for Monitoring and Evaluation of Long-Range Transmission of Air Pollutants in Europe. Collates information on deposition and emission inventories supplied by member states, and develops transport models for pollutants.
Helsinki	1985	1987	The Protocol on the Reduction of Sulphur Emissions or Their Transboundary Fluxes by at least 30%. Committed parties to a 30% cut in SO ₂ emissions by 1993 based on 1980 levels.
Sofia	1988	1991	The Protocol concerning the Control of Emissions of Nitrogen Oxides or Their Transboundary Fluxes. Commits member states to bring back NO _x emissions to their 1987 levels by 1994
Oslo	1994	Not Yet	The Second Protocol on the Further Reduction of Sulphur Emissions. Requires different % reductions from member states based upon an effects based concept know as the critical loads approach. The UK is required to reduce sulphur emissions by 80% against 1980 levels, by 2010.

The structure of this report is as follows. The contribution of palaeolimnology to the acidification debate is described in Chapter 2. Palaeolimnology has been instrumental in confirming the cause-effect relationship between acidification and deposition across Europe and North America. Chapter 2 also describes the techniques used to reconstruct hydrochemical variables from fossil data, and reviews the biological groups preserved in fossil records. The concept of ecosystems restoration is discussed in Chapter 3. Chapter 4 describes the technique of analogue matching and describes a preliminary application of the approach for northern European lakes developed by Flower et al. (1997).

The report also considers the question of what makes a suitable restoration target given that in the UK there are very few pristine ecosystems. Human activities have influenced the natural

landscape for thousands of years. There is also increasing evidence that acidification may be influenced by climate change in remote alpine lakes. The degree to which climate change has impacted upland lakes in Europe and the UK in general is addressed. Chapter 5 details the available data in this area, as well as outlining the other assumptions of the modern analogue approach.

Chapter 6 briefly outlines ways in which the method can be applied and improved, and how the validity of the assumptions of the approach can be tested.

Recovery from acidification will be, in most cases, restricted to natural recovery. Identifying a period when recovery has taken place, to the satisfaction of both the environmental groups and those with economic interests, is required if recovery can be defined as being complete. The modern analogue approach and palaeolimnology allows us an opportunity to investigate recovery in acidified systems, and to evaluate the success of emission reductions.

Describing the biological target for recovery is more difficult. Directly, palaeolimnology is limited to identifying communities of those organisms that preserve well in sediments. Palaeolimnology, however, can help to select modern lakes that are analogous to the pre-impact conditions of presently acidified lakes. When coupled with the detailed knowledge of aquatic ecosystems gained from investigating analogue lake systems, the tools required for setting targets for recovery and evaluating the progress made towards those targets would then be at our disposal.

Chapter 2: Palaeolimnology

2.1 What is Palaeolimnology?

Palaeolimnology is the study of the history of lakes. It is concerned with how lakes have changed over time and with understanding those changes, primarily through an assessment of the historical record contained in the sediments of lakes. Palaeolimnology has been used extensively to study the development of lake systems (ontogeny).

Over the last few decades numerous sampling and analytical techniques have been developed that enable palaeolimnologists to obtain sediment cores containing undisturbed sediment records (e.g. Glew, 1991, Charles et al., 1994). These can then be analysed for a variety of fossil organisms and geochemical markers using appropriate laboratory methods (Berglund, 1986). Dating techniques using ^{210}Pb (Appleby et al., 1986) and other radiometric methodologies (Charles et al., 1994) can be used to apply chronologies to the changes observed in the sediment record. This allows the timing, magnitude and rate of change to be determined.

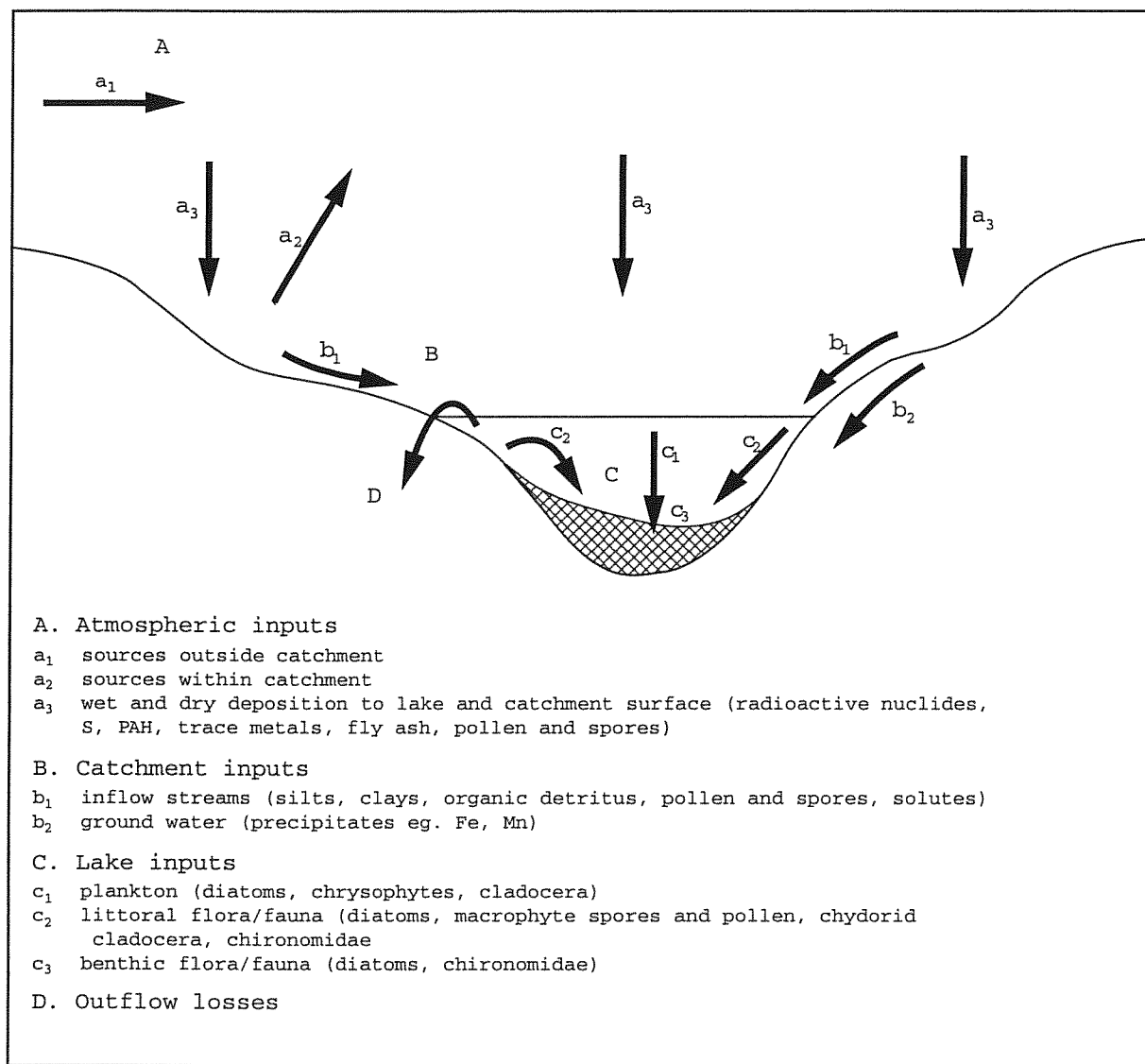
Sediments found within lakes are derived from three main sources; atmospheric inputs, catchment inputs and lake inputs (Figure 1). Atmospheric sources include wet and dry deposition both from sources within the catchment and those from outside. Inflows from streams and groundwater contributions make up the catchment component, and the planktonic, benthic and littoral flora and fauna of the lake contribute to the lake input fraction (Battarbee et al., 1988).

Organisms that are preserved in the sediment of a lake, such as diatoms or cladocera, can be analysed for changes in community structure over time. Assessment of the modern distribution of these organisms can be related to the hydrochemistry of the waterbody, and optima and tolerance ranges for individual species can be calculated (Battarbee, 1991). These optima and tolerance ranges can be applied to the fossil communities by means of transfer functions (e.g. see Battarbee, 1991; Bennion, 1994; Birks et al., 1990b; Charles et al., 1994; Dixit & Smol, 1994). These enable changes in the hydrochemistry of a lake to be determined from the fossil community assemblages deposited to the sediment at that time (Anderson, 1995).

These techniques are now widely used within palaeolimnology. They have been used to determine changes in waterbodies related to many hydrochemical variables, including acidity (pH) (Dixit et al., 1992; Charles et al., 1990; Allott et al., 1995; Cumming et al., 1994; Dixit et al., 1992), total phosphorus (TP) (Bennion, 1994; Dixit & Smol, 1994), Dissolved Organic Carbon (DOC) (Kingston & Birks, 1990), salinity (Fritz, 1990), aluminium (Al) (Dixit et al., 1992), as well as enabling changes in climate and temperature to be inferred from the sediment record (Lotter et al., 1997).

Analyses of the sediment record for contamination by pollution indicators seen as heavy metals (Pb, zinc etc.) (Davis et al., 1983) spherical carbonaceous particles (SCP) (Rose, 1996; Rose et al., 1994) and inorganic ash spheres (IAS) (Rose, 1996), have allowed the causal mechanisms for pollution induced changes to be determined, especially in the case of the acute acidification of sensitive surface waters in Northern Europe and North America.

Figure 1: Schematic cross-section of a lake and its catchment showing sources and pathways of material found in lake sediments (from Battarbee et al., 1988).



