
The Causes of Lake Acidification, with Special Reference to the Role of Acid Deposition [and Discussion]

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The causes of lake acidification, with special reference to the role of acid deposition

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Acid surface waters ($\text{pH} < 5.5$) occur throughout western and northern Europe. The claim that many of these waters have been acidified in recent decades and that the acidification results from acid deposition has been well-substantiated by palaeolimnological studies. At almost all sites acidification post-dates 1800 A.D.; it is accompanied by increases in the concentration of trace metals and carbonaceous particles and the spatial pattern of acidified lakes coincides with areas of high acid deposition (greater than $0.5 \text{ g sulphur m}^{-2} \text{ a}^{-1}$). Very sensitive sites ($\text{Ca}^{2+} < 50 \mu\text{eq l}^{-1}$) in areas of low acid deposition are not acidified.

Palaeolimnological tests to evaluate the contribution of other factors suggest that leaching and paludification processes are important on a post-glacial time scale but imperceptible over the last 200 years, and that alterations to catchment burning and grazing regimes over this time scale have little or no effect. Only the afforestation of sensitive catchments in areas of high sulphur deposition appears to be significant, an effect attributed to the enhanced sulphur-scavenging efficiency of the forest canopy rather than to the direct effect of forest growth.

INTRODUCTION

Palaeolimnologists have shown that recent (post-1800) lake acidification is a common phenomenon in many parts of Europe and North America (Renberg & Hellberg 1982; Davis *et al.* 1983; Flower & Battarbee 1983; Charles 1985). They have used the lake sediment record to evaluate alternative explanations for the causes of lake acidification (Battarbee *et al.* 1985). In addition to the acid deposition hypothesis initially proposed by Odén (1968), these have included land-use change (Rosenqvist 1977, 1978; Krug & Frink 1983) and the long-term effect of natural processes (Pennington 1984).

Although there is now overwhelming palaeolimnological evidence for the importance of acid deposition as the main cause of recent acidification (Battarbee *et al.* 1988*a*) it is nevertheless important to assess the extent to which these other factors may have contributed to the overall result.

The Surface Water Acidification Project (SWAP) Palaeolimnology Programme has included two studies that describe the post-glacial history of acidified or sensitive lakes (Atkinson & Haworth and Renberg, this symposium) and several studies that were designed to assess the impact of land-use and land-management change (Patrick *et al.*, Birks *et al.*, Anderson & Korsman, Kreiser *et al.* and Renberg *et al.*, this symposium). This paper summarizes the results of these studies and describes the palaeolimnological evidence that directly supports the acid-deposition hypothesis.

LONG-TERM ACIDIFICATION

Long-term lake acidification has been a theme in palaeolimnological research for over 50 years. There are now many studies, reviewed by Battarbee (1984) and Charles & Norton (1986), mainly based on diatom analysis, showing that many presently acidic lakes were characterized in their early history by alkaline conditions. Some, however, have been acidic throughout their history (Jones *et al.* 1989). In all cases where acidification has taken place, the period of most rapid change occurs in the earliest stage following deglaciation (Whitehead *et al.* 1986; Atkinson & Haworth and Renberg, this symposium). Most authors, by using pollen analysis and sediment chemistry (Pennington *et al.* 1972; Digerfeldt 1972; Engstrom & Wright 1985) attribute this change to a combination of the removal of easily leached base cations from unweathered catchment soils and the development of acidic organic soils as a vegetation succession from arctic tundra to forest took place in the early Post-glacial period.

Although detailed trajectories in the latter part of the Post-glacial period vary from site to site depending on geology, soils, vegetation and early human influence, recent studies by using pH-inference techniques have shown that acidification during this period is very slow indeed (Renberg & Hellberg 1982; Atkinson & Haworth and Renberg, this symposium), with rates often less than 0.1 pH unit per 1000 years. In some cases, pH has not changed despite acidification and paludification of catchment soils (Jones *et al.* 1989). This mismatch between soil acidity and lake acidity in pre-industrial periods may be due to a combination of in-lake neutralizing processes, the inflow of relatively alkaline groundwater to lakes, the high partial pressure of CO₂ in soils and the lack of additional mobile anions to transfer acidity in surface soil horizons to watercourses.

