
Diatom Analysis and the Acidification of Lakes [and Discussion]

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Diatom analysis and the acidification of lakes

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Diatom microfossils preserved in lake sediments can be used to provide evidence for lake acidification. Unlike documentary methods of historical reconstruction, the sediment record is potentially useful at all sites, it is usually continuous and it can be extended backwards in time as desired. Cores can be taken with little disturbance, by freezing *in situ* if necessary, and sediments deposited since about 1800 A.D. can be dated using ^{210}Pb analysis. The strong relationship between diatom occurrence and water pH allows pH to be reconstructed with a standard error of between *ca.* 0.25 and 0.5 pH unit using either an index system or multiple regression analysis.

Although there is evidence that long term acidification is a natural process for lakes in areas of resistant base-poor bedrock, diatom analyses from NW European and North American sites shows that rapid acidification has taken place within the last 150 years. The first major sign of acidification is the loss of planktonic diatoms at pH values between about 5.5 and 5.8. Acidification below pH 5.5 leads to the decline of species typical of circumneutral water, such as *Achnanthes microcephala* Kütz. *Cymbella gracilis* (Rabh.) Cleve and *Anomooneis vitrea* (Grun.) Ross, and the expansion of acidophilous taxa such as *Tabellaria flocculosa* (Roth.) Kütz. and *Eunotia veneris* (Kütz.) O. Müller. Acidobiontic species become common at pH values less than 5.5 and as the pH drops to 4.5 these taxa begin to replace acidophilous taxa in the assemblage. *Tabellaria binalis* (Ehr.) Grun. is probably the most faithful member of this group.

Diatom analysis for four acid lakes in Galloway, SW Scotland show that the beginning of acidification has varied from 1840 (Loch Enoch) to 1925 (Loch Grannoch) and that pH has declined by between *ca.* 0.5 units (Loch Dee) and *ca.* 1.2 pH units (Loch Grannoch) in these lakes. Since lakes without afforested catchments have been acidified and lakes with afforested catchments were acidified before afforestation it can be concluded that afforestation is not responsible for acidification in this region.

INTRODUCTION

Claims that acid precipitation has led to the acidification of lakes and to the loss of salmonid fish populations (Jensen & Snekvik 1972; Almer *et al.* 1974) have promoted a need for unambiguous empirical evidence of recent lake acidification. Since the process is too slow to be detected by short-term monitoring, evidence must be based either on long-term monitoring, in the expectation of continuing change, or on historical reconstruction. A number of methods have been used for historical reconstruction including the use of chemical records (Wright 1977; Watt *et al.* 1979; Sutcliffe *et al.* 1982), fisheries data (Jensen & Snekvik 1972; Beamish & Harvey 1972), algal records (Berge 1976, 1983; van Dam & Bloklund 1978; van Dam *et al.* 1981) and microfossil analysis of lake sediments (for example, Davis & Berge (1980); Renberg & Hellberg (1982); Flower & Battarbee (1983)).

Chemical records, especially pH records, are potentially the best means of reconstructing recent changes in lake acidity but records exist only for a small number of lakes and they are rarely continuous. Moreover estimates of the degree of pH change based on occasional measurements arbitrarily located in time can be misleading since pH measurements can vary

seasonally and annually, and since analytical techniques are not always compatible (cf. Howells 1983).

Recorded changes in fishery status have also been used as evidence for acidification in both Europe and North America. It is assumed that those lakes that have lost or have experienced substantial reductions in salmonid populations have been acidified since salmonid species fail to reproduce successfully in highly acid waters. Fisheries data are unlikely to yield precise information on the onset and rate of acidification for any particular lake because of the general lack of quantitative records and because of the delayed response of fish populations to acidification, but trends towards fishlessness have been observed in regions sensitive to the effects of acidification (for example, Sevaldrud *et al.* 1980; Harriman & Morrison 1984).

Algal communities respond to pH change, and differences between the floristic composition of old diatom samples and modern diatom samples can be used to assess whether acidification has taken place. If old samples are available for examination it is possible to eliminate taxonomic problems that would otherwise devalue such past–present comparisons (cf. Battarbee (1979a, 1981a)). There are other problems in using this approach. First, although probably more of these records exist than of other kinds, the algal collections are limited by chance to certain sites and dates. Second, unless detailed site descriptions are available it is often difficult to relocate the sample habitat with the precision necessary to make a confident modern comparison. And third, unless a more or less continuous series of samples are available neither the timing nor rate of change can be assessed.

However Berge (1976, 1983), van Dam & Bloklund (1978) and van Dam *et al.* (1981) have made good use of this approach and have been able to show that some lakes and ponds had become more acidic between the dates of sampling.

Lake sediments preserve a chemical and biological record of the history of a lake that can be revealed by analysis and sediment dating. As a method for assessing acidification it has several unique advantages. It is potentially useful at all sites, the record is usually continuous, and it can be extended backwards in time, if necessary, up to the age of the lake basin. Moreover if good dating control is available the timing and rate of change can be assessed. Disadvantages include differential preservation of organisms both within and between species groups, problems associated with the origin of various fractions of the sediment and the lack of ecological information for some taxa and groups of taxa.

Although there is a literature on geochemical methods (Norton & Hess 1980; Davis *et al.* 1983; Tolonen & Jaakkola 1983), on invertebrate analysis (Brakke 1980; Henrikson *et al.* 1982; Davis *et al.* 1983) and on chrysophyte scale analysis (Smol *et al.* 1984), the most widely used palaeolimnological technique for indicating acidification history is diatom analysis. This paper reviews progress so far in the use of diatoms to reconstruct past pH values.

METHODS

Methods needed to carry out diatom analysis include sediment coring, sediment dating and diatom preparation. Specific techniques need to be those appropriate for dealing with recent sediments.

Most acid lakes are oligotrophic and have slow sediment accumulation rates (often $< 1 \text{ mm a}^{-1}$). Any record of recent acidification therefore is likely to be held within the uppermost sediment. Corers that cause minimal surface sediment disturbance are therefore

needed (for example, Mackereth (1969)). In some cases it is advantageous to freeze the sediment *in situ* (for example, Huttunen & Meriläinen (1978); Renberg (1980)). Subsamples of 0.5 cm thickness allow a sample resolution of perhaps 2–10 years depending on the accumulation rate and the degree of bioturbation.

Accurate sediment dating is essential not only to construct a chronology but also to calculate rates of change. Where they can be found lakes with annually laminated sediments allow the most accurate and precise chronologies (cf. Battarbee (1981*b*); Tolonen & Jaakkola (1983)). More usually dating is carried out using ^{137}Cs analysis (cf. Pennington *et al.* ((1973)) and ^{210}Pb analysis (Krishnaswamy *et al.* 1971). ^{137}Cs analysis allows the concentration pattern of this fallout radionuclide in the sediment to be matched with reported fallout. A peak in 1963 is associated with maximum fallout levels (cf. Cambray *et al.* (1979)). This technique, however, may not be reliable in acid lakes since in certain cases there is evidence for post-depositional movement of the isotope (cf. Longmore *et al.* (1983); Davis *et al.* (1984)).

The most useful dating technique is ^{210}Pb dating. ^{210}Pb is a naturally occurring isotope with a half life of 22.26 years and it can be conveniently used for sediments deposited within the last 150 years or so. Dates are usually calculated assuming a constant rate of supply of unsupported ^{210}Pb to the sediment through time (cf. Oldfield *et al.* (1978); Appleby & Oldfield (1978, 1983)).

Diatoms are well preserved in acid conditions and occur in acid lake sediments in high concentrations. They are separated from the sediment by oxidation of the organic matrix (Battarbee 1979*b*) and identified and counted at high magnification ($\times 1000$) after mounting in a medium of high refractive index. Both the percentage frequency of the constituent taxa and the concentration of valves (Battarbee & Kneen 1982) are usually calculated.

DIATOMS AND pH RECONSTRUCTION

Diatoms can be used as good indicators of pH, and there is a substantial literature on diatom distribution in relation to pH (Hustedt 1937–39; Jørgensen 1948; Foged 1948, 1953, 1954; Meriläinen 1967, 1969; Chohnoky 1968). However this good relationship may not be owing only to the effect of H^+ concentration since pH is also correlated strongly with other important factors such as ionic strength and nutrient availability (for example, Patrick (1977)).

Despite the lack of ecological understanding attempts at reconstructing pH from diatom assemblages have been remarkably successful.

(a) *The Hustedt classification*

Most attempts to organize diatoms into pH classes start with the Hustedt system set out in a classic monograph on the diatom flora of Java, Bali and Sumatra (Hustedt 1937–1939). The diatoms were divided into five categories according to their individual pH tolerances as follows.

- (1) Alkalibiontic: occurring at pH values > 7 ;
 - (2) alkaliphilous: occurring at pH about 7 with widest distributions at pH > 7 ;
 - (3) indifferent: equal occurrences on both sides of pH 7;
 - (4) acidophilous: occurring at pH about 7 with widest distribution at pH < 7 ;
 - (5) acidobiontic: occurring at pH values < 7 , optimum distribution at pH = 5.5 and under.
- Hustedt then characterized the various pH intervals according to this scheme.

(1) pH 7: the frequent forms (die Massenformen) consist almost exclusively of alkalibiontic and alkaliphilous together with indifferent forms.

(2) pH 6–7: the majority consists of alkaliphilous species which begin to disappear within this interval, the indifferent forms are frequent, whereas about 30% represent acidophilous species.

(3) pH 5–6: alkaliphilous and indifferent forms much less numerous, the frequent forms comprise up to 75% acidophilous and acidobiontic diatoms.

(4) pH 4–5: alkaliphilous forms have disappeared, the indifferents still comprise only about 20% of the frequent forms, whereas about 80% are acidophilous and acidobiontic diatoms.

(5) pH 4: the number of forms is very small, and these are solely acidobiontic.

Although these characterizations are now known to be somewhat inaccurate, the pH categories and the terminology presented by Hustedt have found more or less general acceptance and use among diatomists. However a number of authors have pointed out that the term 'indifferent' is a misleading one since it applies to those taxa that prefer water of circumneutral pH. The word 'indifferent' and the definition 'equal occurrence on both sides of pH 7' imply that taxa in this category are insensitive to pH changes. This is not the case and was clearly not intended by Hustedt, as his characterization of the pH scale above clearly shows. Renberg (1976) defines forms belonging to this category as those 'which occur at around pH 7'. The term 'circumneutral' is less ambiguous and should be adopted instead.