Palaeolimnological techniques have been used to assess changes in both the biology and, more recently, the chemistry of lakes. They have been used over timescales ranging from millions of years (e.g. the study of Lake Baikal) to sub-decadal (e.g. Allott et al., 1992). The techniques used in palaeolimnology have also been used in monitoring projects attempting to evaluate current changes in aquatic systems. Dixit and Smol (1994), present four transfer functions that can be used to reconstruct the hydrochemistry of lakes from the enumeration of siliceous microfossils from lake sediments. In this way, inferences about the current, changing, hydrochemical nature of surface waters can be made using the biological record of organisms contained within recent lake sediments.

The short-time series used in many recent ecological studies are often unsuitable for monitoring studies (Smol 1995). Natural variability is difficult or impossible to distinguish from data of insufficient quality or time span. Without such data, environmental managers often have difficulty in determining the trajectory or the causes of degradation, let alone the likely effects of recovery or targets for their mitigatory efforts (Smol, 1995). Smol (1992) argues that the task of assessing ecosystem health is made harder without suitable and available long-term data. Palaeolimnological data provides such long-term environmental, hydrochemical, lithological and biological data, enabling the baseline (pre-impact) conditions of a surface water to be established (see Chapter 3). These baseline states of surface waters provide invaluable information on the degree of change present at a site. Where anthropogenic pollution is suspected or known to have degraded a surface water, targets for the restoration of the site can be established with regard to these baseline conditions.

Palaeolimnological studies are often based upon studies of the fossil remains of biological indicators, the aquatic flora and fauna living in a lake. Palaeolimnological sediment records of change are averaged over time, the species composition dynamically in tune with the predominate or average hydrochemical or physical conditions of the lake. Short-term cycles, such as diurnal or diel changes, can influence the water chemistry or the biological sample taken during spot sampling measures (Anderson & Battarbee, 1994). However, the historical record of lake sediments is time averaged, and, therefore, does not contain the degree of noise inherent in spot sampling records.

The wide variety of organisms preserved in the sediments and the range of chemical, lithological and biological assays that can be applied to the sediment allows for a huge range of experiments and hypotheses to be tested using palaeolimnological techniques.

2.2 Palaeolimnology and lake acidification

Palaeolimnology played a key role in the study of lake acidification. During the early 1980s, there was considerable debate surrounding the cause of fish stock declines in upland lakes. Many scientists, especially those from Scandinavia where the effects of acid deposition were first described, believed that acid emissions were to blame for the recent acidification of surface waters in Europe and North America. A number of alternative hypotheses were formulated, however, in what was a highly charged scientific and political debate.

One such claim was that acid lakes were the result of long-term, natural acidification processes (Pennington, 1984). It was claimed, that after lakes were formed when the glaciers retreated and climate warmed at the start of the Holocene, lakes acidified as weathering of soils led to the progressive leaching of base cations. Soils would gradually acidify and the subsequent

runoff would slowly lower the pH of lakes. The basis behind this hypothesis was that there are many lakes that are presently acid but that had no historical record of fish stocks, evidence for long-term acidification.

Other claims surrounded the large-scale changes in land-use within upland areas of Northern Europe (Rosenquist 1978). Decreased grazing in lake catchments may have led to an increase in heathland vegetation and with it an increase in acid soils. Evidence from Sweden (Renberg et al. 1993a,b) has shown considerable influence on lake hydrochemistry by changes in land-use. Many lakes in southern Sweden were shown, by hydrochemical reconstruction using a diatom-pH transfer function, to have an alkaline phase prior to the onset of acidification in the 19th Century. The alkaline phase was the result of land-use practices that no longer exist in modern day Sweden.

More credibility was placed upon claims that recent acidification of surface waters was the result of recent afforestation. Streams draining from afforested catchments have been shown to be more acidic than those draining non-afforested areas (e.g. Harriman & Morrison, 1982; Stoner & Gee, 1985). Many of the afforested regions of the UK are located in areas of high sensitivity to acid deposition with base-poor bedrock, slow weathering rates and high rainfall (Kinniburgh & Edmunds, 1986). Consequently, many areas of afforested upland Britain have low acid buffering capacities. Tree growth processes (Nilsson et al., 1982), enhanced scavenging and foliar uptake of sulphur dioxide (Lindberg & Garten, 1998), as well as land improvement measures prior to tree planting (Hornung & Newson, 1986), have all been proposed as mechanisms which promote acidification.

Palaeolimnological studies have demonstrated that, whilst afforestation can result in enhanced acidification, it cannot account for the widespread, rapid acidification of European surface waters. Kreiser et al. (1990) studied four Scottish lochs, two with afforested catchments (Loch Chon and Loch Doilet) and two with moorland catchments (Loch Tinker and Lochan Dubh). Loch Tinker and Loch Chon are located in the Trossachs region of Scotland, an area of high acid deposition, whereas Loch Doilet and Lochan Dubh are located in an area that receives low levels of acid deposition. Kreiser et al. (1990) demonstrated that acidification occurred gradually from c. 1850 at Loch Tinker. When the site was afforested in the 1950s, there was an increase in the rate of acidification. The most rapid period of change occurs in the 1960s following canopy closure when pH fell from 5.8 pH units to 5.2. Loch Tinker shows some signs of early decline in the planktonic *Cyclotella* flora of the lake, an early biological indication of acidification. There is further acidification up to the 1930s, but the acute, recent (post 1960) acidification and associated diatom changes shown to occur in Loch Chon do not occur in Loch Tinker. There was no overall change in inferred pH or diatom composition above a depth of 8cm (1930) in the sediment core. pH reconstruction at Loch Tinker shows that pH fell from 6.6 to 5.7 by 1930, but then fluctuated around 5.6-5.7 until the present day. Carbonaceous particle records from the two lochs show that both were receiving high levels of atmospheric contamination by the 1940s, which is prior to the afforestation of Loch Tinker.

The study by Kreiser et al. (1990) demonstrated that in areas of high deposition afforestation can lead to further (or more severe) acidification of soils and surface waters. The tree canopy increases scavenging of particles from the atmosphere, which can enhance the flux of atmospherically derived acidity to the catchment. Afforestation in the catchment of Loch Chon had such an effect leading to further acidification after c. 1960s. Loch Tinker stopped acidifying after c. 1930, even though it was experiencing similar levels of deposition as Loch

Chon. The diatom sub-fossil flora of Loch Doilet, the other Loch in the study with an afforested catchment, does not indicate any further acidification following afforestation. Loch Doilet receives lower levels of deposition than Loch Chon and the scavenging effect of the canopy has had little or no effect on the chemistry of Loch Doilet. The two Lochs with moorland catchments both acidified after c. 1850, precluding afforestation as the primary cause of surface water acidification.

Palaeolimnological techniques were also used to evaluate the claims that acidification of surface waters was the result of deposition of atmospherically derived acids to lakes and the catchments draining into them (e.g. Battarbee et al., 1990). Recent lake acidification is strongly correlated with evidence of atmospheric contamination from lake sediments (Battarbee, 1990). Records of carbonaceous particles document the impact of atmospheric contamination to surface waters (Rose et al., 1995; Rose, 1996). Created during high-temperature burning of fossil fuels in power stations, carbonaceous particles first enter the sediment record during the nineteenth century as the industrial revolution took hold in the UK. Carbonaceous particle concentrations drastically increase in the post-war period (1950s), reaching a peak around 1970. Concentrations decline after the 1970s where oil availability was reduced during the Oil Crisis and because of international efforts to curb emissions in the late 1970s and early 1980s.

Battarbee (1990) demonstrates that the trend in surface water acidification seen in UK lakes parallels the trends in carbonaceous particle concentration identified from the sediment record. The first indication of atmospheric contamination occurs around c.1850 and rarely before 1800 in UK lakes. This is also the period where a response of the diatom flora to the contamination is first observed. Contamination levels increase rapidly after c. 1940, and the diatom flora shows a rapid change to more acid tolerant species after this time. Lakes that have little evidence of atmospheric contamination in the sediment record, such as Loch Corrie nan Arr (Battarbee, 1990), are also those that have not acidified, for those areas that receive high loadings of atmospheric deposition acute acidification of the surface waters has been identified from the sediment record (Battarbee, 1990)

Palaeolimnology has played a crucial role in determining the cause and effects of lake acidification. Without the high resolution historical record contained in the sediment of lake basins and interpreted by palaeolimnologists it would have been particularly difficult or impossible to prove the role emissions from power stations played in acidifying surface waters.

Since SWAP and PIRLA, the emphasis has shifted towards identifying chemical and biological recovery from acidification. Dixit et al. (1989) showed clear recovery in Swan Lake using chrysophytes, a siliceous type of algae (see below). Swan Lake, situated near to the Sudbury smelters in Ontario, had acidified because of the large amount of SO₂ and toxic metal pollution emitted from the smelters. Dixit et al. (1992) demonstrated similar recovery in three other Sudbury lakes, this time using both diatom and chrysophyte remains. Recovery was attributed to the reduction in emissions from the Sudbury smelters where emissions of SO₂ had fallen by 50%. This reduction has been attributed to a reduction in the activity at some of the smelters and improvements in emissions control and taller stacks at the others. Evidence of recovery in sites remote from point source pollution was described by Allott et al. (1992) for the Round Loch of Glenhead. In cores with high sedimentation rates recent reversal of the diatom floristic trend towards dominance by a few acid tolerant forms was seen. These studies indicate that recovery from acidification should happen in lake systems following emissions

reductions, though detecting this may be more difficult in sites remote from point-source emissions. It may take many years before significant changes in the hydrochemistry and biology of lakes are observed.