LAND-USE AND LAND-MANAGEMENT CHANGE

Heathland and/or forest regeneration

The possibility that recent lake acidification is caused by changes in catchment land-use and management was proposed most explicitly by Rosenqvist (1977, 1978). He argued that a decline in farming intensity, especially in burning and grazing, would lead to regeneration of heathland and forest vegetation causing an increase in the accumulation of acid humus in soils and the release of protons into surface waters by cation exchange processes.

There have been many attempts to test this hypothesis by using palaeolimnological techniques, either by careful choice of sites or by using past analogues. It can be argued that the hypothesis cannot have universal validity as recent lake acidification has occurred in regions where there has been either no change or, in some cases, an increase in farming intensity (Timberlid 1980; Battarbee *et al.* 1985; Patrick *et al.*, this symposium). In addition, sites with only very small catchments in relation to their surface area (Birks *et al.*, this symposium) have been recently acidified as have montane sites above the limit of summer sheep grazing (Patrick *et al.* 1989, this symposium). In all these cases there is a clear evidence of atmospheric contamination.

At sites where a decrease in burning and grazing has occurred in the catchments of acidified lakes, it is sometimes possible to test the hypothesis by comparing the timing of land-use change with the timing of acidification. For example at Llyn y Bi in Wales Fritz *et al.* (1990) showed

that acidification began in the late-nineteenth century well before the landowner's decision to stop *Calluna* burning in the 1930s.

Although these examples illustrate cases where land-use changes cannot have caused lake acidification, none examine the efficacy of the mechanism itself. Where there has been a decrease in burning and grazing and an increase in acid deposition over approximately similar timescales, the effect of the former can be assessed by using analogues in space or in time. A space analogue would involve a study of land-use effects in sensitive systems in an area of very low acid deposition (less than $0.5 \text{ g S m}^{-2} \text{ a}^{-1}$). There have been no palaeolimnological studies of this kind, but it can be observed that very acid clear-water lakes ($\text{pH} < 5.5$) do not occur in areas of very low acid deposition despite well-documented histories of land-use change.

An alternative approach is to use past analogues, employing palaeoecological techniques to examine the relation between vegetation and land-use changes, inferred from pollen analysis, and lake response, inferred from diatom analysis, at times well before the increase in acid deposition (pre 1800 A.D.). Although detailed land-use inferences are more conjectural with past analogues the advantage is that lakes that are currently acidified, and therefore of proven sensitivity, can be used. In this way Jones *et al.* (1986) showed that a major shift from woodland to peatland in the catchment of the Round Loch of Glenhead approximately 5000 years BP caused little change in lake pH. Renberg *et al.* (this symposium) did pollen and diatom analysis of eight sensitive sites in Sweden and showed that there was little or no water pH response to the spread of *Picea* forest 2500–3000 years ago, and Anderson & Korsman (this symposium) similarly showed that no acidification followed the decline in farming that took place in an area of northern Sweden during Iron Age times.

The heathland and forest regeneration hypothesis is irrelevant at many sites and where and when an appropriate land-use change has taken place there is no related evidence for significant water acidification.

Conifer afforestation in the United Kingdom

In this discussion it is appropriate to separate afforestation of moorland and its effects from the land-use changes described above because afforestation processes involve an intensification of catchment interference and management, often including deep drainage of soils, hydrological manipulation, use of fertilizers and pesticides and closely-spaced planting of trees. Moreover it has been observed that forests are efficient scavengers of dry sulphur deposition (D. Fowler, personal communication) and that streams with afforested catchments have more acidic water and poorer fisheries than adjacent streams with moorland catchments (Harriman & Morrison 1982; Stoner & Gee 1985).