(b) *The Nygaard pH indices*

Hustedt's system was followed by subsequent diatomists (for example, Jørgensen (1948)), and in 1956 Nygaard attempted to quantify the system. To do this, instead of taking only the dominant or most frequent forms (die Massenformen) into consideration he included all taxa, as well as their relative frequencies, arguing that rare species may be as informative as common ones. He categorized his taxa according to the Hustedt pH system using pH preferences reported in the literature. On the assumption that acidobiontic and alkalibiontic forms were much better ecological indicators than their respective acidophilous and alkaliphilous counterparts, he suggested that these groups should be awarded more significance in statistical calculations. He therefore weighted the relative frequencies of each acidobiontic and alkalibiontic taxon by an arbitrary factor of five. By adding the totals of alkalibiontic and the totals of acidobiontic and acidophilous taxa he produced sum totals of 'alkaline' units and 'acid' units respectively. The 'indifferent' taxa were not included. On this basis he proposed three indices as follows: index α = (acid units)/(alkaline units); index ω = (acid units)/(number of acid species); index ϵ = (alkaline units)/(number of alkaline species).

Nygaard demonstrated the applicability of the indices by calculating index α , ω , and ϵ for a series of 14 lakes with pH values less than 4 to over 7.5 and obtained a fairly good relationship between measured pH and the trends of the various indices.

Some of the limitations of this approach have been pointed out by recent authors (for example, Meriläinen (1967); Digerfeldt (1972); Renberg (1976); Battarbee (1979b)). The need for accurate information regarding the pH preference of any taxon included in the index is clear as is the need for accurate identification of taxa. However, in a situation where certain groups (acidobiontic and alkalibiontic taxa) are preferentially weighted it is even more critical that taxa are correctly allocated. The pH range and pH optimum of many taxa have still to be fully determined and literature sources can be fallible. Renberg (1976) points out the obvious

misclassification of *Anomoeoneis exilis* (Kütz.) Cleve as alkalibiontic by Hustedt (1957) for a taxon that is most common in acid waters, while *Eunotia bactriana* Ehr. classed as an acidophilous taxon (Foged 1977) seems to be restricted to waters less than pH 5.5 and should probably be regarded as an acidobiontic form (R. J. Flower, personal communication). Second, and more specific to the system proposed by Nygaard, the exclusion of indifferent (circumneutral) taxa from the indices can lead to large fluctuations in an index, especially, for example, in acid lakes that contain very few alkaliphilous or alkalibiontic taxa, and scores of infinity are possible. Third, both index ω and index ϵ are related to the number of taxa identified per sample. Since this is dependent on the total number of valves counted at any sample and on taxonomic conventions varying from person to person, the use of these indices for comparative purposes and for assigning pH changes is limited (cf. Digerfeldt (1972); Renberg (1976)). Meriläinen (1967) ascribes some of the differences between the indices for Finnish lakes and Danish lakes of the same pH to such variations in taxonomic methodology. A fourth criticism is that Nygaard's test of the indices was based on modern pH measurements gathered without due regard to seasonal fluctuations in water pH.

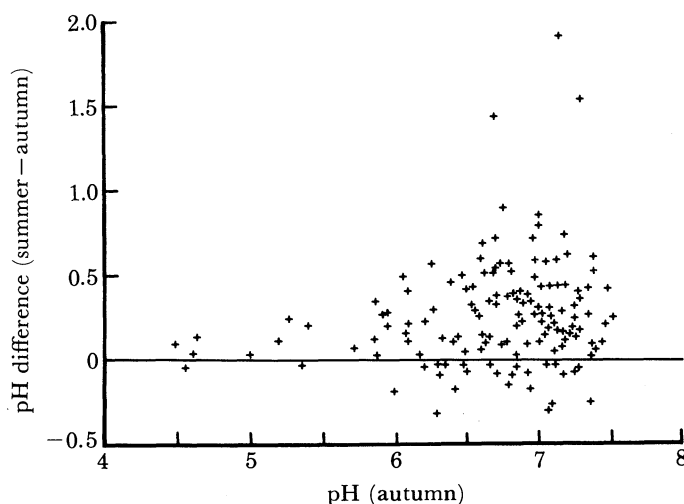


FIGURE 1. Autumn pH against pH difference (summer–autumn) for 150 lakes in Eastern Finland (from Huttunen & Meriläinen (1983)).

(c) *Meriläinen's calibration*

Meriläinen (1967) tested the applicability of the Nygaard scheme to Finnish lakes and realized that a better agreement between predicted and measured pH could be obtained if pH measurements of the lakes used in the test series were standardized by only using values from the autumn overturn period. Although the pH of acid lakes ($< \text{pH } 5.5$) show relatively little seasonal variation, more productive but poorly buffered lakes with higher alkalinity values can show substantial changes especially in summer as a result of algal photosynthesis. This relation between autumn and summer pH values for a series of lakes of differing acidity has recently been convincingly shown by Huttunen & Meriläinen (1983) (figure 1). In extreme cases the difference between autumn and summer pH can be as high as 2 pH units.

The calibration curves for each of the Nygaard indices, calculated from surface sediment diatom assemblages are shown in figure 2. For index α values were found to vary from almost

zero to infinity, with the index for acid lakes being both very high and very variable. When transformed to a logarithmic scale the values were grouped along a straight line determined by linear regression analysis with the equation $\lg \text{index } \alpha = 1.08x + 7.16$, where $x = \text{pH}$. Graphs for index ϵ and index ω also show a relation with pH. Meriläinen concluded that the indices were capable of predicting pH with an accuracy of about ± 0.5 pH unit.

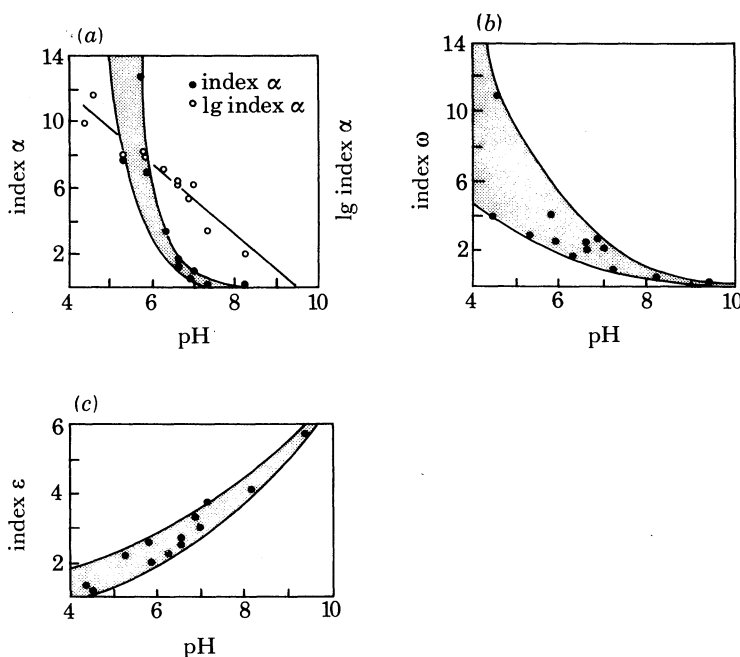


FIGURE 2. Graphs of index α , index ω , and index ϵ against measured autumn pH for 14 Finnish lakes (from Meriläinen (1967)).

A revival of interest in Nygaard's indices followed Meriläinen's paper. Foged (1969) used all three indices to reconstruct pH changes in the development of Grane Langsø. Digerfeldt (1972) considered index α to be the most reliable of the three indices and calculated its values for a Lake Trummen core. Although he found the pH indicated to be rather too high throughout, the variation in values accorded well with the lake history as inferred from other sources. Renberg (1976) also used this system and found that the calculated values for pH from the contemporary diatom flora agreed well with the measured summer pH for Prästsjön of between 5 and 6, and that the reconstructed values for the former pH levels in the lake also appeared credible.

(d) *Index B – modification by Renberg & Hellberg*

Although Renberg (1976) found that $\lg \text{index } \alpha$ predicted the contemporary pH of Prästsjön quite well he was concerned about the variability of the index caused by the exclusion of indifferent (circumneutral) taxa. Consequently Renberg & Hellberg (1982) proposed an index B in which indifferent (circumneutral) taxa have a place in both the numerator and denominator of the equation and in which the weighting coefficients are objectively calculated by the least squares method:

$$\text{index B} = \frac{\% \text{ ind.} + (5 \times \% \text{ acp.}) + (40 \times \% \text{ acb.})}{\% \text{ ind.} + (3.5 \times \% \text{ alk.}) + (108 \times \% \text{ alb.})}$$

where ind. = indifferent, acp. = acidophilous, acb. = acidobiontic, alk. = alkaliphilous, and alb. = alkalibiontic.

By using both new and literature data for the diatom content of the uppermost sediment of 30 lakes in Scandinavia, Renberg and Hellberg computed a linear function for pH calculated (figure 3) with the equation:

$$\text{pH} = 6.40 - 0.85 \lg \text{index B.}$$

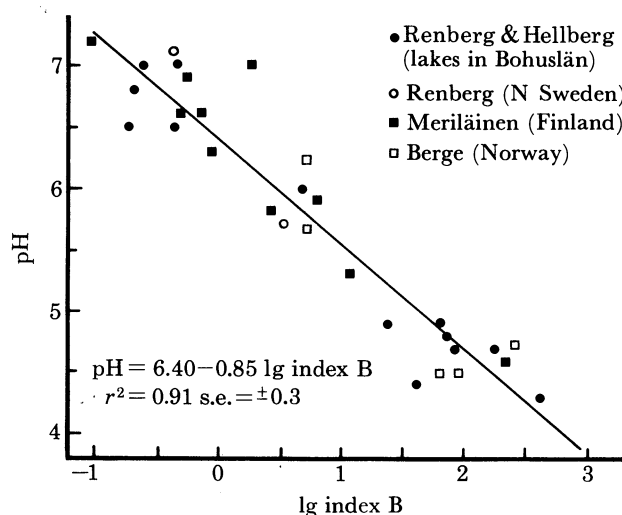


FIGURE 3. \lg index B against measured pH for 30 Scandinavian lakes (from Renberg & Hellberg (1982)).