2.3 Hydrochemical Reconstruction using Palaeolimnology

It has long been recognised that by studying the autecology of biological indicators for various hydrochemical variables you can classify specific groups of organisms that are found together under similar hydrochemical conditions. Taking this a step further allows us to determine the hydrochemistry of surface waters through an analysis of the organisms living in them. Earlier attempts at hydrochemical reconstruction were based on qualitative assessment of species. In recent years, however, more sophisticated statistical techniques have been used to provide accurate, robust hydrochemical reconstructions from fossil records that have a firm ecological basis.

The Surface Waters Acidification Project (SWAP) (Mason, 1990) and the Palaeoecological Investigation of Recent Lake Acidification (PIRLA) (Charles & Whitehead, 1986) projects developed new methods of reconstructing hydrochemical variables from the diatom species assemblages found within the sediment record.

Early approaches to pH reconstruction had been made qualitatively using Hustedt's pH classification system using diatoms. Nygaard (1956, cited in Battarbee et al., 1986) further enhanced this approach by developing indices. These indices were based on ratios of the percentages of diatom valves in Hustedt's pH categories. Meriläinen (1967) further developed quantitative approaches for reconstructing the acidity of surface waters using the relationship between the \log^{10} of index values (e.g. index α) and the measurements of lake water pH using regression analysis. The slope and intercept of the regression equation are then used to predict lake pH. Renberg and Hellberg (1982) derived Index B, again based on pH categories, which can be used to predict pH. Index B uses more information than Index α , and is less reliant on alkaline taxa that are rare or absent in acid lakes. Multiple linear regression of the optima and tolerance ranges of biological indicators for pH has also been used successfully to reconstruct the historical record of pH change from sedimentary records (Charles, 1985; Flower, 1986).

Whilst multiple regression has been useful in providing accurate pH reconstructions, it has two major drawbacks; the technique is not ecologically realistic and, because class-interval groups are used, individual species data may be ignored.

The technique that was developed for SWAP and PIRLA (e.g. Birks et al., 1990b) was based around weighted averaging (WA) regression and calibration (ter Braak, 1987). Weighted averaging regression and calibration overcome many of the problems of other calibration methods (Korsman & Birks, 1996). WA assumes a unimodal relationship for the response of species to explanatory variables. This is considered to be a sound ecological assumption (ter Braak & Prentice, 1988). Other regression and calibration methods assume linear responses to explanatory variables and, therefore, do not represent ecological functioning as well as WA. WA also maximises the covariance between the diatom data and the measured environmental variables (Korsman & Birks, 1996). This is the same approach used in direct gradient analyses such as canonical correspondence analysis (CCA). Indirect gradient methods attempt to maximise the variance only with the diatom data and, consequently, some information may be

lost when only the first few components are used for regression. If more components are used, multi-collinearity problems are introduced to the analysis (Korsman & Birks, 1996).

There are some weaknesses in WA, but these have recently been addressed with the development of a new variation of WA; Weighted Averaging Partial Least Squares (PLS) regression. WA-PLS uses the residual structure of the species data within the regression and calibration procedure to improve the predictions made using the technique (ter Braak & Juggins, 1993). Simple WA fails to accommodate this extra data and the predictive power of the WA calibration models produced using this method may not be as accurate as those developed using WA-PLS.

The calibration approach involves a two-step process. Firstly the relationship between the species data and the measured environmental variables (the predictor variables) is established using WA or WA-PLS regression. The relationships derived from the regression technique are then regressed or calibrated against the fossil data to predict pH from the fossil assemblages using inverse WA or WA-PLS regression.

To test the transfer function model, the predicted results are compared to a set of observed data. Strictly, the model should be tested against independent data, not against the data from which the model was derived. However, Birks et al. (1990b) demonstrated the techniques of 'Bootstrapping' or 'leave-one-out jack-knifing' that estimate the true error of the model. These methods achieve this by taking a sub-sample of the training set to compare with the observed data, thus forming an independent test of a model's predictive power. These techniques are computer intensive, with c. 1000 calibrations being run to test the error of prediction in a transfer function. Bootstrapping is especially suitable as the whole dataset is used to test the model.

2.4 *Sub-fossil remains found in lake sediments*

Palaeolimnological techniques are dependent upon the preservation of organisms in the sediments of lake systems. Not all conditions are suitable for the preservation of organisms and the fossils found in lakes can be dissolved or broken (Cameron, 1995; Flower, 1993). Some groups of organisms are not preserved within the sediment record. Preservation can also be dependant on the environmental characteristics of the lake. For example, ostracods are not found in surface waters that have low calcium concentrations because in these conditions, their ability to grow a strong calcite shell is impaired and they are less protected against predation. However, a number of important species groups are reliably preserved in the sediments of low alkalinity lakes. It is upon these organisms that the majority of the development of the techniques has taken place. These groups are described below with examples of their use in palaeolimnological studies.

2.4.1 Diatoms (Bacillariophyceae)

Diatoms are single celled, golden-brown algae characterised by an external siliceous cell wall, or frustule, in which silicic acid has been dehydrated and polymerised to form silica particles (Wetzel, 1983). This frustule consists of two overlapping valves connected by bands of silica known as girdle bands. The frustule is often highly ornamented with various species exhibiting different employment of features. These include the raphe (a slit in the length, or part of, the cell), the pseudoraphe (a depression in the axial areas of the cell wall) or striae (lines on the valve face composed of holes of complex structure within the cell wall). It is this

ornamentation on the valve and its general shape and nature that form the basis of all taxonomic separation of diatom species (Barber & Haworth, 1981). Cell morphology varies between genera and species, and even allows varieties and forms of species to be distinguished.

Diatoms are found almost everywhere that light and moisture occur, including virtually all marine, brackish and freshwater environments, as well as soils, ice, and attached to rocks and other substrates within spray and splash zones near water. Diatoms live singly or form colonies, usually secreting a mucilaginous material that covers the frustule and allows the diatom cells to attach to one another or to the benthos (Wetzel, 1983). Diatoms live on a variety of substrates, as well as in the plankton of lakes. They occupy habitats on rocks and stones (the epilithon), on aquatic macrophytes (epiphyton), sand grains (epipsammon), the sediment (epipelon) and on mosses (epibryon).

Due to the siliceous nature of the frustule, it is resistant to a certain degree of chemical attack and is usually well preserved within lake sediments. The relative ease with which diatom samples can be taken and analysed, and the variety and beauty of the various forms of frustule, have resulted in diatoms being a long studied member of the limnic flora (Battarbee et al., 1986). Diatom autecology has largely been described in the literature and observations regarding tolerances and optimal abundances for a variety of hydrochemical variables have been made. Diatoms have been demonstrated to respond to a number of environmental and hydrochemical variables (e.g. Battarbee, 1984; Dixit & Smol, 1994), including those not related to the acidified nature of many lakes in Northern Europe and North America, such as salinity (Fritz, 1990) and TP (Bennion, 1994; Engstrom et al., 1985).

The response of the diatom community to acidification has been examined via inferences from the sediment record and from laboratory and whole lake experiments. It has been demonstrated that diatoms respond markedly and quickly to changes in pH (e.g. Flower & Battarbee, 1983; Battarbee, 1984; Flower, 1986), alkalinity (Charles et al. 1994), Al and DOC (Kingston & Birks, 1990). These variables represent hydrochemical components that change during the acidifying process.

2.4.2 Chrysophytes (Chrysophyceae)

Chrysophytes are another type of algae, found in the plankton, and are characterised by a golden brown colour. Most of the Chrysophycean algae are unicellular organisms possessing one or two flagella (Wetzel, 1983). Many species do not possess a cell wall and are bounded instead by a cytoplasmic membrane or are covered by a coating of tiny siliceous or calcareous plates and scales. These siliceous scales are well preserved in the sediment and form the basis of species identification being distinguishable to species level.

Chrysophytes have been used as biological indicators (Kristiansen, 1986) and in palaeolimnological investigations including studies of pH change and lake acidification (e.g. Siver & Hamer, 1990; Facher & Schmidt, 1996). For example, Smol et al. (1984) demonstrated the use of Mallomonadacean microfossils in the analyses of past changes in lake acidity. The genera *Mallomonas* are a group of flagellated algae of the class Chrysophyceae. In the upper sediments of a core from Deep Lake, Adirondacks, USA, the contribution of *M. crassisquama* to the total (%) scale count decreased, whilst the contributions of *M. hindonii* and *M. hamata* increased, the former becoming the dominant source of chrysophycean scales

to the sediment. This shift in species contributions to the total scale count was attributed to a progression from circumneutral conditions to those of lowered pH. From the 38 lakes in the Adirondacks assessed by Smol (1980) for Mallomonadaceae microfossils, *M. hindonii* and *M. hamata* were found to be indicative of acidic lakes, whilst *M. crassisquama* was rarely found in lakes with a pH <5.0. Smol et al. (1984) substantiated their hypothesis of a response in the mallomonadaceae microfossils to increasing acidity with an analysis of the diatom microflora of Deep Lake, which showed a similar response to the decreasing pH.

Dixit et al. (1992) used changes in the chrysophyte and diatom communities to infer long-term trends in pH and metals in three lakes in the Sudbury region of Ontario, Canada. The chrysophyte community was shown to respond more quickly and more markedly to acidification than the diatom communities in the lakes. Vernal blooming and euplanktonic chrysophytes tracking spring depressions in pH are thought to account for the closeness of the chrysophyte inferred data to the observed pH trend (op cit.). Chrysophytes may infer lower pH values than those of diatom inferred data because the chrysophytes respond to the spring water chemistry when pH values are lower and aluminium levels higher, whilst diatoms infer average, ice free conditions (op cit.).