Diatom analysis of sediments from very sensitive lakes ($\text{Ca}^{2+} < 50 \text{ } \mu\text{eq l}^{-1}$) have shown that acidification invariably precedes afforestation (Flower & Battarbee 1983; Flower *et al.* 1987), but the study of Kreiser *et al.* (this symposium) comparing the diatom histories of afforested and moorland lakes at higher Ca^{2+} levels ($80 \text{ } \mu\text{eq l}^{-1}$) indicates that afforestation led to a rapid acceleration in acidification at a time when the moorland control site (Loch Tinker) changed little.

To evaluate whether the forest effect was caused by forest growth factors or by scavenging Kreiser *et al.* (this symposium) then compared a sensitive afforested and moorland site in an area to the north with lower acid deposition. Although it was not possible to separate clearly the forest effect from the direct effect of acid deposition, as the moorland site was also slightly

acidified, the results show that the afforested site in the north (Loch Doilet) was much less acidified than the afforested site (Loch Chon) in the south, despite the presence of a more mature forest at the former site. These data require replication but nevertheless support the scavenging hypothesis as the main mechanism to explain the 'forest effect.'

ACID DEPOSITION

A major effort has been made to consider the role of factors other than acid deposition as a cause of lake acidification. As these factors have been shown to be of very minor importance it is now necessary to assess how well acid deposition can explain recent lake acidification. Potential cause-effect relations are easier to disprove than to prove, but the palaeolimnological evidence supporting the acid deposition hypothesis is substantial. It can be demonstrated that recent lake acidification is almost perfectly correlated with acid deposition surrogates in both time and space; we can show that a dose-response relation exists between sulphur loading and the sensitivity of lakes to acidification.

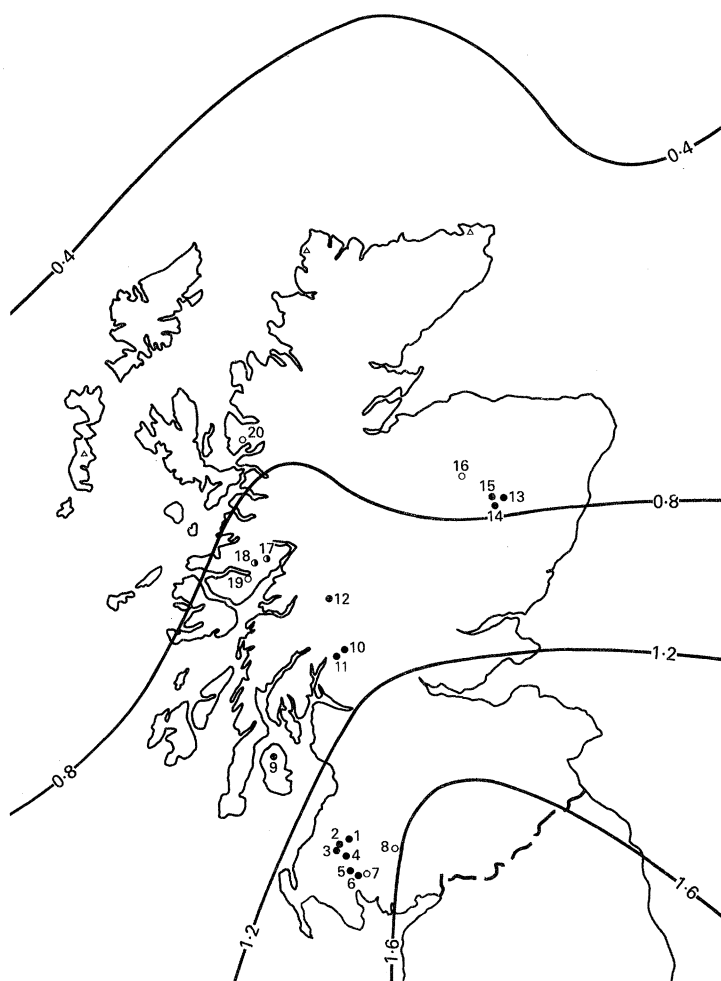


FIGURE 1. Change of pH at core sites in relation to the modelled deposition of S ($\text{g m}^{-2} \text{a}^{-1}$) for Scotland (from Derwent *et al.* (1988)); (●), pH decrease 0.4–1.2; (◐), pH decrease < 0.4; (○), no decrease; (Δ), work in progress.