This relationship has a high r^2 value of 0.91 and a standard error of ± 0.3 pH unit. The index has been used by Flower & Battarbee (1983) and Tolonen & Jaakkola (1983) with results giving close agreement with measured pH.

(e) *Approaches using multiple regression analysis*

Alternative approaches to the use of a system based on Nygaard's indices have been developed by Davis & Berge (1980); Davis *et al.* (1983); Davis & Anderson (1984), and Charles (1982), and involve the use of multiple regression equations using either the Hustedt pH groups or individual taxa as predictors of pH.

In the case of multiple regression analysis by pH group, Charles (1982) shows a very good predictive relationship between measured and inferred pH for an Adirondack lake data set ($r^2 = 0.93$) (figure 4) with the equation:

$$\text{pH} = 10.0389 \text{ acb.} - 0.349 \text{ acp.} - 0.00871 \text{ ind.} - 0.00134 \text{ alk.} + 7.99.$$

However, this relationship was only slightly better than the predictive power of the Nygaard–Meriläinen \lg index α with the same data set ($r^2 = 0.91$) (figure 5). The advantage of the multiple regression approach over the Nygaard method is that the coefficients are derived objectively rather than allocated subjectively. On the other hand the coefficients in index B are also derived objectively and the choice between these two approaches therefore is not based on computational accuracy but on whether a good local diatom set is available to generate locally suitable multiple regression equations. In the absence of such a data set it is appropriate to use pH categories from literature sources and to use index B.

A multiple regression analysis approach using selected taxa as predictors of pH has been developed by Davis and his coworkers. It requires the use of a calibrated local data set since despite the universal distribution of most diatom taxa the species composition of diatom assemblages can differ from region to region in a way that is not understood. Little work has

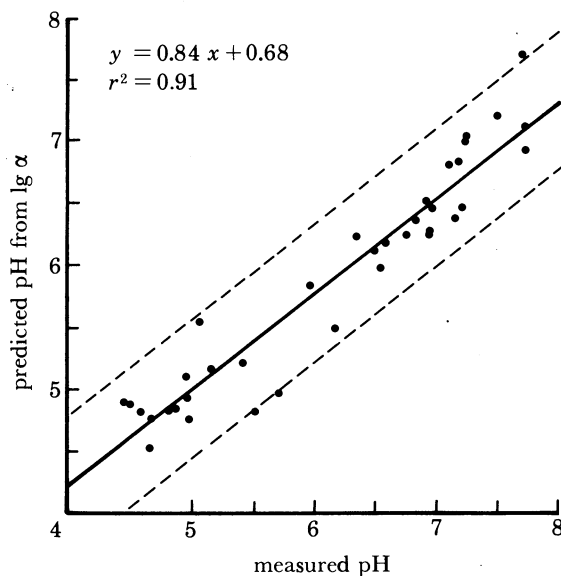


FIGURE 4. $Lg \alpha$ against measured pH for lakes in the Adirondack Mountains (Charles 1982).

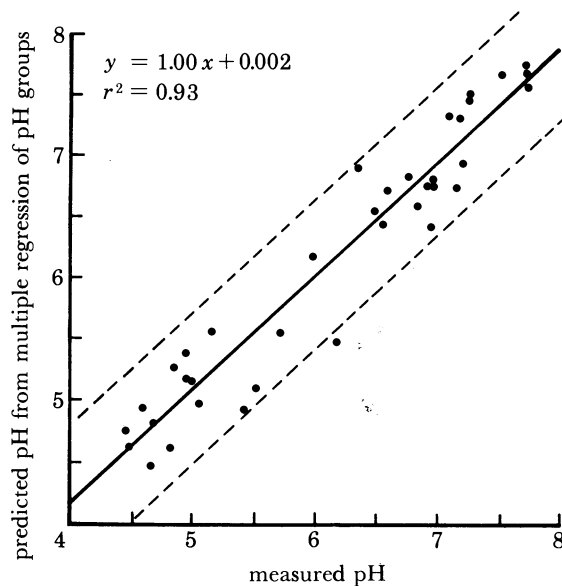


FIGURE 5. pH calculated from multiple regression of pH groups against measured pH for lakes in the Adirondack Mountains (Charles 1982).

been done on the possible ecological significance of morphotypic variation. The considerable advantage of this approach is that taxa are not placed into broad artificial groupings. The disadvantage, on the other hand, is that only a relatively small number of taxa are included resulting in a potential loss of information from the exclusion of less frequent taxa. This

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approach can also produce a high correlation between observed and predicted pH values as Davis & Anderson show (cf. figure 6) but when the various methods are compared on a sediment core (figure 7) the pH values reconstructed in this manner tend to have greater scatter because of the inherent variability of certain taxa in lake or sediment data sets. However this variability can be reduced by using instead the regression equation of the first principal component of the individual taxa data against measured pH (Davis & Anderson 1984).

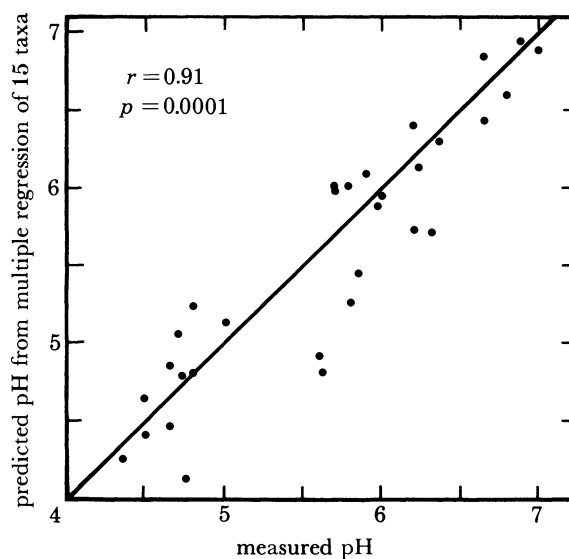


FIGURE 6. pH calculated from multiple regression of 15 taxa against measured pH for lakes in New England (from Davis & Anderson (1984)).

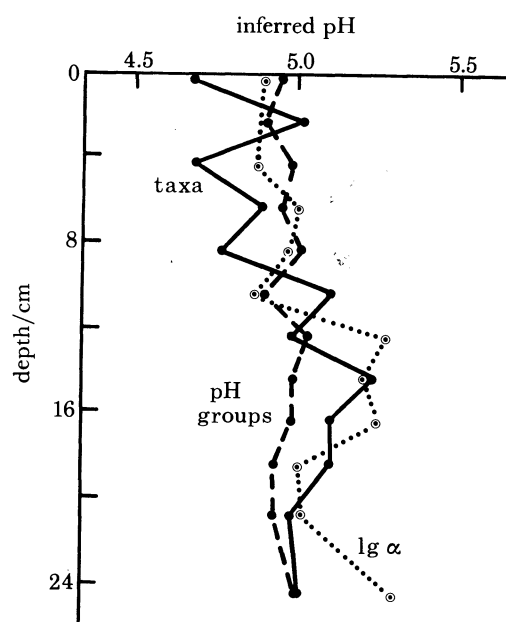


FIGURE 7. Comparison of downcore pH for Speck Pond, Maine, U.S.A., calculated from multiple regression analysis of taxa and pH groups, and from $\lg \alpha$ (from Davis & Anderson, unpublished).

(f) Errors in pH reconstruction

Whether the index system or multiple regression analysis is used the pH of a diatom assemblage from the surface sediment of most lakes can now be predicted with a standard error of *ca.* ± 0.25 to *ca.* ± 0.5 pH units (Davis & Anderson 1984; Renberg & Hellberg 1982). Since pH varies seasonally and annually and since there are also errors in the measurements of pH, especially in waters of low ionic strength (Howells 1983) further substantial reduction in the errors of pH reconstruction are unlikely. Remaining systematic errors are likely to be associated with taxonomic and ecological problems such as failure to recognize morphologically similar but ecologically different species and ecotypes and, when using the Hustedt categories, in the wrong classification of taxa. Errors may also be associated with factors other than pH. Davis & Anderson (1984), for example, have noted that those lakes that deviated most from measured values in their Norwegian data set were lakes at low elevations, low pH (\leq pH 5.5), small surface area, and high concentrations of SO₄, Al, P and Ca, while J. Ford (personal communication) has suggested that acidophilous taxa are more abundant in shallow lakes than in deeper ones at the same pH. Sedimentological factors may also be important if certain taxa are preferentially transported to the point of sediment accumulation or allochthonous diatoms are quantitatively important (R. W. Battarbee & R. J. Flower, unpublished).

However, as Davis and Anderson have pointed out, some of these factors, such as lake area, elevation, and morphometry, can be regarded as constant over substantial time periods, at least during the time period of expected recent acidification, and the shifts in pH indicated by these models are probably more accurate and precise than the statistics for the surface sediment data would suggest. The diatom content of the sediment core in most cases provides a continuous, integrated and internally consistent record of lake acidity.

DIATOM ANALYSIS OF SEDIMENT CORES AND LAKE ACIDIFICATION

The recent developments in the reconstruction of pH from diatom assemblages noted above have occurred because of the need to evaluate the timescale and extent of recent lake acidification. However, this work builds on and supplements research into limnological change that has been going on for over 50 years (Pearsall 1921; Lundquist 1924) and that recognizes that lake acidity in the longer term is controlled by climatic and edaphic as well as anthropogenic factors. It is therefore necessary to set recent (0–150 B.P.) changes into the wider context of the full post-glacial development.