2.4.3 Chironomids (Chironomidae)

Adult chironomids are delicately built, long-legged, frequently brightly coloured members of the non-biting midge group. The family Chironomidae is typified by a thorax that overhangs the headpiece of the adult chironomid. They commonly form swarms hovering about water bodies at dusk.

Nearly all chironomid larvae are aquatic. They have worm like bodies with a pair of pro-legs on the prothorax and another pair on the caudal segment, with the penultimate segment sometimes bearing filamentous gills. They are found in a variety of colours, with some species (Chironominae) adopting a 'blood-red' colour; the result of haemoglobin-like pigments, which allow the larva to survive in oxygen depleted environments.

Chironomids are found in two habitats within lakes; the littoral benthic and the offshore, or profundal, benthic. Thienenmann (1920 cited in Frey, 1988) described a successional series of lakes based upon chironomid taxa from the offshore communities. At one end were the oligotrophic lakes represented by the *Tanytarsus* community, and at the other were the lakes with low dissolved oxygen levels, where communities of *Chironomus* were found. Today, oligotrophic lakes are characterised by a *Tanytarsus lugens* community because in Thienenmann's original work complications arose as the genus *Tanytarsus* primarily occupies the shallower water inshore, with only a few, specialised species occupying the offshore areas (Frey, 1988). Most of the littoral species don't move into the profundal range at all. This is probably due to low water temperatures in the profundal benthic region (Frey, 1988).

The remains of chironomid larvae found in lake sediments are derived from two sources; the actual remains of dead larvae and their molt stages (Frey, 1988). The chironomid larvae have four stages, the skin and head capsule are split allowing the larvae to eject a stage, thus allowing it to continue growing. The type of species will determine the degree of preservation of the head capsule in lake sediments (Brooks, pers comm). Species from the subfamily Tanypodinae have more robust head capsules and are usually found intact. The head capsules

of species from the Orthoclaadiinae, on the other hand have a more brittle suture on the central axis and are often found to be split in the sediment.

Chironomids are useful as environmental indicators (Seather, 1979). The head capsules are chitinous and are found abundantly in the sediments of most lakes and can be identified to generic level. The chironomid fauna of lakes is species rich, which allows the transfer function approach to be applied for environmental reconstruction.

Chironomid remains have been largely used to infer changes in climate on a Holocene timescale and to construct chironomid-based climate prediction transfer functions (Walker et al., 1991; Walker et al., 1991; Wilson et al., 1993; Lotter et al., 1997). Lotter et al. (1998) used chironomids as part of a multi-proxy study of nutrients in the Swiss Alps. They found a strong relationship between chironomid communities and TP, with their predictive model using chironomids having a $r^2 = 0.68$ (op cit.).

Chironomids have also been used to investigate lake acidification (Henrikson & Oscarson, 1985; Henrikson et al., 1982), and studies have attempted to look at the responses of chironomid populations to increasing acidity (Griffiths, 1992). However, chironomids may not be responding to changes in acidity, rather to degrees of environmental stress (Brooks, pers comm.). The removal of acid sensitive species allows other species to dominate the chironomid fauna. When pH returns to higher levels (e.g. through liming, Brooks, pers comm.) these (so called) acid tolerant species remain in the sediment record after liming, even though the pH is now much higher. Therefore, the response shown by some chironomid species appears to be not just a response to hydrochemical change but to some sort of disturbance criteria.

2.4.4 Cladocera

Cladocera, or water fleas, are small crustaceans. The thorax of a cladoceran is enclosed within a folded oval carapace, and bears five or six pairs of limbs. Some species of cladoceran occur all the year round, others only develop from eggs during warmer periods or more-favourable conditions. The females reproduce rapidly, producing 'summer' eggs that require no fertilisation from the male. These eggs develop quickly from the brood pouch contained behind the carapace (Wetzel, 1983). When winter approaches or unfavourable conditions arrive, the females produce 'winter' eggs that require fertilisation from the males. These 'winter' or 'resting' eggs have a thick shell and can persist in the sediment for many years until conditions are suitable for their development. These eggs are resistant to freezing and desiccation, and are easily transportable.

Cladocera are represented in lake sediments by a variety of body parts. Cladoceran species can be identified by differentiating between the morphology of headshields, shells, post-abdomens, claws, and ephippia (Scourfield & Harding, 1958), which accumulate in the sediment. Cladocera remains recovered from lake sediments are primarily found to be derived from the littoral-benthic dwelling family Chydoridae (Nilssen & Sandøy, 1990). The planktonic species are usually restricted to *Bosmina* spp. Sedimentary records of Cladocera have previously been used to show the development from oligotrophy to eutrophy in lakes (Nilssen & Sandøy, 1990). Clear changes in the species composition of the Cladoceran assemblages, where there is distinct replacement of species within the Chydoridae and the genus *Bosmina*, can be related to

changes in fish predation pressure and lake trophy (e.g. Kerfoot, 1974; Boucherle & Züllig, 1983).

The direct response of the cladocera to acidification is more problematic. Interpreting changes in species assemblages and abundances over small changes in pH is complex, and detailed knowledge of cladoceran autecology is poorly understood below pH 5.5 (Nilssen and Sandøy, 1990). Changes in species composition often related to acidification could reflect indirect effects, resulting from changes in predation pressure and vegetation type and cover that are brought about via changes in pH.

A number of palaeolimnological studies have, however, demonstrated changes in cladoceran assemblages related to acidification (Steinberg et al., 1988; Uimonen-Simola & Tolonen, 1987; Nilssen & Sandøy, 1990). Increasing acidity has been demonstrated to effect changes in community interactions, the loss of acid sensitive species and of species richness as a whole, and changes in total individual numbers of cladocera (Paterson, 1994).

2.4.5 Other fossil groups found in lake sediments

The fossil organisms described above are not the only biological indicators that are preserved in the sediment record of lakes, although they are the most widely studied groups in acid lakes. Lakes support a rich and diverse flora of living organisms and many of these are represented in the sediment record.

In well-buffered systems the calcite, bivalve shells of the group ostracoda are preserved. These shells have been used to infer changes in lake water O₂, salinity and other climate related variables (DeDecker et al. 1988). In low alkaline systems mandibles from the genus *Chaoborus* have been used to effectively infer changes in fish predation pressure, and presence and absence of fish populations (Uutala 1990 & Uutala et al. 1994). A number of other algal groups are preserved in the sediment as non-siliceous fossils. These include the *Pediastrum* (Chlorococcales) group, which are the best preserved of the other algal groups (Charles et al., 1994). In suitable and very rare conditions, however, green, blue-green and dinoflagellate vegetative cells and resting spores can also be found (Livingston, 1980). The reproductive structures of charophytes (oospores) are also preserved. Biochemical fossils such as photosynthetic pigments are also being investigated to attempt to trace past changes in algal and bacterial populations that do not leave reliable morphological fossils (Leavitt et al. 1989).

Little work has been done, however, to assess the degree to which many of these fossil organisms quantitatively and qualitatively represent the structure and productivity of the ecological community from which they were derived (Battarbee, 1991). Lack of representation stems from problems in the preservation of the organism post mortem (taphonomy) and because whole biotic groups appear to be missing or greatly underrepresented in the sediment record (op cit.).

Chapter 3: Concepts of Ecological Restoration

From an ecological standpoint restoration of aquatic ecosystems should represent the restoration of biological activity: achieving working ecosystems in which macrophytes, zooplankton, plankton and other aquatic fauna are functioning within their normal range of activity.

Freshwaters are perturbed by two different sets of impacts (Battarbee, 1997); contamination of the surface waters from a diverse range of pollutants and habitat disturbance such as physical alterations to the shoreline or catchment (e.g. hydroelectric dam construction). How pollution and habitat disturbance interact to determine the nature of environmental change in freshwaters needs to be considered if the restoration of freshwaters is to be attained. Battarbee (1997) illustrates the relationship between pollution effects and habitat disturbance in the form of a naturalness matrix (see Figure 2)

Ecosystems can be related to each other in terms of their structure and their function. The number of species and the organisational complexity of the ecosystem define ecosystem structure. Ecosystem function is a combination of the biomass and nutrient content of that ecosystem (Bradshaw, 1984). Degradation of an ecosystem will result in the reduction of ecosystem function or structure or both. Bradshaw (op cit.) represents this in the form of a diagram (Figure 3). Natural ecosystem processes will move a degraded ecosystem along a theoretical pathway back to its original state. This process will take a long time to complete and can only take place if the forcing that led to the degradation has been removed. If this has not been removed further degradation of the ecosystem may take place. Moving the ecosystem along the pathway to the original state artificially is the act of restoration. Bradshaw (op cit., & 1996) proposes a number of options for where restoration may not be possible; the original state of the ecosystem may be the result of human land use practices that are now outdated and no longer practised, for example. These options are rehabilitation and replacement. Rehabilitation is progress made towards the original state that is not complete. Ecosystem function and structure have been improved, but the pre-disturbance conditions have not yet been achieved. Alternatively, another ecosystem can be substituted for the degraded one. The substitute ecosystem is generally less complex than the original state of the degraded one. This process is known as replacement. True restoration may be unrealistic in many situations. It may be expensive or inappropriate. In these circumstances, rehabilitation or replacement may be better suited to providing an ecosystem more valuable in function and complexity than the degraded ecosystem.

