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The recent palaeolimnology of two sites with contrasting acid-deposition histories

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A palaeoecological comparison is made between geologically sensitive sites chosen from an area of low sulphur deposition (Lochan Dubh) and area of high sulphur deposition (Round Loch of Glenhead). Pre-industrial (pre-1800) acidities of the lakes were similar but the pH of the Round Loch of Glenhead has subsequently dropped by over 0.5 of a pH unit whereas the pH of Lochan Dubh has only decreased slightly. The record of atmospheric contamination confirms that the Round Loch of Glenhead is a more heavily polluted site than Lochan Dubh. The increased degree of lake acidification and higher levels of atmospheric contamination at the Round Loch of Glenhead are correlated with the greater sulphur deposition levels at this site.

Introduction

Palaeoecological evidence based principally on diatom analysis (Battarbee 1984) suggests that many lakes in the British Isles have rapidly acidified over about the last 150 years (Flower & Battarbee 1983; Battarbee *et al.* 1988). Possible reasons for lake acidification have been evaluated by using palaeolimnological techniques, which include land-use change (Jones *et al.* 1986; Patrick *et al.*, this symposium), afforestation (Flower *et al.* 1987) and long-term soil acidification (Jones *et al.* 1989). These factors cannot be used to explain present-day acidities at the sites investigated so far, although afforestation may enhance lake acidification at certain sites (Kreiser *et al.*, this symposium). However, there is strong evidence for atmospheric contamination of acidified lakes by trace metals (Rippey, this symposium), spheroidal carbonaceous particles (Wik & Natkanski, this symposium) and magnetic minerals (Oldfield & Richardson, this symposium). This evidence of increasing atmospheric contamination, from the mid-19th century onwards at many sites, is correlated with an increase in national sulphur emissions (Barrett *et al.* 1983) and at sensitive sites (cf. Edmunds & Kinniburgh 1986) with a trend of lake acidification. For the palaeolimnological programme of the Surface Water Acidification Project (SWAP), sites were carefully chosen to evaluate the relation between the history of atmospheric contamination, and the degree of acidification and ecological change.

Site descriptions and methods

Two lakes are examined in this study. The Round Loch of Glenhead is situated in the Galloways Hills, southwest Scotland, an area currently subjected to high levels ($1.24\ g\ S\ m^{-2}\ a^{-1}$) of sulphur deposition. Lochan Dubh forms part of the Loch Doilet catchment in the Sunart area of central western Scotland, and lies about 200 km north of the Round Loch, where levels of atmospheric deposition are considerably lower ($0.78\ g\ S\ m^{-2}\ a^{-1}$).

The two sites have Ca^{2+} levels less than 50 μeq l^{-1} (see table in Battarbee & Renberg, this symposium) indicating that both are very susceptible to acidification (Battarbee *et al.* 1988). However, the Round Loch of Glenhead has a lower pH, a lower alkalinity and over twice the concentration of excess non-marine SO_4^{2-} compared with Lochan Dubh. This difference in non-marine SO_4^{2-} is likely to be mainly attributable to atmospheric sulphate deposition (cf. Gorham *et al.* 1986). The relatively high Na^+ and Cl^- levels at the sites reflects a substantial marine influence.

The lakes are both of glacial origin situated on granite (Round Loch of Glenhead) and quartzite (Lochan Dubh) bedrock, both of which have a low weathering rate. Catchment vegetation consists of moorland communities dominated by *Molinia caerulea* and *Calluna vulgaris* overlying blanket peat. Neither site shows any pollen or documentary evidence for recent land-use or land-management change (Patrick *et al.*, this symposium).

Piston cores were obtained from the Round Loch of Glenhead (core RLGH3) in 1984 and from Lochan Dubh (core LOD2) in 1986. Analytical techniques and diatom analysis are outlined in Stevenson *et al.* (1987). Diatom nomenclature follows the SWAP protocol (Munro *et al.*, this symposium), and pH reconstruction follows Birks *et al.* (this symposium).