(a) Post-glacial trends in lake acidification

For lakes situated in upland areas and, or, in regions of resistant base-poor bedrock palaeolimnological evidence suggests that oligotrophication and acidification are natural processes. This view first suggested by Lundquist (1924) but formulated more specifically by Quennerstedt (1955) and Mackereth (1965, 1966) argues that early post-glacial soils were base-rich and unweathered and that lakes were alkaline owing to the supply of base cations. However, as leaching of soils continued and as acid organic soils developed in lake catchments lake waters became more acidic and primary production decreased, a process appropriately termed meiotrophication by Quennerstedt (1955). The timing and rate of this process seems to vary with location. At Kentmere in the English Lake District, Round (1957) showed that

TABLE 1. DIATOMS CHARACTERISTIC OF THE SUCCESSIVE POLLEN ZONES FROM KENTMERE (FROM ROUND 1957)

depth from the surface cm	pollen analytical zone	period	type of deposit	characteristic diatoms
50–205	VIIb, VIII	post-Atlantic period	diatom-Gyttja	<i>Cymbella</i> <i>Achnanthes</i> <i>Gomphonema</i> <i>Anomoeoneis</i> <i>Eunotia</i>
205–315	VIIa	Atlantic	diatom-Gyttja	<i>Cymbella</i> <i>Achnanthes</i> <i>Eunotia</i> <i>Gomphonema</i> <i>Cyclotella</i>
315–360	VI	Boreal	diatom-Gyttja	<i>Fragilaria</i> <i>Epithemia</i> <i>Cyclotella</i>
360–375	IV, V	Preboreal	diatom-Gyttja	<i>Fragilaria</i> <i>Epithemia</i>
375–395	III	late-glacial	clay	
395–415	II		diatom-Gyttja	<i>Fragilaria</i> <i>Epithemia</i> <i>Melosira arenaria</i> <i>Navicula järnefeltii</i>
415–475	I		diatom-Gyttja grading into clay	<i>Fragilaria</i> <i>Epithemia</i> <i>Navicula järnefeltii</i> <i>Melosira arenaria</i> <i>Cymbella thumensis</i>

the beginning of acidification was coincident with the Boreal–Atlantic transition (about 7500 B.P.): a time when upland blanket peat was beginning to form in the Pennines.

Although he did not attempt to quantify the pH change indicated, the diatoms changed from a predominantly alkaliphilous *Fragilaria–Epithemia* assemblage in the late-glacial and Boreal periods, through a *Cyclotella–Melosira* stage and thence, by the mid-Atlantic period to an acidophilous *Achnanthes–Anomoeoneis–Eunotia–Cymbella–Gomphonema* assemblage (table 1). In Denmark a similar analysis of Støre Gribsø sediments (Nygaard 1956) showed that acidification did not begin until about 400 B.C. or the beginning of the sub-Atlantic period. He ascribed the changes to climate deterioration leading to the leaching of Ca^{2+} ions, an increase in the formation of raw humus in the adjacent beech forest floor and an increase in the influence of water from surrounding bog areas. Using his index α (see above), he concluded that the pH of the lake had changed from between pH 7 and 9 in the Atlantic period to about pH 4–5 by about 400 B.C. Many papers have since been published identifying acidifying trends in lakes in NW Europe including Scotland (Alhonen 1968; Pennington *et al.* 1972), NW England (Round 1957, 1961; Haworth 1969; Evans 1970), North Wales (Crabtree 1969; Evans & Walker 1977), Sweden (Digerfeldt 1972, 1973, 1975, 1977, Renberg 1976, 1978), and Finland (Huttunen *et al.* 1978). In most cases the main period of change has been in the early post-glacial period.

More recently and in order to provide a context for the study of recent acidification Renberg

& Hellberg (1982) have made quantitative reconstructions of the pH of a lake over the post-glacial time period. Figure 8 shows the changes for Lake Gårdsjön over a 12500 a year period and includes a pH curve reconstructed using index B. Despite a rather irregular period in the late-glacial sediment at the base of the profile the progressive decrease in pH from > 6.5 to about 5.7 can be seen as alkaliphilous taxa are replaced by acidophilous ones. The recent acidification from about 1950 (Renberg & Hellberg 1982) is not shown on this diagram.

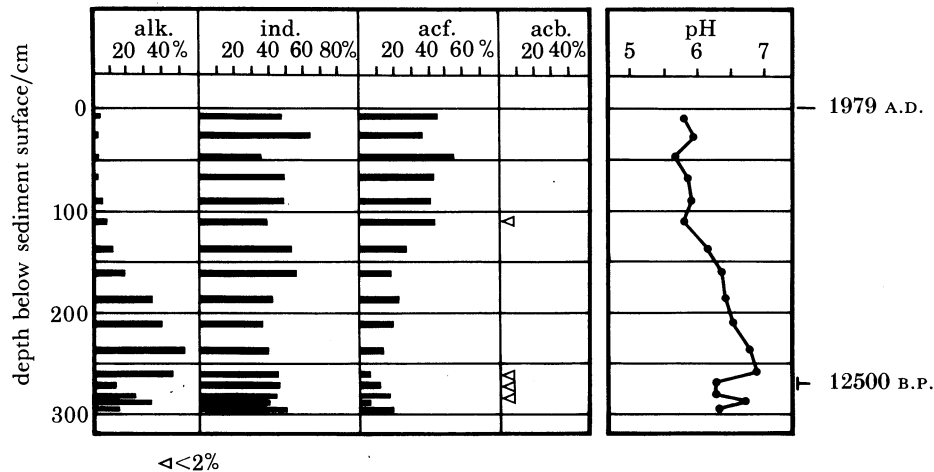


FIGURE 8. Variations in pH and pH groups over the late and post glacial history of Gårdsjön, Bohuslän, Sweden (from Renberg & Hellberg (1982)).

It is clear that the long term tendency for lakes in areas of acid bedrock is to become more acid. In most cases the period of most rapid change occurs during the first part of the post-glacial period although further changes often take place and these tend to be associated with soil-vegetational changes in the catchments. Changes when they do occur tend to be towards a lower pH but there is no evidence to suggest that these decreases are necessarily progressive and continuous, and there are no examples, so far, of a lake falling to a pH of less than 5 during these earlier periods.

(b) *Diatom evidence for recent acidification (0–150 B.P.)*

(i) *Sites outside the United Kingdom (table 2)*

Evidence for recent acidification from diatom changes in lake sediment was first presented by Miller (in Almer *et al.* (1974)). She reported a strong decrease in the abundance of valves in the upper sediment of Stora Skarsjön in SW Sweden and noted that planktonic diatoms decreased and the relative frequency of a number of acidobiontic taxa, including *Tabellaria binalis* (Ehr.) Grun. and *Amphicampa hemicyclus* (Ehr.) Karsten increased. Berge (1975) carried out a similar investigation in Langtjern, Norway, and although no dating was carried out he suggested that the 80 cm core represented a period in excess of 800 years. The diatom analysis of the core shows that the lake was highly acid (about pH 5) throughout the whole period although a slight expansion in the uppermost 2 cm of *Amphicampa hemicyclus*, *Tabellaria binalis* and *Navicula subtilissima* Cleve is quoted as evidence for a slight recent acidification.

Clearer evidence of acidification was presented by Berge (1979) from the analysis of three lakes in Agder County, S Norway and two in Hordaland County, on the west coast of S Norway.

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TABLE 2. ACIDIFICATION DATA FOR LAKES FROM NORWAY, SWEDEN, FINLAND, THE U.S.A. AND CANADA

lake†	region	year of core	modern pH (observed)	approx. onset of acidification	pre-acidification pH (predicted)	modern pH (predicted)	pH decline	pH reconstruction	methods dating
Norway									
¹ Langtjern	Buskerud	1975	4.68–5.16	—	4.3–6.2	< 5.5	—	index ω	—
² Övre Målmesvatn	Vest-Agder	1977	4.53	1927–37	4.4–6.5	< 5.2	30.5	index ω	¹³⁷ Cs
² Övre Botnatjønn	Hordaland	1977	4.74	—	< 5.0	< 5.1	—	index ω	—
² Rödlivatn	Hordaland	1977	6.23	—	5.1–6.8	5.2–6.9	—	index ω	—
² Högteivvatn	Aust-Agder	1977	4.47	—	4.7–6.6	< 6.3	—	index ω	—
² Risvatn	Aust-Agder	1977	5.66	—	5.7–7.2	5.4–7.1	—	index ω	—
^{4,5} Hovvatn	S Norway	⁷ 1978	4.4	1918	4.8–5.4	3.9–4.4	0.75	⁶ MR	²¹⁰ Pb
^{4,5} Blåvatn	nr Bergen	⁷ 1978	5.1	1930	5.2	5.1	0.1	⁶ MR	²¹⁰ Pb
⁵ Norre Målmesvatn	Vest-Agder	1979	4.6	1890	84.9–5.3	84.4–4.5	0.6	⁶ MR	²¹⁰ Pb
⁵ Holmvatn	—	1978	4.7	1927	4.8–5.2	4.5	0.5	⁶ MR	²¹⁰ Pb
⁵ Dorsvatn	—	1978	5.0	—	⁸⁵ 0	⁸⁵ 0	0.0	⁶ MR	²¹⁰ Pb
⁵ Brårvatn	—	1979	5.2	1850	85.3–6.1	85.2–5.3	0.3	⁶ MR	²¹⁰ Pb
⁵ Botnavatn	—	1978	5.7	1920	85.9	85.7	0.2	⁶ MR	²¹⁰ Pb
⁵ Opplosvatn	—	1979	5.8	—	85.8	85.8	0.0	⁶ MR	²¹⁰ Pb
⁵ Grønivatn	—	1978	6.5	—	86.5	86.5	0.0	⁶ MR	²¹⁰ Pb
Sweden									
⁹ Stora Skarsjön	W Coast	1972	4.5	—	6.0	4.5	1.5	index ω	—
¹⁰ Gårdsjön	Bohuslän	1979	4.6	1950	6.0	4.5	1.5	index B	¹¹ oilsoot
¹⁰ Härsvatten	Bohuslän	1979	4.4	—	5.9	4.2	1.7	index B	—
¹⁰ Lysvatten	Bohuslän	1979	¹² 5.9	—	6.1	¹³ 5.2	0.9	index B	—
Finland									
¹⁴ Hauklampi	N Espoo	1982	4.75–4.9	1961–4	6.0–6.4	5.1–5.4	1.05	index α/B	²¹⁰ Pb
¹⁴ Orajärvi	N Espoo	1982	4.7–4.8	?	6.3	4.9	1.4	index B	—
¹⁴ Haklajärvi	N Espoo	1982	4.88–5.10	?	6.30	5.60	0.7	index B	²¹⁰ Pb
U.S.A.									
¹⁵ Honnedaga	Adirondacks	1976	4.7	1966	6.2	5.6	0.6	index α	¹³⁷ Cs
¹⁵ Woodhull	Adirondacks	1976	5.2	—	6.0	6.0	0.0	index α	¹⁶ —
¹⁵ Seventh	Adirondacks	1976	6.5	—	6.5	6.5	0.0	index α	—
¹⁷ Big Moose	Adirondacks	1982	4.7	1950	5.7	4.7	1.0	¹⁸ MR/index α/B	²¹⁰ Pb
⁵ Solitude Pond	New Hampshire	1979	4.8	1920	⁸⁵ 1	⁸⁴ 8	¹⁹⁰ 3	⁶ MR	²¹⁰ Pb
⁵ Speck Pond	Maine	1978	4.7	1920	84.9–5.2	84.7–5.0	0.2	⁶ MR	²¹⁰ Pb
⁵ Ledge Pond	Maine	1979	4.5	1905	84.9	84.5	0.4	⁶ MR	²¹⁰ Pb
⁵ Tumbledown Pond	Maine	1978	4.8	1970	85.05	84.8	¹⁹⁰ 25	⁶ MR	²¹⁰ Pb
⁵ E. Chairback Pond	Maine	1979	5.0	1960	85.2	85.0	¹⁹⁰ 2	⁶ MR	²¹⁰ Pb
⁵ Klondike Pond	Maine	1978	5.9	1945	86.1	85.9	0.2	⁶ MR	²¹⁰ Pb
Canada									
²⁰ B	Ontario	1981	5.2	1962	6.2	4.7	1.5	index α	¹³⁷ Cs
²⁰ Cs	Ontario	1981	5.2	1954	7.1–7.3	6.4	0.8	index α	¹³⁷ Cs