Bradshaw (1996) has also proposed that by aiming at ecosystem restoration we are setting too high a target for ourselves. In current ecological terminology, the word 'ecosystem' describes both the biological and the non-biological elements that occur together in a given area. Restoration of an 'ecosystem' then should revolve around restoring the function, structure, and the interaction of the whole system. Consequently, it is now common to talk about 'habitat' restoration where the emphasis is placed upon restoring the 'place' where organisms live (the habitat) rather than the processes of a degraded ecosystem.

Good ecological restoration entails negotiating the best possible outcome for a specific site based on ecological knowledge and the diverse perspectives of interested stakeholders (Higgs, 1997). It should be noted then, that simply restoring a disturbed ecosystem to its former state

Figure 2: Naturalness matrix showing: (1) hypothetical change over baseline for three lakes (a, b, & c): (2) a 'state-changed' classification system related to change over baseline, with 1=most changed, 9=least changed and 10=pristine (from Battarbee, 1997).

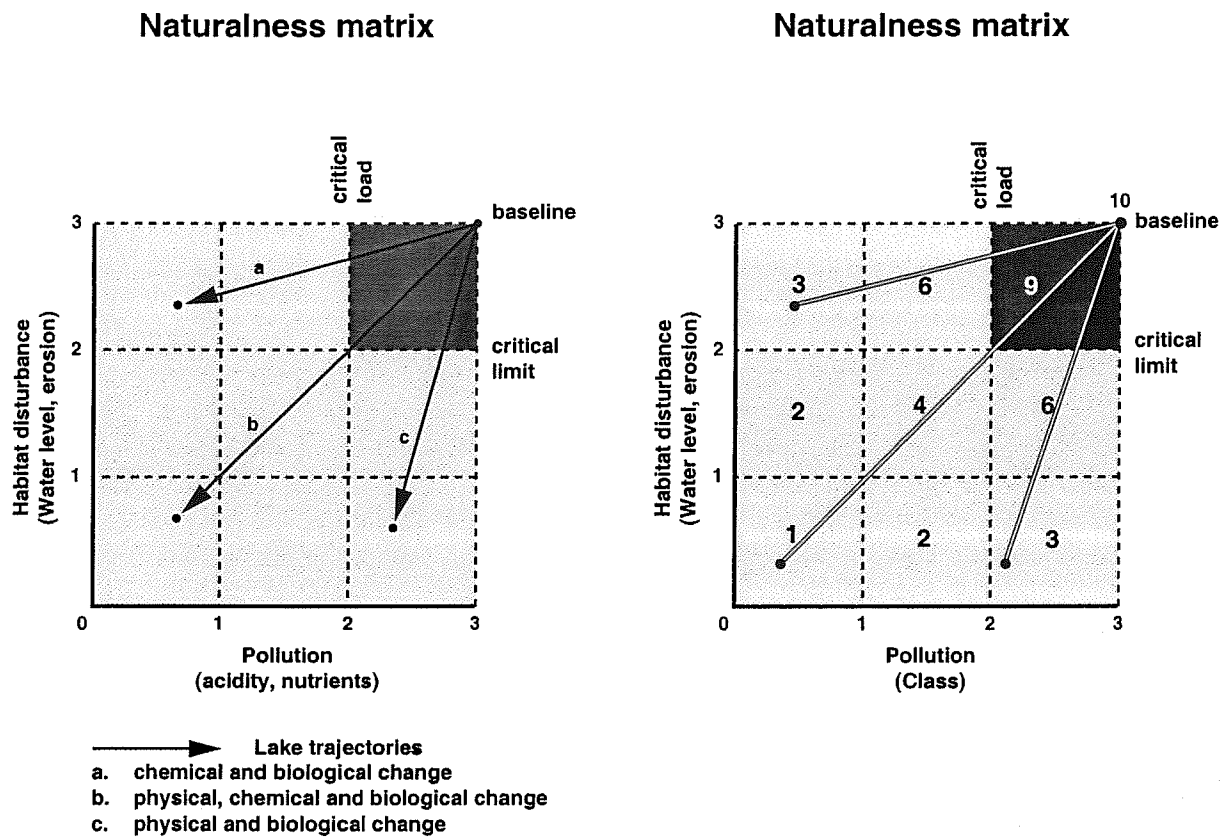
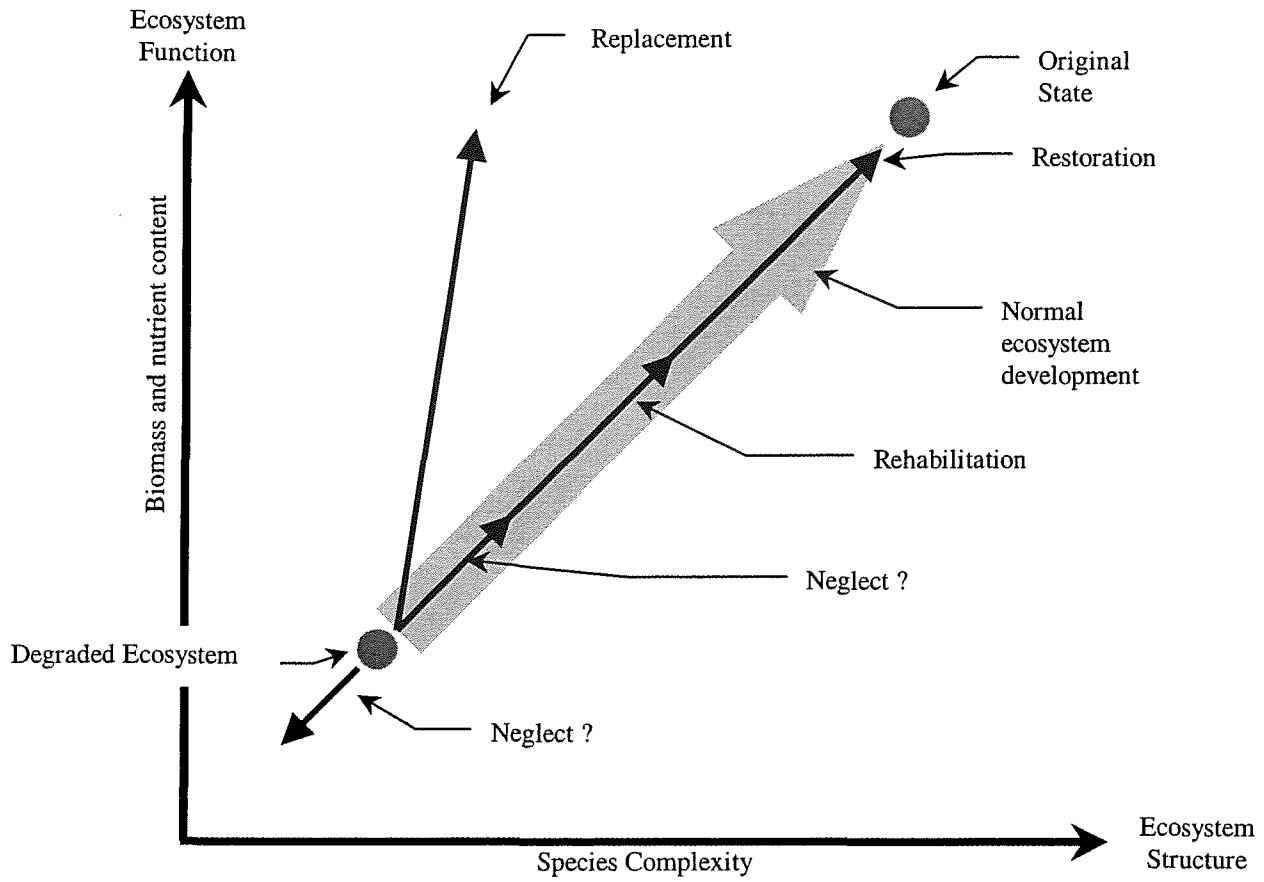


Figure 3: Changes in ecosystem function and structure for a hypothetical degraded ecosystem. Note the various forms of 'restoration' and the changes associated with true restoration (after Bradshaw, 1984).



does not always form the most appropriate use of the available funds or the most appropriate restored community.

3.1 The use of baseline conditions as restoration targets or reference states

Most surface waters in Scotland, Wales and Northern England have existed for around 10 000 years. They were formed as glaciers retreated when the climate began to warm at the end of the last glacial. During this period natural climatological, geological, geomorphological and biological change has taken place. More recently, anthropogenic changes have begun to affect UK surface waters, most notably the eutrophication and acidification of lakes and rivers. Catchment processes or the hydrochemistry of certain lakes may have buffered some or all of the effects of change. Other, more sensitive, surface waters may have been altered by the natural and anthropogenic changes they have been exposed to.

The potential influence of natural factors has clear implications when we consider defining the baseline state of degraded (acidified) surface waters. The baseline state of the surface water could be defined as the state prior to the onset of acidification, or alternatively as the state immediately following the retreat of the glaciers and the lake formed. The potential effect of climate change on lake water acidity is discussed in more detail below. Such natural responses complicate the process of defining baseline state for acidified lakes, let alone describing the hydrochemical and biological status of such a state.

Bradshaw (1984) used the term 'original state' to define the pre-impact state of an ecosystem. In palaeolimnology, this state can be described as the 'Baseline State' of surface waters. The baseline or pristine state of a surface water is that which combines an absence of habitat disturbance and an absence of pollution. It can be argued that this represents the highest quality of surface water that is attainable in the UK, and that these conditions should be the main goal of any restoration project.