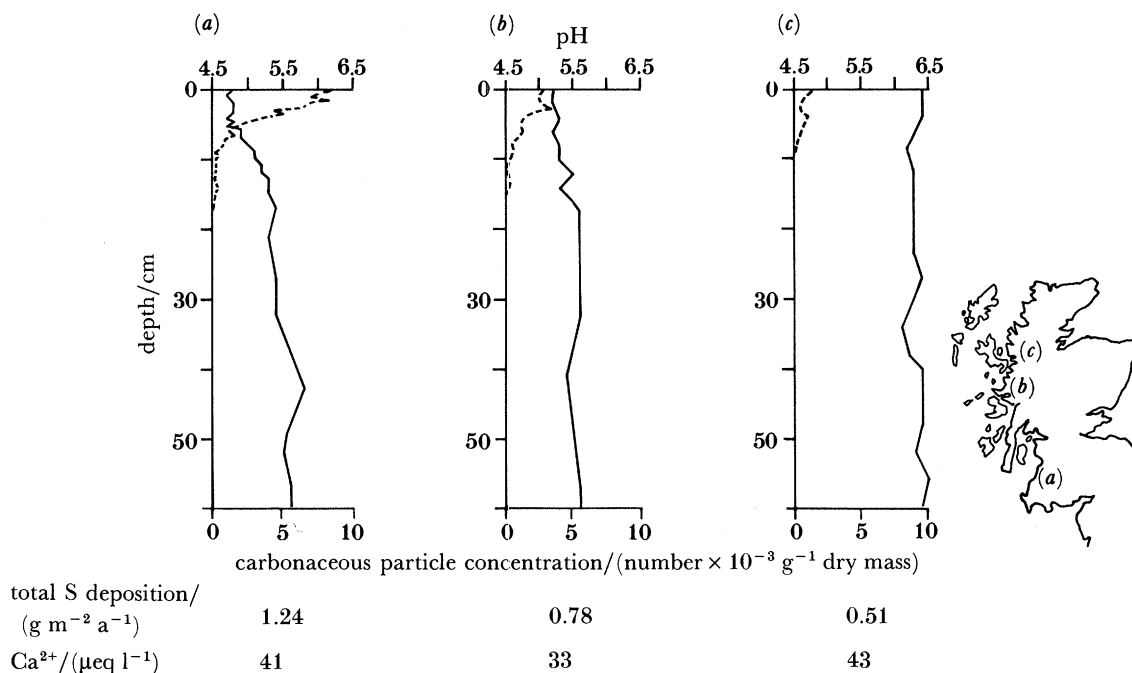


FIGURE 2. Comparison of pH trends (—) and carbonaceous particle profiles (---) for three sites with moorland catchments from high to low S deposition and approximately constant lake-water Ca^{2+} values. (a) Round Loch of Glenhead, (b) Lochan Dubh, (c) Loch Corrie nan Arr.

These relations can be illustrated by data from Scotland where there is a strong sulphur deposition gradient from south to north. Figure 1 shows the distribution of sites for which we have diatom-based pH reconstructions and indicates the extent of estimated pH decline at each site. At each acidified site the first point of pH decrease is never before 1800 (cf. Battarbee *et al.* 1988a) and no acidified sites have been found in the areas of low acid deposition.