† 1, Berge (1975); 2, Berge (1979); 3, From author's interpretation. A clear acidification is observed at this site despite the ambiguous index ω predictions; 4, Davis & Berge (1980); 5, Davis *et al.* (1983); 6 Multiple regression techniques including pH groups, taxa and first principal component regressions; 7, Lake also referred to in Davis & Berge (1980). Here the revised data in Davis *et al.* (1983) are presented; 8, Davis *et al.* (1983) do not indicate predicted pH values, only the predicted fall in pH. These values are back-calculated assuming predicted pH = modern pH; 9, Almer *et al.* (1974); 10, Renberg & Hellberg (1982); 11, Renberg & Wik (1984); 12, Elevated value after liming in 1974; 13, 1974 pH before liming; 14, Tolonen & Jaakkola (1983); 15, Del Prete & Schofield (1981); 16, By comparison of ¹³⁷Cs from Woodhull; 17, Charles (1984); 18, Multiple regression of pH groups; 19, Some regression methods show no change; 20, Dickman *et al.* (1983).

Except for ^{137}Cs dates for Övre Målmesvatn these data were also limited by lack of secure dating. Of the five lakes only Övre Målmesvatn (samples in 1977 with a pH of 4.53) was analysed in some detail and this was the only lake to show a clear acidification from a level of about 6 cm in the sediment, thought, from the ^{137}Cs analysis to date to 40–50 years B.P. (1927/37). Indifferent (circumneutral) taxa such as *Anomoeoneis exilis*, *Navicula cocconeiformis* Gregory and *Cymbella hebridica* (Gregory) Grun. decline and acidobiontic taxa such as *Tabellaria binalis* and *Amphicampa hemicyclus* increase. Throughout this study Berge uses index ω of Nygaard to reconstruct pH and produces pH values that appear to be inaccurate. In a cooperative study with Davis, however (Davis & Berge 1980) the results from two additional Norwegian lakes, Hovvatn and Blåvatn, are presented, and pH values are reconstructed using a variety of multiple regression techniques (Davis & Anderson 1984). These show clear recent acidification at Hovvatn from pH 4.7–5.3 down to 3.9–4.4, but little change in Blåvatn, although a small increase in *Eunotia exigua* (Bréb.) Rabh. was noted in the upper 3 cm. The Hovvatn change, on the other hand, is associated with a strong increase in acidobiontic taxa in the upper 4 cm with the first signs of acidification at 8.5 cm. In the 1980 paper Davis and Berge estimate from ^{137}Cs dating, that initial acidification dates from 1945 at Hovvatn and 1950 at Blåvatn. More recently, however, these dates have been revised to 1918 and 1930 respectively (Davis *et al.* 1983).

Diatom evidence for acidification of additional Norwegian lakes is presented by Davis *et al.* (1983) and the changes are dated using ^{210}Pb analysis, a more reliable and comprehensive method than the ^{137}Cs they had previously used. The data from Holmvatn are given in detail and show that on the basis of diatom changes acidification began in the 1920s. Six other lakes in Norway were also studied (table 2) and Davis *et al.* (1983) compute pH decreases of 0.3–0.8 units starting between 1890 and 1930.

In Sweden Renberg & Hellberg (1982) carried out diatom analyses on three acid lakes in Bohuslän. They reconstructed former pH values using their index B and dated the recent changes using counts of oilsoot particles in the sediment (Renberg 1984). All three lakes show recent periods of rapid acidification. In Gårdsjön the change is confined to the upper 7 cm from 1960 to the present showing a change from pH > 6 to about pH 4.5. The uppermost sediment is characterized by the acid taxa *Anomoeoneis seriens* (Bréb.) Cleve, *Eunotia bactriana*, *E. denticulata* (Bréb.) Rabh, *E. exigua*, *E. veneris* (Kütz.) O. Müller, *Navicula subtilissima*, and *Tabellaria binalis*. In addition, as noted by Almer *et al.* (1974) the diatom plankton decreases rapidly as acidification progresses. Härsvatten and Lysevatten also show near surface declines in pH from about 5.9 to about 4 and from about 6.1 to 5.2 respectively. Unfortunately no dating is available for either of these two cases but a minimum of pH at 3 cm followed by an increase to the surface in Lysevatten is thought to represent the effect of liming in 1974.

There has been considerable work in Finland on the diatoms of naturally acid lakes (Meriläinen 1967; Meriläinen & Huttunen 1984), and Tolonen & Jaakkola (1983) have recently reported studies of lake sediments for a number of sites in SW Finland that have experienced recent pH declines (Pätilä 1982). For two of the four lakes (Sorvalampi and Häkljärvi) insufficient stratigraphical detail is presented for clear conclusions to be reached, but for Hauklampi and Orajärvi the trend of recent acidification can be seen from index B computations showing a decline in pH from about 6 to about 5.1–5.4 for Hauklampi and a decline from about 6.3 to about 5.0 for Orajärvi. No dating is available for the latter case but for Hauklampi ^{210}Pb dating shows the decline in pH to start from about 1961–1964.

In the U.S.A. data are available on recent sediments from the Adirondack Mountains (del

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Prete & Schofield 1981; Charles 1984) and from lakes in New England (Davis *et al.* 1983). Del Prete and Schofield selected three lakes, Honnedaga, Woodhull and Seventh, of which only Honnedaga showed a significant pH decrease in the uppermost part of the core (from about 6.2–5.6). The authors suggest, by comparison with the ^{137}Cs based sediment accumulation rate of nearby Woodhull lake, that the change in Honnedaga lake occurred at least ten years before the core was taken (1976).

Charles (1984) has analysed a core from Big Moose Lake using ^{210}Pb analysis for dating and a comparison of index α , index B and a multiple regression equation of pH preference categories to reconstruct pH. He concludes that from about 1800–1950 pH was about 5.7 and that after 1950 pH dropped steadily and rapidly over a ten year period to its present value of about 4.7. The present surface sediment is dominated by *Stauroneis gracillima* and an acid variety of *Fragilaria virescens*.

TABLE 3. ACIDIFICATION DATA FOR GALLOWAY LAKES BASED ON INDEX B pH RECONSTRUCTION (RENBERG & HELLBERG 1982) AND ^{210}Pb DATING

Galloway lakes, SW Scotland				
	Altitude m	percentage afforestation	date of planting	date of acidification
Loch Enoch	493	0	—	1840
Round Loch of Glenhead	299	0	—	1850
Loch Grannoch	210	69.5	1962, 1976	1925
Loch Dee	225	20.8	1976	1890
	pre- acidification pH	modern pH (predicted)	modern pH (ob) (observed)	pH decline
Loch Enoch	5.2	4.3	4.4–4.7	0.9
Round Loch of Glenhead	5.7	4.7	4.5–5.0	1.0
Loch Grannoch	5.6	4.4	4.4–4.9	1.2
Loch Dee	6.1	5.6	4.9–5.9	0.5

Sediments from six New England lakes have been studied by Davis *et al.* (1983). They also used ^{210}Pb dating and a variety of pH reconstruction techniques. The data are presented in table 2. All lakes showed some signs of recent acidification but at E Chairback Pond, Tumbledown Pond and Solitude Pond some pH regressions indicated no change in pH. The earliest evidence of the onset of acidification was 1905 at Ledge Pond. This site also showed the largest decline in pH of about 0.5 unit.

In Canada two sites in Ontario have been investigated by Dickman *et al.* (1983), on the basis of index α and ^{137}Cs dating they conclude that the pH of one lake dropped from 6.2 to 5.2 over the last 20 years while the pH of the second lake had dropped from 7.1 to 5.2 over the last 30 years.