The baseline can also be thought of as the state prior to a specific impact. In the UK, evidence of acidification is rarely found before the 1850s. This period can be defined as the baseline for acidification but does not necessarily reflect the state where there has been no anthropogenic influence on the lake. In this way baseline conditions can be used to define future hydrochemical and biological reference conditions, or if unobtainable, due to irreversible change, alternative targets that approximate natural conditions can be defined (Battarbee, 1997). In such cases, some of the most significant baseline biological characteristics can be defined from the sediment record contained within the lake.

3.2 Hydrochemical and Biological Targets for Restoration

Studies of disturbance in lakes often revolve around the decline or accumulation of a certain chemical variable or suite of associated variables in the lake water (for example, total phosphorus (TP) (Bennion, 1994), pH (Flower & Battarbee, 1983) and Al and DOC (Kingston & Birks, 1990)). Targets could be set with reference to certain hydrochemical or biological criteria. Reconstructing the hydrochemical characteristics of surface waters has often been achieved through the use of indicator species (e.g. Birks et al., 1990b); the hydrological change being defined by the response of a particular group of organisms to that change. Other studies (e.g. Henriksen et al., 1990) have used point source measurements to determine the degree of

change between studies in surface water hydrochemistry, or to identify those surface waters that are sensitive to environmental change.

Consequently, because of this need to quantify biological change in response to environmental change for hydrochemical reconstruction, the biological response to acidification is fairly well understood (e.g. Havas & Rosseland, 1995). The effects of lake acidification have long been identified, with the major loss of fish populations and aquatic vegetation within acidified systems being the most immediately identifiable of responses of the system to decreasing pH (Uutala, 1990; Kingston et al., 1992; Havas & Rosseland, 1995). Other studies have looked at the effects on algal populations, such as diatoms or chrysophytes (Battarbee, 1984), whilst other authors have described the change in insect (Brodin & Gransberg, 1993) or zooplankton (Havas & Rosseland, 1995) populations associated with acidification.

The return of fish stocks to lakes is an easily identifiable, ecological (biological) target for scientists, politicians and the environmentally aware public to focus upon, as well as being an issue of great economic importance. The reliance upon this sort of target, and the use of solely hydrochemical parameters to define it, has slowed progress towards adopting relevant biological reference states that address the whole lake system rather than a small part of a lake's ecosystem. Henriksen et al. (1990) sampled many lakes across Norway for the 1986 1000-lakes survey. The project was designed to determine the surface water chemistry and the status of fish populations of lakes from acid sensitive areas of Norway. The data was used to identify changes in water chemistry and fish status since 1974 and 1975 (c.f. Henriksen et al., 1990) and to act as baseline characteristics against which the effects of emission's reductions (e.g. LRTAP protocols) could be judged. The only biological aspect incorporated in the 1000-lakes survey was the fish status of the surveyed lakes. It was only later, through further study of the other biological groups found in aquatic systems, that the degree to which other organisms were affected by acidification was identified (Havas & Rosseland, 1995).

3.3 Liming: an example of lake acidification restoration

A key technique employed by those attempting to restore acidified surface waters has been to lime either the water directly or the catchment that drains into the acidified lake (Renberg & Hultberg, 1992). Although the impetus behind such restoration methods has been a biological one it is still essentially a hydrochemical approach in nature. The aim of liming is an elevation of the pH of the water (and the increase in acid neutralising capacity (ANC) and the decrease in Al toxicity that go with it) that will allow fish populations to be re-introduced or re-colonise.

Each year approximately 300 000 tons of powdered limestone is used to treat lakes, rivers and wetlands in order to increase the pH of surface waters across the globe (Henriksen et al., 1995). This costs between 40-50 million US\$ per year (Henriksen et al., 1995). There are over 11 000 lakes that are routinely limed in Sweden and Norway alone.

Originally, the main aim of liming was to restore fish stocks to impacted lakes for economic and recreational fishing (Henriksen et al., 1995). These aims have now been broadened to encompass the preservation and recovery of biodiversity and human health. The official aim of the liming projects in Sweden (Svensson et al., 1995) and Norway (Romundstad & Sandøy, 1995) are shown in Table 3.

In the UK there has been far less use of liming to mitigate the impacts of acid deposition (c.f. Howells, 1995). At Loch Fleet, Southwest Scotland, an extensive study of liming applications and the biological and chemical responses to the treatment was begun in 1986. Around 450 tonnes of CaCO₃ was applied to various parts of the catchment, and the water quality of the loch quickly improved following the treatment. pH rose to 6-7, calcium to 4 mg l⁻¹ and labile Al was greatly reduced (Howells, 1995).

Table 3: The primary aims of liming of surface waters in Sweden and Norway (Source: Muniz, 1991).

Sweden	The biological aims are to detoxicate the water for the preservation or recolonisation of the natural flora and fauna
	The chemical aims are to raise the pH above 6.0 and alkalinity to levels exceeding 0.1 meq l ⁻¹
Norway	To improve the conditions for recreational fishing
	To preserve biological diversity

The water quality proved suitable for the return of trout through restocking, and 18 months after initial treatment the restocked fish have shown good growth and fecundity (op cit.). There have also been changes in the invertebrate fauna of Loch Fleet, a decline in predatory beetles (thought to be due to fish predation) and the return of some acid-sensitive species. Flower et al. (1990) conducted a palaeolimnological study of Loch Fleet using diatoms to determine the biological response of this important group to the liming treatment. They concluded that the pH of the lake had risen to a level that had never been reached throughout the history of the lake (Flower et al., 1990; Anderson et al., 1986). The diatom taxa present after liming contain species that were previously rare or absent from the diatom sedimentary record.

This demonstrates the crux of the problem with liming as a restoration tool. Far from restoring the pre-acidification flora and fauna of acidified lakes the results show that conditions created in lakes after liming are often different to any condition previously seen in the lakes through the palaeolimnological record. This cannot be considered restoration in the sense of Bradshaw, rather it is replacement. In essence a degraded ecosystem is being replaced by one that restores a target organism to the treated lake but which does not reflect the original state of the lake in terms of structure or function. This may be an appropriate target for which to aim if the biological target is the restoration of fish stocks to impacted lakes. However, it should not be taken to represent the original state of the degraded ecosystem. For true restoration to be achieved the biology of the original state of the degraded ecosystem must be restored. Identifying such targets or reference states, therefore, should involve biological as well as

hydrochemical variables and should be related to conditions that are similar to those previously exhibited during the history of any given lake. Palaeolimnology can define such targets or reference states in terms of the hydrochemistry and selected biological indicators.

3.4 Defining baseline states and restoration targets

To define a baseline or reference state for a surface water hydrochemical reconstructions have traditionally been used to provide information about the pre-disturbance hydrochemistry of surface waters. There are two potential ways that hydrochemical reconstructions can be made; dynamic modelling and transfer functions. Dynamic modelling uses information about the hydrological and chemical influxes to a catchment and other variables such as soil CO₂ partial pressures, soil porosity, bulk density, and cation exchange capacity, to model hydrochemical changes for given deposition patterns. One such dynamic model is MAGIC (Model of Acidification of Groundwaters in Catchments) (Cosby et al., 1985a,b). Using known historical deposition levels (usually only for the last decade or so) the models can be used to hindcast the historical hydrochemistry of surface waters. However, most often the models are used to predict future hydrochemical targets for given levels of emission abatement such as the LRTAP conventions discussed above. The other main way in which hydrochemical reconstructions are made is using palaeolimnological techniques and hydrochemical transfer function. As discussed above, certain biological indicators (usually the siliceous algae, diatoms and chrysophytes) have strong responses to hydrochemical variables, primarily pH, phosphorus and salinity. These can be quantified, and using weighted averaging calibration, used to infer the historical hydrochemistry of surface waters from the sediment record. This then acts as a record of the hydrochemistry of a surface water. Then a pre-disturbance period can then be identified and the hydrochemistry inferred from the sub-fossil remains of the sediment record.

The use of the palaeolimnological record to set biological targets for restoration is considered in the next chapter.

Chapter 4: Setting Biological Targets for Ecosystem Restoration Using Palaeolimnology and the Modern Analogue Approach

One approach to defining biological targets or reference states using palaeolimnological data is the modern analogue approach. The modern analogue approach uses robust statistical methods to compare fossil assemblages to modern assemblages. This approach was developed by Flower et al. (1997).

4.1 Modern Analogues

Modern Analogues are lakes that have modern chemistries and biological communities that match the past conditions of a lake. Defining analogue sites can provide the chance to examine the ecology of selected lakes so that the biological characteristics of the analogue lakes can be used to define biological targets or reference states for recovery from acidification. Space-for-time substitution is the basis of selecting sites as modern analogues. This assumes, however, that the aquatic communities currently existing in sites closely resemble those that existed in similar but now acidified habitats (Flower et al., 1997). The sites and the biological communities they contain can vary greatly through subtle differences in geology, climate or other biogeographic and chemical variables.

The methodology behind modern analogues has its foundations in the field of palynology (Overpeck et al., 1985). Fossil pollen spectra derived from palaeoecological studies have been compared to the pollen spectra retrieved from pollen traps and taken from modern sites. This has allowed palynologists to match modern forest ecosystems to those that existed in the past (e.g. Wright, 1967). From this, they could interpret the type of ecosystem and vegetation that once existed and evaluate the changes that have brought the ecosystem to that which is found today. Early studies used qualitative matches of the various pollen spectra. Modern studies use statistical methods to quantify the degree to which modern sites match the fossil flora (c.f. Overpeck et al., 1985). These statistical methods are known as dissimilarity coefficients.