If lake sensitivity is kept constant, but sulphur loading is varied, it can also be shown (figure 2) that the degree of pH decline is correlated with the degree of atmospheric contamination. The three sites shown in figure 2 are all extremely sensitive to acidification with Ca^{2+} between 30 and 50 $\mu\text{eq l}^{-1}$. The Round Loch of Glenhead lies in an area of high sulphur deposition ($1.2 \text{ g m}^{-2} \text{ a}^{-1}$), has undergone a pH decline of approximately 1 pH unit and is strongly contaminated by carbonaceous particles. At Lochan Dubh in an area of medium sulphur deposition ($0.8 \text{ g m}^{-2} \text{ a}^{-1}$) acidification and atmospheric contamination are correspondingly lower, and at Loch Corrie nan Arr there is only slight atmospheric contamination and no evidence of a pH decline.

If the data-set is enlarged to cover all sites that have diatom-based pH reconstructions (cf. Battarbee *et al.* 1988a), the acidification status of any one site can be determined by using this relation of sensitivity (as Ca^{2+} concentration) and loading (as sulphur deposition). Figure 3 shows data for moorland sites where a line describing a Ca^{2+} :sulphur deposition ratio of between approximately 50 and 70:1, effectively separates acidified from non-acidified sites. All sites where the diatom data indicate questionable or very slight acidification fall within the bounds of the ratio.

This empirically derived relation has not yet been independently validated, and it has not been modified to include sites with afforested catchments, where sulphur deposition may be

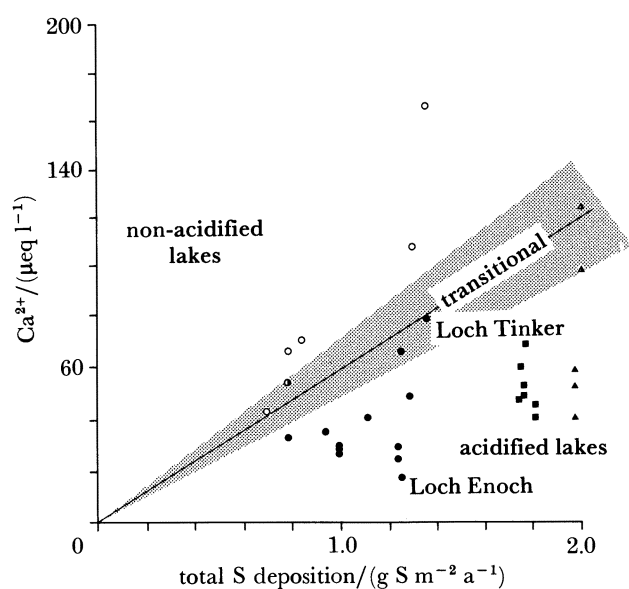


FIGURE 3. The relation between lake-water Ca^{2+} concentration and total S deposition for core sites in the United Kingdom. The acidification status of each lake is shown and the data can be used to derive the critical S load for each site. Scottish sites are shown in figure 1, other sites and all site data are presented in Battarbee *et al.* 1988a; ($\circ\bullet$), Scottish sites; ($\Delta\Delta\Delta$), English sites; (\blacksquare), Welsh sites; ($\bullet\blacktriangle$), pH decrease 0.4–1.2; ($\bullet\Delta$), pH decrease < 0.4; ($\circ\Delta$), no decrease.

enhanced, but it does underline the importance of sulphur deposition as the main cause of lake acidification.

The ratio can also be regarded as the 'critical ratio' for the acidification of moorland sites as it allows the critical sulphur load for any one site to be calculated. On this basis the critical load for Loch Enoch, assuming no change in Ca as S is reduced, is about $0.3 \text{ g S m}^{-2} \text{ a}^{-1}$ (75% lower than the present sulphur deposition).

The ultimate dose–response test of this relation is the recovery of lakes following a reduction in deposition. Already there is evidence to suggest that following the decline in U.K. and European emissions of SO_2 in the last decade, lake acidification has stopped and in some cases is reversing (Battarbee *et al.* 1988b).