(ii) *Sites in Galloway, SW Scotland*

Index B (Renberg & Hellberg 1982) and ^{210}Pb dating is being used to reconstruct former pH values of a group of lakes in the upland granite areas of Galloway, SW Scotland. So far data are available for four lakes (table 3), two of which are reported in Flower & Battarbee (1983). Three lakes, Loch Enoch, Round Loch of Glenhead and Loch Grannoch have mean pH values below 5 and have catchments entirely on granite, while the fourth, Loch Dee, is at a lower altitude and has some of its catchment on metamorphic rocks. It has a contemporary seasonal

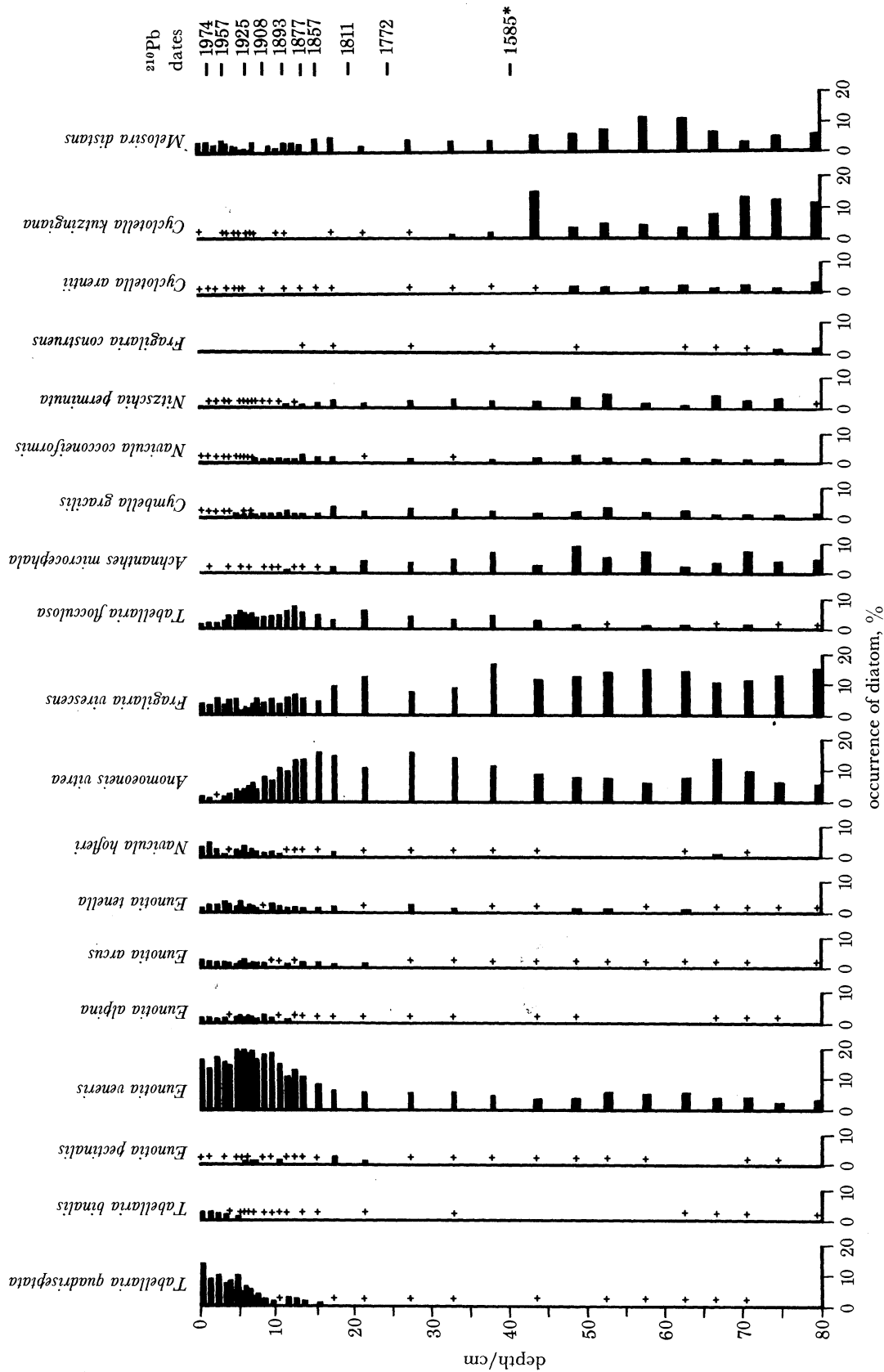


FIGURE 9. Diatom diagram and ^{210}Pb dates for Round Loch of Glenhead, Galloway, Scotland (from Flower & Battarbee (1983)). * Extrapolated date.

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pH range of 5.1–5.9. All lakes show very clear acidification trends in the recent sediment. In Loch Dee the acidification has not yet caused the development of an acidobiontic flora but it has led to the progressive loss of diatom plankton. In this lake the decline in pH has been from about 6.1 to 5.6 starting in about 1890. In the other three lakes pH changes from about 5.7 to less than 5.0 have occurred and have involved the development of *Tabellaria binalis*–*Tabellaria quadriseptata* Knudsen communities in all cases. However, dates for the onset of acidification range from 1850 at Round Loch of Glenhead and Loch Enoch to 1920–30 at Loch

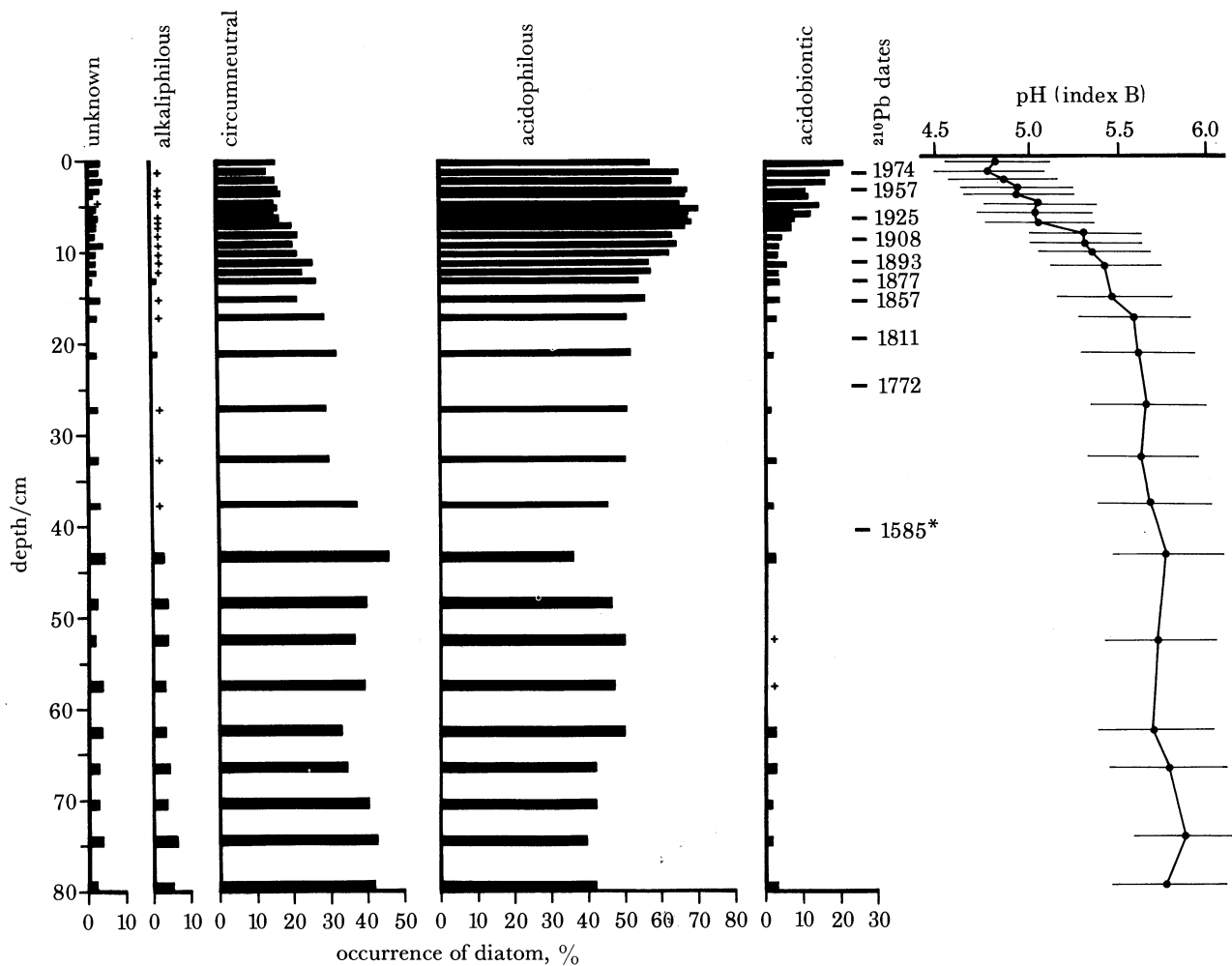


FIGURE 10. pH groups and pH reconstruction (index B) for diatoms from Round Loch of Glenhead, Galloway, Scotland. * Extrapolated date. Error bars indicate ± 0.3 pH unit s.e. quoted for the index B method (Renberg & Hellberg 1982).

Grannoch. The diatom diagrams from Round Loch of Glenhead (figures 9 and 10) show the typical progression of species at these sites and the shift to acid dominated communities.

The earliest assemblage shown in figure 9 from 80–40 cm represents the period of time covered by the core before approximately 1600 A.D. Both periphytic taxa (*Anomoeneis vitrea* (Grun.) Ross, *Eunotia veneris*, *Fragilaria virescens* Ralfs) and planktonic taxa (*Cyclotella kützingiana* Thwaites) are common. Between 45 and 40 cm there is a very marked decline in the planktonic *Cyclotella* species. The pH reconstruction suggests that there was a slight decrease in pH from

about 5.7 to 5.5. The loss of plankton at this stage accords with the observations of Charles (1982) and Davis *et al.* (1983) that planktonic *Cyclotella* taxa are rare below pH values of about 5.5.

This change is followed by a 250 year period (1600–1850) of relative stability with a pH of about 5.5. However at about 15 cm (1850) the beginning of an acidification process that culminates with the modern lake situation is registered by the decrease in the circumneutral taxa *Fragilaria virescens* and *Achnanthes microcephala* (Kütz.).

Decline in other circumneutral taxa (*Nitzschia perminuta* (Grun.), *Navicula cocconeiformis* and *Cymbella gracilis* (Rabh.) Cleve) follow and are replaced by acidophilous and acidobiontic taxa (*Tabellaria quadrisepata*, *T. binalis*, *Eunotia* sp. and *Navicula höfleri* Cholnoky). The changes represent a drop in pH from 1850 to the present day of almost one unit from about pH 5.5 to pH 4.6 (figure 10).

DISCUSSION

(a) *Diatom responses to acidification*

Studies of the relation between diatoms in the surface sediment of lakes and measured pH show that diatom assemblages can be used to predict lake water pH with a standard error of between *ca.* ± 0.25 and *ca.* ± 0.5 pH unit. Whether an index system or a multiple regression approach is used this relationship allows past pH levels to be inferred with some confidence. There are some weaknesses in the methods, associated with occasional faulty categorization of diatoms, taxonomic difficulties with some groups and in some cases slight biases in the representativity of the sediment sample site. However, these problems mainly affect the accuracy of pH prediction; they do not weaken the ability of the palaeolimnological approach to detect trends directly from the way species and groups of species change in the core (*cf.* figures 9 and 10). In almost all studies (Almer *et al.* 1974; Renberg & Hellberg 1982; Flower & Battarbee 1983; Charles 1982; Davis *et al.* 1983), the diminution and loss of a diatom plankton component can be seen to be one of the first major signs of acidification occurring at the relatively high pHs of between 5.5 and 5.8. *Cyclotella* species seem to be especially vulnerable. The reason for the loss of diatom plankton is not clear. Although acid waters tend to have very high transparencies other non-diatom plankton, especially Dinophyceae and Cryptophyceae, are abundant and are successful competitors in these conditions suggesting that N and P nutrient availability is not the main factor explaining the diatom decrease. The decline may be related to silica availability, a nutrient specific to diatoms, since concentrations tend to be low in acid waters and co-precipitation of silica with Al^{3+} occurs (Dickson 1980). Si uptake by benthic diatoms is also likely to cause reduction in Si available to plankton during the growing season but this may be more an effect following the loss of plankton than a cause of the plankton decrease.