Dissimilarity coefficients measure the difference between multivariate (e.g. species abundances) samples such as those produced in palaeolimnological studies. Dissimilarity coefficients have a number of advantages:

1. They are a precise method of comparison between samples;
2. The process may be automated or computerised;
3. Dissimilarity Coefficients allow the opportunity to calibrate the dissimilarity scale in terms of the underlying biological or environmental differences.

Other, multivariate, approaches also exist (e.g. using principle components analysis or canonical variates analysis) but these do not directly compare the degree of analogy that dissimilarity coefficients measure.

Prentice (1980) suggests that there are three types of dissimilarity coefficient, the simple or unweighted, the equal weight, and the signal-to-noise measures. Simple methods tend to be heavily influenced by taxa that have wide ranges, and the rare types have little influence upon the results. Equal weight measures upweight the rare taxa and downweight the common species. A potential problem is that these tests may give extra weight to the insignificant, yet

potentially noisy, taxa. The method of upweighting is dependent upon the statistical test being used. Signal-to-noise measures avoid this by weighting the data so that the signal component of the difference between fossil and modern samples is emphasised at the expense of the noise component. The problem with signal-to-noise measures is that they are scale dependent (Webb et al., 1978; Delcourt et al., 1984). Equal weight measures may prove more appropriate if certain minor taxa are judged important to the analysis. Overpeck et al. (1985) tested various dissimilarity coefficients for application in analogue matching tests. They found that the signal-to-noise methods provided the more robust analysis because rare taxa are down-weighted and the floristic signal drawn out. Their results also showed that the three signal-to-noise measures they tested all provided similar results.

There has been very limited use of modern analogues, especially in any quantitative way, within the field of palaeolimnology to tackle the issues of recovery in acidified lake systems.

An exception is the work of Flower et al. (1997), who identified modern analogues for two Scottish lochs from a modern dataset of lakes from northern Europe. Flower et al. (1997) used the squared chi-square distance measure in their study of diatom based modern analogues for the pre-acidification status of recently acidified lakes in northern Europe. They compared the fossil diatom assemblages in two acidified lakes from the Galloway region of Scotland (the Round Loch of Glenhead and Loch Dee) with modern surface sediment samples from over 200 lakes in Britain, Ireland, Sweden and Norway. The squared chi-square analysis identified several modern analogue sites within the dataset. Loch Teanga in the Hebrides and Lough Claggan, Ireland had the most similar diatom floras to the pre-acidification floras of the Round Loch of Glenhead and Loch Dee respectively. These sites, however, can not be considered as being pristine sites (i.e. sites that have not been impacted by atmospheric deposition). Loch Teanga and Lough Claggan are located in areas of low to moderate acid deposition ($0.4-0.8 \text{ g S m}^2 \text{ yr}^{-1}$) (Flower et al., 1997; Battarbee et al., 1988; Flower et al., 1994). Although the diatom communities of these lakes do not show any response to this level of deposition other biological communities may have been affected by deposition or trace metal contamination. Flower et al. (1997) also looked at the other hydrochemical determinants of the lakes selected as analogue matches for the Round Loch of Glenhead and Loch Dee. The Round Loch of Glenhead had no modern analogues that had a similar lake-water calcium concentration. Loch Dee, however, had some analogues with similar calcium concentrations (e.g. Loch Howie, Scotland, and Lough Brockagh, Donegal) and these sites may represent better analogue sites than Lough Claggan.

The approach has since been applied to other Acid Waters Monitoring Network Sites (Allott, pers. comm.).

With the adoption of robust statistical techniques, the application of modern analogues to identifying potential targets for the restoration of acidified surface waters has become a potentially powerful tool.

The approach used by Flower et al. (1997) raised a couple of interesting points regarding the suitability of analogue sites selected in this manner. These will be discussed further in the following section.

Chapter 5: Key Assumptions and Problems of the Modern Analogue Approach

The modern analogue approach makes a number of assumptions about the data used and the theoretical concepts of analogue matching (c.f. Flower et al., 1997). These assumptions need to be tested before analogue matching techniques are applied to the study of recovery, as they are inherent in understanding the validity of substituting space for time in biological studies.

The key assumptions used in the modern analogue approach are:

1. The fossil group (or groups) used for analogue matching effectively represents ecosystem variation.
2. The modern surface sediment dataset contains suitable analogue lakes for the acidified lakes that will be matched with them (e.g. that there are enough pristine, high calcium concentration sites to provide an adequate range of matches).
3. The hydrochemistry and biological communities of analogue sites accurately represent the pre-acidification conditions of acidified lakes.

5.1 Do diatoms effectively represent ecosystem variation?

It has to be assumed that the fossil group used in analogue matching (e.g. diatoms in the Flower et al., (1997) approach) is representative of the whole trophic cascade and variation in other biological groups found in aquatic systems (e.g. other algal groups, aquatic macrophytes, other invertebrate groups, or fish). Furthermore, the variation in the fossil record is assumed to accurately reflect changes occurring to the whole biology of a given lake. This has to be assumed because not all groups leave remains that are preserved in the sediment record of lakes (see Palaeolimnology section earlier). Therefore, analysing change in other biotic groups is impossible without these inferences unless historical records are available.

It is well known that diatoms respond quickly and with definite pattern to changing lake-water acidity. Given this information, it is likely that a matching process based solely on sub-fossil diatom data is selecting analogues with similar pH characteristics to those inferred from the surface sediments of presently acidified lakes rather than on the whole hydrochemical signature. It is likely, therefore, to expect that the results do not necessarily reflect whole ecosystem characteristics.

5.2 Does the modern data set contain a large enough range of sites from a range of hydrochemical characteristics?

In the Flower et al. (1997) application, the lakes selected as the closest analogues to the Round Loch of Glenhead and Loch Dee were lakes that had, somewhat, been impacted by acid deposition. They were from areas receiving low to moderate acid deposition. Flower et al. (1997) suggest that this was due to there being insufficient pristine sites in their dataset. Another problem they encountered was that the two selected analogue lakes had very different hydrochemistries apart from their pH levels (as inferred from the diatom data). Consequently, it is important to assume that the dataset being used for analogue matching has sufficient geographical range and hydrochemical scope for the matching process to be worthwhile.

Flower et al. (1997) recognise that there is a need to incorporate surface sediment data from more pristine sites. In their dataset, there were very few pristine sites, the majority found in northern Norway. Including more pristine sites would improve the chances of pristine lakes being selected as modern analogues. This is problematic because there are very few areas of Northern Europe that can be considered pristine as there are large areas of northern Europe that have been impacted somewhat by acid deposition.

5.3 Can baseline conditions be adequately defined?

For the process of space-for-time substitution to be accepted as an adequate model, it must be assumed that the hydrochemistry and biology of analogue lakes are a true reflection of pre-acidification conditions observed in acid lakes. This assumes that anthropogenic acidification is the only or the major forcing mechanism affecting environmental change upon acidified lakes in the UK over the Holocene period. This also assumes that climate change or other anthropogenic influence (e.g. habitat disturbance) since pre-acidification times (c. AD 1850) has had negligible effects upon the overall hydrochemical and the biological functioning of the lake systems.

There is some degree of evidence that climate changes and other anthropogenic influences (e.g. catchment disturbance or land-use change) have had profound impacts on the hydrochemistry and biology of some studied lakes across northern Europe.

5.4 Natural variability in surface water systems

Nature is inherently dynamic: A constantly changing or fluctuating climate has been a major driving force in determining the present day distribution of ecosystems and the physical appearance of the landscape that they occupy. Among this background of climate change and natural variability anthropogenic activities have influenced and changed ecosystems, predominately since the industrial revolution, but also during the forest clearances of the Neolithic period and later clearances as land was prepared for agricultural use (Roberts, 1998).

An important consideration in the ecological restoration of acidified sites is the degree to which the pre-acidification hydrochemical and ecological conditions may have changed. There are three main themes that need addressing when considering the fluctuating hydrochemical and ecological conditions of surface waters, the climate effect, catchment disturbance and land-use change effects, and the apparent stability of UK lakes prior to anthropogenic acidification.

5.4.1 Climate

For mountain lakes, Skjelkvåle & Wright (1998) suggest that palaeolimnological analogues may be of little use because future climate changes are likely to cause 'conditions never previously experienced on earth, such as high atmospheric CO₂ levels and high UV-B radiation.' (Ibid. pp-285). A number of other studies also describe the likely effects of a changing climate for acid lakes (e.g. Schindler et al., 1996; Psenner & Schmidt, 1992; Leavitt et al., 1997). These effects are summarised in Table 4. Such dynamism would compromise the applicability of the modern analogue approach and question whether targets for recovery from anthropogenic acidification are realistic goals.

To assess the impact or influence that fluctuations in climate have had upon lakes, long core stratigraphies that contain past records of climate fluctuation are required. Analysis of the cores for changes in biological assemblages over time and dating of these changes can answer questions surrounding whether upland acid waters were chemically and ecologically stable prior to anthropogenic acidification or are continually changing, dynamic systems.