I thank all members of the Palaeoecology Research Unit at University College London and other colleagues in the SWAP Palaeolimnology Programme for providing the data and contributing to the ideas in this paper. I am also grateful to Ron Harriman, Ron West and David Tervet for chemical analyses; sulphur deposition data were kindly provided by Dick Derwent.

REFERENCES

- Battarbee, R. W. 1984 Diatom analysis and the acidification of lakes. *Phil. Trans. R. Soc. Lond. B* **305**, 451–477.
 Battarbee, R. W., Flower, R. J., Stevenson, A. C. & Rippey, B. 1990 Lake acidification in Galloway: a palaeoecological test of competing hypotheses. *Nature, Lond.* **314**, 350–352.
 Battarbee, R. W., Anderson, N. J., Appleby, P. G., Flower, R. J., Fritz, S. C., Haworth, E. Y., Higgitt, S., Jones, V. J., Kreiser, A., Munro, M. A. R., Natkanski, J., Oldfield, F., Patrick, S. T., Richardson, N. G., Rippey, B. & Stevenson, A. C. 1988a *Lake acidification in the United Kingdom 1800–1986: evidence from analysis of lake sediments*. London: Ensis Publishing.

- Battarbee, R. W., Flower, R. J., Stevenson, A. C., Jones, V. J., Harriman, R. & Appleby, P. G. 1988*b* Diatom and chemical evidence for reversibility of acidification of Scottish lochs. *Nature, Lond.* **332**, 530–532.
- Charles, D. F. 1984 Recent history of Big Moose Lake (Adirondack Mts, N.Y., U.S.A.) inferred from sediment diatom assemblages. *Verh. internat. Verein. Limnol.* **22**, 559–566.
- Charles, D. F. & Norton, S. A. 1986 Paleolimnological trends for evidence for trends in atmospheric deposition of acids and metals. In *Atmospheric deposition: historical trends and spatial patterns*, ch. 9, pp. 335–434. Washington, D.C.: National Academy Press.
- Davis, R. B., Norton, S. A., Hess, C. T. & Brakke, D. F. 1983 Paleolimnological reconstruction of the effects of atmospheric deposition of acids and heavy metals on the chemistry and biology of lakes in New England and Norway. *Hydrobiologia* **103**, 113–124.
- Derwent, R. G., Hopper, S. & Metcalfe, S. E. 1988 *Computer modelling studies of the origins of the acidity deposited in Scotland*. Harwell: Atomic Energy Research Establishment, report no. 13328.
- Digerfeldt, G. 1972 The post-glacial development of Lake Trummen. Regional vegetation history, water-level changes, and palaeolimnology. *Fol. Limnol. Scand.* **16**, 1–104.
- Engstrom, D. R. & Wright, H. E. 1985 Chemical stratigraphy of lake sediments as a record of environmental change. In *Lake sediments and environmental history* (ed. E. Y. Haworth & J. W. G. Lund), pp. 1–68. University of Leicester Press.
- Flower, R. J. & Battarbee, R. W. 1983 Diatom evidence for recent acidification of two Scottish lochs. *Nature, Lond.* **20**, 130–133.
- Flower, R. J., Battarbee, R. W. & Appleby, P. G. 1987 The recent palaeolimnology of acid lakes in Galloway, south-west Scotland: diatom analysis, pH trends and the role of afforestation. *J. Ecol.* **75**, 797–824.
- Fritz, S. C., Kreiser, A. M., Appleby, P. G. & Battarbee, R. W. 1990 Recent acidification of upland lakes in North Wales: palaeolimnological evidence. In *Acid waters in Wales* (ed. R. Edwards). Cardiff: Welsh. (In the press.)
- Harriman, R. & Morrison, B. R. S. 1982 The ecology of streams draining afforested and non-forested catchments in an area of central Scotland subject to acid precipitation. *Hydrobiologia* **88**, 251–263.
- Jones, V. J., Stevenson, A. C. & Battarbee, R. W. 1986 Lake acidification and the land-use hypothesis: a mid-post-glacial analogue. *Nature, Lond.* **322**, 157–158.
- Jones, V. J., Stevenson, A. C. & Battarbee, R. W. 1989 Acidification of lakes in Galloway, Southwest Scotland: a diatom and pollen study of the post-glacial history of the Round Loch of Glenhead. *J. Ecol.* **77**, 1–23.
- Krug, E. C. & Frink, C. R. 1983 Acid rain on acid soil, a new perspective. *Science, Wash.* **221**, 520–525.
- Odén, S. 1968 *The acidification of air precipitation and its consequences in the natural environment*. Energy Committee Bulletin, 1. Stockholm: Swedish Natural Sciences Research Council.
- Patrick, S. T., Flower, R. J., Appleby, P. G., Oldfield, F., Rippey, B., Stevenson, A. C., Darley, J., Raven, P. J. & Battarbee, R. W. 1989 Palaeoecological evaluation of the recent acidification of Lochnagar, Scotland. Palaeoecology Research Unit, University College London, Research paper no. 34.
- Pennington, W. 1984 Long-term natural acidification of upland sites in Cumbria: evidence from post-glacial lake sediments. *Freshwat. Biol. Ass. Ann. Rep.* **52**, 28–46.
- Pennington, W., Haworth, E. Y., Bonny, A. P. & Lishman, J. P. 1972 Lake sediments in northern Scotland. *Phil. Trans. R. Soc. Lond. B* **264**, 191–294.
- Renberg, I. & Hellberg, T. 1982 The pH history of lakes in southwestern Sweden, as calculated from the subfossil diatom flora of the sediments. *Ambio* **11**, 30–33.
- Rosenqvist, I. T. 1977 *Acid soil – acid water* Oslo: Ingeniørforlaget.
- Rosenqvist, I. T. 1978 Alternative sources for acidification of river water in Norway. *Sci. Tot. Envir.* **10**, 39–49.
- Stoner, J. H. & Gee, A. S. 1985 The effects of forestry on water quality and fish in Welsh rivers and lakes. *J. Inst. Wat. Engineers Scientists* **39**, 125–157.
- Whitehead, D. R., Charles, D. F., Reed, S. E., Jackson, S. T. & Sheehan, M. C. 1986 Late-glacial and Holocene acidity changes in Adirondack (N.Y.) lakes. In *Diatoms and lake acidity* (ed. J. P. Smol, R. W. Battarbee, R. B. Davis, & J. Meriläinen), pp. 251–274. Dordrecht: Dr. W. Junk