Following the decline of planktonic diatoms in a lake undergoing progressive acidification, periphytic diatom taxa that prefer circumneutral water are the first to decline at a pH of about 5.5. Typical taxa are *Achnanthes microcephala*, *Fragilaria virescens*, *Cymbella gracilis*, *Nitzschia perminuta* and *Anomoeoneis vitrea*. By the time the lake reaches a pH of about 4.5 these taxa are likely to constitute less than 10% of the total flora. Below about pH 5.5–5.0 these taxa tend to be replaced in the diatom communities by the gradual expansion of acidophilous species such as *T. flocculosa* (Roth) Kütz., *Frustulia rhomboides* (Ehr.) de Toni and *vars.*, *Eunotia veneris* and *Achnanthes marginulata* Grün. Continuing acidification beyond pH 5.0, however, leads to the expansion of acidobiontic taxa most of which are not to be found at all in waters with pH values

higher than 5.5. Typically only a small number of taxa are involved and these vary somewhat from region to region. *Tabellaria binalis* seems to be the most faithful acidobiontic form increasing in the surface sediments of all lakes so far reported in Norway, Sweden, Finland, Scotland and U.S.A. (cf. table 2). *Semiorbis hemicyclus* (= *Amphicampha hemicyclus*) increases parallel to *T. binalis* in Norwegian lakes but this acidobiontic diatom is very rare in sites in Galloway where *T. quadriseptata* is the dominant acidobiontic taxon. Other acidobiontic forms that are more or less common include *N. subtilissima*, *Eunotia exigua* and *Anomoeneis serians*.

The almost complete restriction of these acidobiontic taxa to waters with pH less than 5.5 suggests that they prefer conditions where alkalinity is completely lacking. As such they are excellent indicators of extreme acidity and their rapid expansion in these acid waters perhaps suggests that lake pH drops rapidly after the loss of bicarbonate buffering associated with waters of pH > 5.5. If this is the case, then the characteristic steepening of the reconstructed pH curve (cf. figure 10) after many years of slow change does not necessarily represent any acceleration in an acidifying influence, merely the rapid crossing of a physico-chemical threshold. Interpretations of reconstructed pH curves in terms of changes in acid emissions should consequently make allowance for this process.

(b) Chronology of recent acidification

From the techniques of pH reconstruction described above and the ^{210}Pb dating of recent sediments it is possible to describe the acidification processes of almost any lake. So far there are few sites that have both detailed stratigraphic diatom data and reliable dating but some trends can be discerned (tables 2 and 3).

The first striking observation concerns the range of dates reported for the beginning of recent acidification, even taking into consideration the difficulty of deciding at which stratigraphic point the decrease begins. The dates vary from country to country and within countries. SW Finland and Sweden have dates of about 1960 (Renberg & Hellberg 1982; Tolonen & Jaakkola 1983), Norwegian dates range from 1890 to 1930 (Davis *et al.* 1983) while Scottish dates vary from 1850 to 1920 within a region of 400 km² (Flower & Battarbee 1982). In almost all cases thus far the pH of the lake before recent acidification has been pH 6.3 or less showing that most acidified lakes reported were already substantially acidic before the recent changes. The change in pH has varied from zero to a decline of up to 1.7 units, but in many cases the decline has been less than 1 pH unit. The long history and slow rate of acidification in SW Scotland shows why chemical records of the last 20–30 years for the upland tarns of the English Lake District are unlikely to show significant change (Sutcliffe *et al.* 1982).

(c) Causes of recent acidification

Throughout the recent debate lake acidification has been widely attributed to an increase in the acidity of precipitation as a result of emissions from fossil fuel combustion (for example, Jensen & Snekvik (1972)). Other hypotheses have also been presented, some as alternatives and others as additional causes. These include the suggestion that long-term natural acidification is an important factor (Pennington 1981) and that land use changes within lake catchments could be acidifying influences (Rosenquist 1978). It is possible to test some of these alternative hypotheses using lake sediments, by site comparison, and by careful examination of the historical data.

It can be seen clearly from the sediment data that long term natural acidification cannot be regarded as a major cause of the observed recent acidification. Significant shifts in pH, mainly

downwards, have occurred throughout the history of many acid lakes associated with climatic, edaphic and vegetational changes, but these changes tend to lead to readjustments of water acidity from one more or less stable situation to another rather than to continuous decreases. In almost all published accounts described above, the reconstructed pH of the water before recent acidification was unchanging for long periods and evidence for even longer periods of stability is restricted more by the length of the core taken (usually less than 1 m) rather than by evidence for earlier change.

The possible effects of land use change are less easy to identify unless lakes with undisturbed catchments or with a well-documented history of catchment change can be considered. In Scotland one possible cause of acidification is recent afforestation, either as a result of the uptake of base cations during forest growth, drainage operations enhancing runoff, or the effects of conifers trapping and concentrating acid gases and aerosols. Studies of paired catchments (Harriman & Morrison 1982) have shown that afforested catchments produce higher concentrations of H^+ , Al, and SO_4^{2-} , Na^+ and Cl^- in stream drainage than adjacent moorland catchments. However, the sediment data from Galloway shows that lakes with non-forested catchments have been acidified and that lakes with afforested catchments were acidified before afforestation (Flower & Battarbee 1983), demonstrating that afforestation could not be regarded as a sole or primary cause of acidification in this region. Unfortunately it is impossible yet to ascertain whether afforestation is an important contributory factor in Galloway since erosion from preafforestation ploughing probably introduces a large number of diatoms from catchment peats into the lakes and distorts the sediment record (R. W. Battarbee & R. J. Flower, unpublished).

The non-afforested catchments in Galloway are characterized by upland moorland vegetation used for sheep grazing. Certain agricultural changes are likely to have taken place over the last 150 years or so involving changes in sheep number and the frequency of moorland burning. However, in the region as a whole, there has been no land abandonment that might have led to the increase of mature *Calluna* heathland and that might have promoted acidification, and there is no evidence in the lake sediments from lithostratigraphic, or pollen analytical data to suggest that there might have been such a change during this time. If indeed such a change has taken place its effects can only be small since lakes throughout the country would have been severely acidified during prehistoric times as upland moorland was created following deforestation by early man (Dimbleby 1962). Nevertheless, the effect of land use change on lake acidity can never be discounted as long as lakes with a proven lack of human disturbance in their catchments cannot be found. In Norway many upland lake catchments have experienced a reduction in grazing and the regeneration of coniferous forest during the last 100 years (Rosenquist 1978), and in N America some of the pH declines observed may be owing to a recovery of lake catchments from eutrophication and elevated pH levels consequent on late 19th century and early 20th century lumbering activities (Davis & Anderson 1984).

Despite some ambiguous situations associated with land use changes the weight of evidence favours acid precipitation as the main cause of recent acidification in the lakes so far studied. Its effect has yet to be disproved at any site and the temporal and spatial patterns of acidification within NW Europe, limited though the data are, are consistent with such an hypothesis. In all cases the onset of acidification postdates about 1800, after the development of coal as a major power source during the industrial revolution, and the later acidification of Swedish and Finnish lakes may be associated with a postwar increase in emissions from oil combustion (Ottar 1977) as well as a change in the pattern of emissions.

A direct relationship between emissions and water pH, however, may not always be expected because of the varying buffering effects of lake catchments (Flower & Battarbee 1983). Evidence for a delayed response has been put forward by Davis *et al.* (1983) and Tolonen & Jaakkola (1983) from comparisons between diatom changes and heavy metal analyses of lake sediments. In Holmvatn, Norway, Pb increases in sediment dated to about 1835 and in Sorvalampi, SW Finland, the Pb increase dates from 1850. The authors believe that these increases are evidence of atmospheric Pb transport associated with early industrialization, and that lakes were likely to be receiving some acid deposition at that time. The effect on water quality and the consequent diatom response, however, was delayed by catchment buffering for up to a century.

Lake sediments can yield a large amount of information on the extent, rate, and causes of lake acidification. Diatom analysis is likely to remain the most effective technique for pH reconstruction but analyses of other chemical and biological parameters are complementary and can enhance ecological and palaeoecological interpretation. The diatom methodology has improved rapidly in recent years by the development of large modern surface-sediment data sets and the introduction of techniques for pH reconstruction. On the other hand the interpretation of the ecological significance of diatom changes requires much more attention, especially in terms of the interrelationships between diatoms and other primary producers in acidifying waters, and the relationship between periphytic diatom communities in particular and the physico-chemical changes that take place as lakes lose their alkalinity.

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Discussion

B. A. THRUSH, F.R.S. (*University of Cambridge, Department of Physical Chemistry, Cambridge, U.K.*). Could Dr Battarbee tell us dates for the onset of acidification obtained from diatom records by other workers?

R. W. BATTARBEE. I have reviewed the available literature for dates of the onset of acidification using diatom records and the data are presented in the written version of this paper (see table 2). There is considerable research activity throughout America and Europe and these data are likely to be supplemented rapidly. At present the total picture is unclear. In some cases the acidification is very recent, i.e. within the last 20 years, in others, as in Galloway, it began in the 19th century. The differences are likely to be due to variations in local versus regional influences and variations in the general buffering characteristics of catchment soils, as well as to regional and national variation in acid emission history.

R. S. CLYMO (*Botany and Biochemistry Department, Westfield College, University of London, U.K.*). There is reason to suppose that some of the effects on aquatic organisms result from short lived episodes – a few days at most – and others from longer term changes in water chemistry. Short-term changes would, presumably, not be detectable directly in the diatom record but (a) do they affect the diatoms at all and (b) do they leave any indirect record?