Psenner & Schmidt (1992) have demonstrated a relationship between colder air temperatures and lower pH of surface waters in two soft-water, high-altitude lakes in the central Alps. The inferred pH data from diatom analysis and correlation with the temperature record was also supported by the results of loss-on-ignition (LOI) and Fe/Mn ratio analysis of the lake sediment. LOI and Fe/Mn are surrogates for the organic content of the sediment. Psenner & Schmidt (1992) found three distinct peaks in the LOI and Fe/Mn data that correspond roughly to the temperature peaks of 1820, 1860, and 1900. Prior to the onset of anthropogenic acidification in the late 19th and early 20th centuries there was a distinct coupling between climate and biogeochemical processes in the two alpine lakes. This coupling has progressively broken down over the last 100-150 years as the deposition of strong acids to surface waters has had greater influence on the biogeochemical processes of the lakes than the fluctuating climate. This has clear implications for assessment of recovery in systems that have climate/biogeochemical coupling. Psenner & Schmidt (1992) suggest that recovery observed in some systems, by a levelling off of the inferred pH decline, may be due to rising temperatures and global warming as much as that due to decreasing acid deposition.

5.4.2 Catchment disturbance and land-use change

For Swedish lakes, Renberg (1990) and co-workers (Renberg et al., 1993a,b) have produced long-core stratigraphies and have analysed the diatom profiles of a great number of lakes. The results show a number of features characteristic of diatom-inferred pH profiles from some of the acidified lakes in Northern and Southern Sweden. Swedish lakes are generally base rich, well buffered and mesotrophic immediately following the end of glaciation. Following this a period of slow natural acidification takes place where progressive leaching and loss of base cations from the soils leads to soil acidification and dilution of the Acid Neutralising Capacity (ANC) of run-off entering the lakes (Renberg, 1993b). The pH of the Swedish lakes immediately following retreat of the ice sheets has been shown to be around 7.0 and the slow process of natural acidification increased acidity to c. pH 5.5.

Acidification continued up until ca. 2300 BP when suddenly, and across a geographic range, the pH of lakes rose quickly to c. 6.5. This has been attributed to a shift in land-use in the catchments of many Swedish lakes known to have taken place around this period of history because of an expansion in the agrarian culture of the Iron Age. Increases in pH have been correlated with the recession of natural forest, expansion of shrub vegetation, increased frequency of cereals and weeds, increased concentrations of charcoal and LOI values, indicating increased soil erosion (Renberg, 1993b). Cultural alkalinisation continued until the industrialisation of the 1900s. Many lakes in Sweden have acidified considerably since the 1950s; many lakes becoming severely acidified with permanently reduced pH values of between 4.5 and 5.0.

Table 4: Possible effects on lake-water chemistry from different possible effects of Climate (Source: Skjelkvåle and Wright, 1998)

Environmental driving variable	Primary Effect	Secondary Effect	Biological impact	Type of Study	References
Climatic warming	Increased water temperature reduced water turnover	higher concentrations of solutes	decreased summer habitat for cold stenothermic organisms	empirical data, 20-year natural climatic cycle (Canada)	Schindler et al., 1990
Climatic warming	Increased water temperature		changes in fish yield	modelling study (Canada)	Minns & Moore, 1992
Climatic warming	Increased water temperature	increased alkalinity generation	changes in diatom communities	palaeolimnological study (Austria)	Psenner & Schmidt, 1992
Stratospheric ozone depletion	Increased UV-B		reduction in photosynthesis and growth of diatoms	mesocosom experiment	Bothwell et al., 1994
Climate warming and acid deposition	Increased mineralization of soil organic matter	increased NO ₃ and acidification		large-scale experiment CLIMEX (Norway) empirical data (Norway)	Lükewille and Wright, 1997, & Wright, 1998 Lydersen, 1995
Climate warming and acid deposition	Decreased DOC concentrations	increased light penetration (UV-B)	photoinhibition of phytoplankton	empirical data (Norway)	Schindler, Bayley & Parker, 1996, & Schindler et al., 1996
More strong winds (Storms)	More 'sea-salt episodes' in lakes in coastal areas	'acidic episodes' in acidified areas	damage to aquatic biota and fish kills	empirical data (Norway)	Hindar et al., 1994 Hindar et al., 1995

Renberg et al. (1993a) also identify a recent liming period in many of the acidified Swedish lakes. Liming has been adopted as a widespread restorative measure in over 6000 lakes (Renberg, 1993a). Liming characteristically increases pH to levels above those of the immediate post-glacial, and the resultant diatom flora is quite unlike anything that has been found in the post-glacial history of the Swedish lakes.

From this type of study, it is clear that considerable anthropogenic influence on Swedish lakes has been experienced since around 2300 BP. This has implications for any attempt to restore these acidified lakes to a former state. The problem now facing environmental managers is that if they are to restore lakes to pre-acidification states then they will be attempting to return lakes to conditions that were inherently dependant upon the cultural changes in land-use associated with the expansion of agriculture in the Iron Age. This type of land-use no longer exists in Sweden so there is little hope of restoring lakes to such a status. Renberg et al. (1993a) have extrapolated the predicted pH response to continued natural acidification given that none of the changes in land-use or deposition chemistry of the last 2300 years had taken place. Arguably, this provides a suitable target that could be attainable given pollution abatement strategies already in place or being negotiated. This is a theoretical attempt to express the expected trajectory following the overriding trend in natural acidification over the previous 10 000 years following deglaciation. However, this provides no more a representative target than the pre-acidification state of surface waters and there is no guarantee that this is a more attainable target given possible changes in the hydrochemical, physical, biological and climatological variables since 2300 BP.

5.4.3 Pre-acidification chemical and biological variation in UK lakes

Atkinson and Haworth (1990) provide a similar study this time of two sites in the UK, Devoke Water in the Lake District, Cumbria, and Loch Sionascaig, Northwest Scotland. Similar trends of slow natural acidification to those found in Swedish lakes were found following deglaciation. However, both lakes are not as sensitive to acid deposition as other upland sites in the UK and the Swedish lakes, and have suffered much less disturbance. Devoke Water has recently (post 1850) begun to acidify to a point where there are considerable biological changes within the waterbody. Loch Sionascaig, on the other hand, is a more well-buffered site and has a stable inferred pH profile for the past few thousand years. In neither of these two sites was a cultural alkalisiation period identified. This suggests that upland UK systems are relatively stable during the Holocene compared to their Swedish counterparts. Further studies also demonstrate the apparent stability of UK upland surface waters prior to the onset of anthropogenic acidification. Jones et al. (1986) presented a diatom-based reconstruction of pH for a Holocene sediment core from the Round Loch of Glenhead, Galloway, Scotland. The diatom stratigraphy of that core showed little change other than that associated with the acidification of the lake shortly after the end of the last glacial, and the recent, anthropogenic acidification. Birks (1996) also demonstrates stability in the Round Loch of Glenhead using rate-of-change analysis to assess the degree of variability in diatom species throughout a Holocene sequence. The only significant period of change was found to be in the last 200 years that is associated with the anthropogenic acidification of the loch (Allott et al., 1992), and the immediate post-glacial as base cation rich soils began to leach, thus influencing the ionic composition of run-off.

Given the relative stability of UK upland acidified sites it should be reasonable to assume that the pre-acidification status of a given lake is an attainable target for restoration measures to

attempt to recreate. However, the apparent stability of UK upland surface waters may be misleading. Only a few sites have been studied in the UK and the resolution of the studies has been biased towards detecting acidification in the upper layers of the sediment record. There has been little high-resolution analysis of pre-1800AD sequences.

Chapter 6: Summary and Recommendations

Palaeolimnological techniques have been widely employed to study lake acidification. This approach has been central in testing the cause-effect relationship between acid deposition and lake acidification, and in assessing the magnitude and extent of surface water acidification across the UK.

Most of these palaeolimnological applications have been based on diatom analysis, and the use of diatom-pH transfer functions to make reconstructions of hydrochemical change in upland lakes associated with acidification.

Following the signing of the Second Sulphur Protocol, attention is now focusing on emissions reductions and the reversibility of surface waters acidification. There is a clear need for criteria against which to evaluate the recovery process.

In order to evaluate future recovery, Flower *et al.* (1997) have proposed a palaeolimnological technique for defining targets for the recovery of acidified surface waters. This is based on the technique of analogue matching of lake sediment diatom assemblages. Multivariate statistical methods are used to identify modern analogues for the pre-acidification diatom assemblages of acidified lakes. The chemical and biological status of modern analogue lakes can then potentially provide recovery targets for acidified systems.

This approach has been successfully applied to several acidified lakes, and modern analogue systems defined for the pre-impact (pre-acidification) status of these impacted sites. An advantage of the approach is that it can provide recovery targets for both chemical and biological status of acidified lakes.

Modern analogue matching as currently applied makes several key assumptions:

1. that analogue matches based on a single biological group (diatoms) effectively represent the hydrochemical and biological variation of low alkalinity systems;
2. that the modern data set used to identify modern analogues contains the range of hydrochemical conditions represented by the fossil assemblages;
3. that a suitable stable 'baseline' (pre-impact) status can be defined.

Prior to more comprehensive application of the modern analogue approach to acidified lakes in Britain, these assumptions require evaluation. Three studies are proposed:

1. Extension of the current modern lake dataset used for analogue matching by the inclusion of minimally impacted low alkalinity sites from northern Scotland.
2. Development of the current technique by including two more fossil groups (chironomids and cladocera) in the modern surface sediment dataset used in the matching procedure. This will allow the assumption that diatoms represent wider ecosystem variation to be tested, and should result in more robust analogue matches.

3. A study of hydrochemical and biological variation in the pre-acidification conditions of acidified lakes through high-resolution palaeolimnological study of selected Acid Waters Monitoring Network lakes. This will allow the stability of baseline (pre-acidification) conditions to be evaluated.

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