Discussion

G. HOWELLS (*University of Cambridge, Cambridge, U.K.*). Is the interpretation by Dr Battarbee of the acidifying effect of forestry at Loch Fleet correct? Ploughing and planting at Loch Fleet took place in 1961–62 and acidification did not occur until the mid 1970s. Further, the area afforested covers only 10% of the catchment, yielding significantly less than this to the runoff, as evapotranspiration from the forest is significantly greater than that from the moorland. The acidity of runoff from the forest, before liming treatment in 1986, was only slightly less than that from the rest (90%) of the catchment and so this contribution could not be responsible for the shift in pH of the loch.

R. W. BATTARBEE. Loch Fleet and Loch Chon are the only two afforested sites we have studied where the main period of acidification follows afforestation. For Loch Chon, which has a largely afforested catchment, we attribute the acceleration in acidification to enhanced scavenging of S by the canopy. I did not give an explanation for the acidification of Loch Fleet. I agree that an insufficient proportion of the catchment has been afforested at Loch Fleet for this interpretation to be valid. We need an additional mechanism. We know that the groundwater in parts of the catchment is extremely Ca^{2+} rich and that a layer of post-1960 peaty sediment up to 1.2 m thick now covers the main bed of the lake, so we have proposed that peat inwash that followed the deep ploughing of the catchment (before planting) has sealed off the lake-water from neutralising contact with sediments. With Dr W. M. Edmunds from BGS we are presently testing this hypothesis by examining pore-water chemistry of cores before and after the inwash period.