Results

(*a*) *Diatom analysis; floristic changes and* pH *reconstruction*

In the Round Loch of Glenhead core, the diatom flora below 20 cm is dominated by indifferent (= circumneutral) taxa, for example, *Achnanthes minutissima*, and acidophilous taxa, for example, *Eunotia incisa* (figure 1). At about 20 cm (*ca.* 1850) the percentages of indifferent species start to decline, and there is an increase in acidophilous forms. At 9 cm (*ca.* 1900) the acidobiontic taxa, for example, *Tabellaria quadriseptata* increase and the values of the indifferent species *Achnanthes minutissima*, *Brachysira vitrea* and *Cymbella lunata* fall sharply. At 5 cm (*ca.* 1953) the acidobiontic species *Tabellaria binalis* becomes important for the first time, and the sum of acidobiontic taxa makes up more than 20 % of the diatom assemblage. Results of diatom-based pH reconstruction suggest that the lake was acid (pH 5.4–5.6) before 1850. A decline of about 0.3 pH units occurred between 1850 and 1900, and there was a further decrease in pH from 1900 to the present day (from pH 5.0 to 4.7). The reconstructed pH is 4.9 at 0–0.5 cm, giving a relatively good agreement with the present pH of the lake water (pH 4.7).

Results of diatom analysis from levels below 80 cm in core RLGH3 indicate that the Round Loch of Glenhead was acid during the entire post-glacial period (Jones *et al.* 1989; Birks *et al.*,

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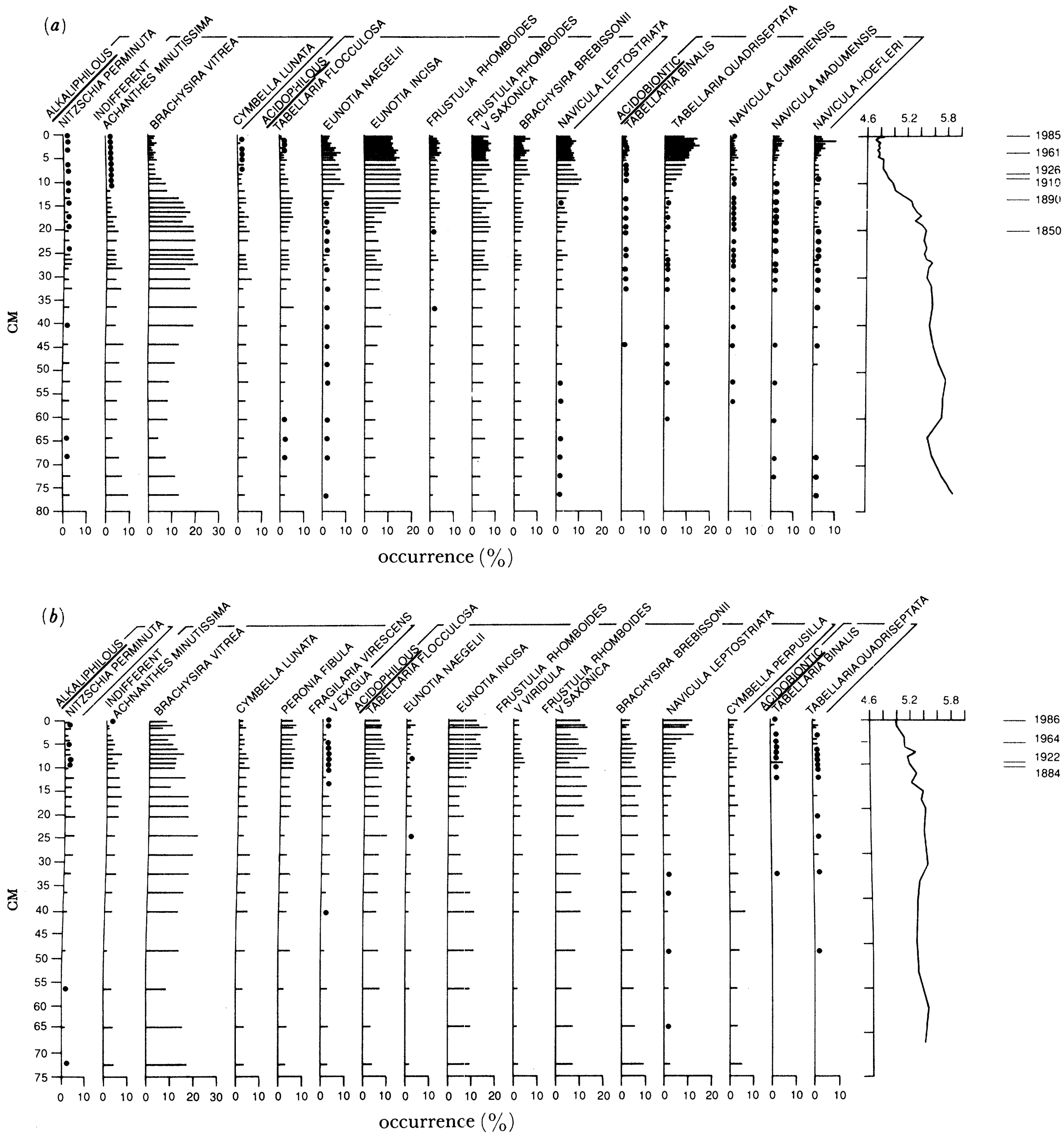