R. W. BATTARBEE. I know of no studies either in field conditions or in laboratory experiments that attempt to evaluate the effect of suddenly lowered pH on diatom growth and survival. However, I suspect that the effect on diatoms in acid lakes is minimal since most of the diatoms occupy periphytic rather than planktonic habitats in such lakes and recent observations have shown that these microhabitats do not experience the sharp fluctuations in pH that can occur in the surface waters during a snowmelt pulse. If there were an effect, short-term seasonal changes would not normally be detectable in the diatom record. This is because sediments in such lakes usually accumulate slowly and even a thin slice of sediment (2–5 mm) is likely to represent a period of time in excess of 1–2 years. Moreover, in these situations only a small amount of bioturbation is necessary to smooth the record. It is likely that this matter could

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only be investigated in lakes that have annually laminated (varved) sediments where techniques of *in situ* freeze coring and thin section or adhesive tape peels were used to examine sediments laid down during successive spring-melt periods.

E. D. LE CREN (*Freshwater Biological Association, The Ferry House, Ambleside, Cumbria, U.K.*). Several decades ago the largest iron works in Europe were in SW Cumbria; they now no longer exist. Perhaps studies of diatoms from the sediments of tarns nearby would be informative. The Freshwater Biological Association has started such studies.

P. GOLDSMITH (*Meteorological Office, Bracknell, Berkshire, U.K.*). Each of the diatom records described are for lakes in fairly remote areas and it would appear that the deduced record of acidity reflects the general background deposition. It would be a rather nice test of the method if a lake near a declining industrial area could be investigated. The diatom record in this case should then reflect the changes in local deposition.

R. W. BATTARBEE. It would indeed be interesting to investigate changes in the recorded diatom flora of a lake after the removal of an acidifying influence, and Mr Le Cren's comment points to a site where this could be carried out. However, there are problems here since although a recovery from a more acid state might point to the overriding importance in the past of the local source, absence of such a recovery might be interpreted either as a failure of the lake to recover or the continued and dominant influence of acid deposition on a regional scale. Ideally, therefore, such a test should be carried out in an area where the acidifying influence could only come from a local source.

K. MELLANBY (*Monks Wood Experimental Station, Abbots Ripton, Huntingdon PE17 2LS, U.K.*). Galloway is an area where we find a rich flora of foliose lichens, showing that the levels of sulphur dioxide in the air are very low. Thus dry deposition cannot be important in the acidification of these lakes.

R. W. BATTARBEE. The abundance of foliose lichens and the presence of acidified lakes in certain parts of Galloway is not necessarily paradoxical. Rainfall in south Scotland has an excess SO_4^{2-} concentration of 60–100 $\mu\text{eq l}^{-1}$ and the annual rainfall is > 1000 mm. Consequently, the total atmospheric SO_4^{2-} loading from wet deposition alone could account for the observed acidification. Lichens, therefore, may be good indicators of atmospheric SO_2 concentration but not of surface-water acidification.

A. D. BRADSHAW, F.R.S. (*University of Liverpool, Department of Botany, P.O. Box 147, Liverpool L69 3BX, U.K.*). While the evidence presented by Dr Battarbee argues very strongly for an acid rain effect, I would like to know what evidence there is for changes in land use in the catchment of these lakes. From 1900 to 1930 hill farming went into a serious decline in many parts of Britain. This would inevitably have led to increased growth of *Calluna* and other Ericaceous species at the expense of grass – which in turn would have altered run off characteristics of the catchment considerably.

P. F. CHESTER (*Central Electricity Research Laboratories Kelvin Avenue, Leatherhead, Surrey KT22 7SE, U.K.*). Dr Battarbee attributes the acidification of Loch Grannoch to industrial emissions of SO_2 . Between 1925 and 1982 your results indicate a 16-fold increase in acidity. Over this period

emissions of SO₂ have not increased by more than a factor 3. How exactly does Dr Battarbee reconcile this with his conclusion?

GWYNETH D. HOWELLS (*Central Electricity Research Laboratories, Kelvin Avenue, Leatherhead, Surrey, U.K.*). Could other changes in land use in Galloway have had some effect on the diatom record of lake sediment cores? Before afforestation, for example, in Loch Grannoch catchment, there was a grazing and moorland economy with a regular (and often irregular) programme of burning. In the light of Professor Rosenqvist's observations, wouldn't this have changed the biomass within the catchment, and so affected the quality of surface waters? What is known about the detailed history of land use–management of the lake catchments?

R. W. BATTARBEE. Except for post-1960 afforestation we have no specific data on changes in land use practice within the catchments of the lakes we have studied. However, taking the area as a whole, we have so far found it difficult to develop a land use hypothesis that might explain the inferred acidification trends at all sites in the way that Professor Bradshaw suggests. This is because: (i) our lithostratigraphic and pollen analytical data, except for the *ca.* 1600 A.D. change at the Round Loch of Glenhead, show no evidence of land-use change and, (ii) although there has been a decline in sheep numbers in the Galloway region since the turn of the century this has been owing to the reduction in grazing acreage as the uplands have been planted with conifers. The sheep density on the areas left unplanted may therefore have changed little. Moreover, although burning has been stopped in the Loch Grannoch catchment the catchments in the Merrick area are still burnt and grazed. In some cases e.g. Round Loch of Glenhead there is overburning, leading to the dominance of *Molinia caerulea* over *Calluna vulgaris*.

In addition, our clearest case of acidification so far is Loch Enoch, at high elevation and an extremely isolated loch where land management has been minimal and where carrying capacity and grazing intensity in the past are likely to have been very low. The situation for Loch Grannoch is less clear. For this site we argue primarily that acidification cannot be due to afforestation since afforestation post-dates acidification. This does not, however, discount an afforestation effect in the more recent past as an additional factor. Unfortunately, we cannot assess the importance of this factor at this site since ploughing before afforestation caused increased soil erosion and, we think, the inwash of diatoms from the catchment and the distortion of the sediment record. As for the pre-afforestation acidification referred to by Dr Chester, our data indicate a decrease in pH from *ca.* 5.6 to 4.4. This decrease may be somewhat exaggerated since the present pH range of the lake is 4.4 to 4.9 and the calibration model is poorly tested at values less than pH 4.5. Taking this and the standard error on our data of ± 0.3 pH unit into consideration the increase of acidity was substantial but probably less than 16-fold. In addition the response of a lake and its catchment to an acid load depends on the degree of buffering within the system, and there is no difficulty, as in the case of Loch Grannoch, in envisaging a small increase in acid load leading to a considerably larger response in H⁺ concentration if the lake is losing its alkalinity. Having said that, however, as Dr Howells suggests, the possibility of an additional land use effect cannot be ruled out at Loch Grannoch, since there is certainly more *Calluna* present in the areas left unplanted at this site than at the Round Loch and Loch Enoch catchments. Whether the *Calluna* developed in response to a cessation of such ameliorating factors as grazing, burning or liming perhaps 30 years or so before the beginning of afforestation can only be evaluated if detailed land use records for the period in question are available.

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A. KERR (*Friends of the Earth (Scotland)*, 53 George IV Bridge, Edinburgh EH1 1EJ, U.K.). I would like to thank Dr Battarbee for presenting an interesting paper and one which, it is hoped, will provide a useful tool in the historical study of lake acidification. I should be grateful if he would elaborate on five points.

(i) He presented a slide showing the 'rate of acidification per 10 years'. Is this an average of ten-yearly rates over the historical time period considered or the rate over the last ten years?

(ii) In outlining the scientific preference for diatom records and the usefulness of sediment core samples, the clarity of delineation and dating resolution were highlighted. Would Dr Battarbee kindly provide information on the annual rates of changes in acidification during the last 10, 20 and 30 years and indicate the mathematical form of changes in the rate?

(iii) Again, an advantage cited for the use of diatom records as a practical investigative tool was the possibility of comparing hypotheses regarding causes of acidification. What hypotheses have been advanced, which tested and what conclusions can be drawn?

(iv) In selecting diatom records as his mode of investigation Dr Battarbee rejected for very good reasons, the use of chemical, fisheries and flora and fauna records. Can he furthermore outline from his results the effect of increasing acidification on the total lake ecosystem?

(v) Finally, can he suggest what other work should be performed to pinpoint the causes of lake acidification and how his work in Britain may be extended in the light of the announcement of the £5 million research programme based in Scandinavia?

R. W. BATTARBEE. (i) and (ii) For simplicity, the rate of change depicted in the slide concerns the rate over the time period considered from the onset of acidification to the present at each site. More detailed calculation of the recent rate of acidification can be made for three sites Loch Dee, Round Loch of Glenhead and Loch Enoch. However, I should point out that these data are subject to the dating errors and pH reconstruction errors that I have already outlined. For Loch Dee the rate of acidification between *ca.* 1940 and *ca.* 1970 was *ca.* $0.013 \mu\text{eq H}^+ \text{l}^{-1} \text{a}^{-1}$, and from 1970 to 1980 the rate increased to *ca.* $0.08 \mu\text{eq H}^+ \text{l}^{-1} \text{a}^{-1}$. For the Round Loch of Glenhead the mean rate from *ca.* 1900 to *ca.* 1970 was *ca.* $0.16 \mu\text{eq H}^+ \text{l}^{-1} \text{a}^{-1}$ while at Loch Enoch the rate since *ca.* 1935 has been *ca.* $0.9 \mu\text{eq H}^+ \text{l}^{-1} \text{a}^{-1}$.

(iii) We have so far tested two hypotheses, that of long-term continuous acidification, and acidification as a result of afforestation. We have found both hypotheses to be inadequate to explain our observations.

(iv) It is difficult to use a diatom record that indicate past changes in other parts of the aquatic ecosystem. For example, although we know that diatom plankton declines rapidly at pH values below *ca.* 5.5 and declines can be seen in the sediment record we cannot conclude that planktonic primary productivity is diminished at these increased acidities since diatoms are replaced by planktonic taxa from other algal groups that often do not leave a clear sedimentary record. An examination of the Cladoceran and Chironomid record of lake sediments might allow some conclusions to be made concerning zooplankton and benthic invertebrate populations, but again interpretations would be qualitative and restricted to those groups and sub-groups that leave a fossil record. So the answer to this question is no.

(v) Our main concern must be to find situations in which various land use hypotheses can be properly tested and to assess the regional extent of acidification in the United Kingdom.