R. A. SKEFFINGTON. (*National Power Technology and Environmental Centre, Leatherhead, Surrey, KT22 7SE, U.K.*). The evidence seems overwhelming that acid deposition has been the major factor in recent lake acidification. But this does not preclude land-use change having a contributory effect in places. Inferences that the effect of afforestation on water quality is mediated only through pollutant capture seem to be being made on the basis of only one site in the 'afforested, low deposition' category. This is surely unsatisfactory and I wonder whether further studies are planned? Our modelling work indicates that afforestation on poor sites should affect water quality simply through cation uptake. I would not like it to be thought that one conclusion of the SWAP palaeoecology programme was that lake catchments could be misused with impunity, acid deposition being the only factor that can affect water quality.

R. W. BATTARBEE. We would like to replicate our Loch Doilet study at sites further north, but unfortunately (or perhaps fortunately!) there are very few afforested sites in Scotland with low Ca^{2+} waters in these areas of low acid deposition in Scotland. Cation uptake may have a role to play but on the basis of the Loch Doilet study, and from observations of water chemistry of lakes with forested catchments in low S deposition areas of Norway, it must be very minor. This does not imply that afforestation has little effect on surface water quality and aquatic ecosystems. On the contrary, we were only concerned with acidification in this study, but we have other data that demonstrate problems of soil erosion and nutrient enrichment associated with afforestation.

A. D. BRADSHAW, F.R.S. (*Department of Environmental and Evolutionary Biology, University of Liverpool, Liverpool L69 3BX, U.K.*) Our memories are often made very short by the pressure of new problems and new experiences. Nowhere is this more true than in science, where ideas can become old in five years. Yet it is those ideas, not long ago so very new, that are the steps to the present.

I was brought up to a tradition of paleoecology – it was called pollen analysis in those days – as an esoteric historical discipline of appeal to an academic few.

So it was, also, with palaeolimnology, until about ten years ago. Then, suddenly, it began to provide some very interesting evidence for the effects of acid rain, an environment problem that was unclear and disbelieved by many, especially in the U.K. This was because the direct evidence, from lake acidification, was based on poor historical data and rather inadequate comparisons of different lakes.

Then, suddenly, I clearly remember, our ideas changed, because palaeolimnology gave clear evidence of major changes in lake quality in recent times that could only be due to acid deposition (Battarbee 1984). Not only were the changes able to be correlated in time with developing industrial activity, but other pieces of evidence could be found, such as carbonaceous particles, to make the arguments incontrovertible. The crucial point was that this evidence was, then, not only enough to persuade ecologists that acid rain was a problem, but politicians also. The need for control of sulphur emissions suddenly became clear.

As a result of all this, palaeolimnology has become an important, practically oriented, area of work, with a great deal of financial support. This has, however, not destroyed palaeolimnology as a scientific discipline, as some academics might have thought, but has strengthened it. The many excellent papers of this meeting make this very clear. There is now a very healthy two-way exchange between the demands of studying acid rain and the science of palaeolimnology, which shows well the value of combining science and practice so that each can feed on the other.

This pleases me greatly, as it is something that has been argued for on previous occasions (Dunnett 1980; Bradshaw 1982). I would like to compliment not only the organizers of this meeting and the speakers, but also the many other scientists, who, together, have taken palaeolimnology into new rewarding areas.

References

- Battarbee, R. W. 1984 Diatom analysis and the acidification of lakes. *Phil. Trans. R. Soc. Lond. B* **305**, 415–477.
Bradshaw, A. D. 1982 Presidential viewpoint: achieving useful ecology. *Bull. Br. Ecol. Soc.* **13**, 112–114.
Dunnett, G. M. 1980 Presidential viewpoint. *Bull. Br. Ecol. Soc.* **11**, 94–96.