FIGURE 1. Diatom frequency diagrams of selected species (expressed as percentage of total diatoms) from (*a*) the Round Loch of Glenhead and (*b*) Lochan Dubh. The order of species left to right represents a gradient of increasing tolerance to low pH; pH reconstruction is calculated by using a weighted averaging technique. The standard error of prediction is based on bootstrap estimation (see Birks *et al.*, this symposium) and is between 0.31–0.32 of a pH unit for both of these sites. Dates are calculated from ^{210}Pb dating.

this symposium). There is little evidence of acidification from 10000 BP to the mid-19th century despite the effects of extensive catchment paludification and long-term soil leaching. Indeed, reconstructed pH never fell below 5.0 until after 1900. Birks *et al.* (this symposium) have also shown that the rate of pH change from 1874–1931 is significantly different from the rate of change over the rest of the post-glacial period.

At Lochan Dubh, the major floristic changes occur between 18 cm and 14 cm, where the proportions of the acidophilous taxa *Navicula leptostriata* and *Eunotia incisa* increase and the indifferent taxa *Brachysira vitrea*, *Achnanthes minutissima* and *Fragilaria virescens* var. *exigua* decline. Although species changes do occur, many taxa remain at stable levels, for example, *Frustulia rhomboides* var. *saxonica*, *Tabellaria flocculosa* and *Peronia fibula*. Below 14 cm (early 19th century) the reconstructed pH lies between 5.3 and 5.4; a slight acidification occurs above this level that continues to the top of the core giving a diatom inferred pH of 5.0. The present measured pH is 5.55.

(*b*) *Other biological indicators of lake water quality*

Chironomid remains in the Round Loch of Glenhead showed decreased diversity, productivity and survival rates after about 1850 that can clearly be related to acidification (Brodin, this symposium). There is also a decrease in the diversity of the cladoceran remains above 10 cm (*ca.* 1913), associated with an increase in the acid-tolerant species *Bosmina longispina*. At Lochan Dubh the absence of acid-sensitive Cladocera (Nilssen & Sandøy, this symposium), for example, *Daphnia longispina*, and the presence of the acid tolerant species *Acantholeberis curvirostris* throughout the core suggests that the lake has always been rather acid (pH < 5.5). However, there is no cladoceran evidence for significant changes in the pH at this site. Both the modern Cladocera and diatom flora from Lochan Dubh suggest more acid conditions than indicated by the measured pH. This may be because of insufficient water chemistry data at this site.

(*c*) *Evidence for atmospheric contamination*

Spheroidal carbonaceous particles (SCP) show similar patterns of contamination in the Round Loch of Glenhead and Lochan Dubh (figure 2), with low pre-20th century values and increasing concentrations from about 1900 onwards. However, in the Round Loch of Glenhead values are over ten-times higher.

Magnetic mineral concentration (saturation isothermal remanent magnetization (SIRM)) values at the Round Loch of Glenhead are low before about 1900 (12 cm), and then increase, probably reflecting atmospheric deposition of magnetic material. At Lochan Dubh there is a small rise in SIRM from about 1900 (9 cm), with a further increase after 1964 (4 cm). The similarity of these trends to the SCP profiles provides good evidence that SIRM is recording the history of fossil-fuel combustion.

At the Round Loch of Glenhead, the first major change in trace metal chemistry occurs at 40 cm where Pb concentrations increase (figure 2). This may possibly be related to local industrial sources. There is a further increase in Pb above 20 cm (*ca.* 1850), where concentrations of Zn also rise. These recent changes in the total concentrations of Pb and Zn cannot be explained in terms of catchment changes and are due to increased atmospheric inputs. The recent (*ca.* 1950) drop in Zn and Pb at the Round Loch of Glenhead is partly due to a decreased contamination flux from the atmosphere, together with the additional effect of a decrease in Zn sedimentary efficiency with falling pH. At Lochan Dubh there is little evidence of any atmospheric contamination by Pb. However, Zn concentrations rise above background levels at about 10 cm (1884), but maximum concentrations are much lower than those found at the Round Loch of Glenhead.

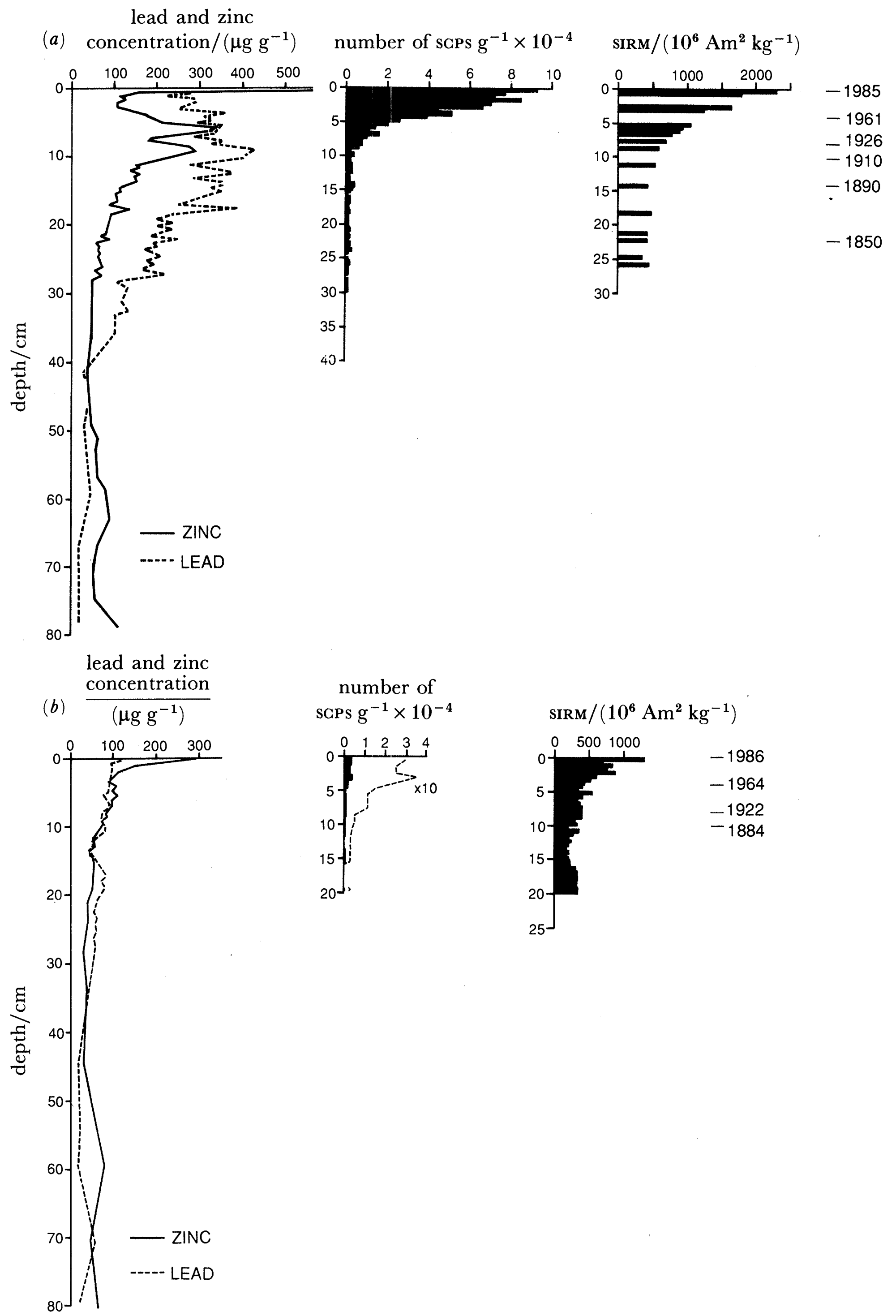

FIGURE 2. Degree of atmospheric contamination. Trace metal, spheroidal carbonaceous particle (SCP) and saturation isothermal remanent magnetisation (SIRM) profiles from (*a*) the Round Loch of Glenhead and (*b*) Lochan Dubh, plotted against depth. Dates are calculated from ^{210}Pb dating.

Discussion

Although distinct floristic changes occur in the diatom flora at both sites, at the Round Loch of Glenhead this involves a substantial increase in both acidophilous and acidobiontic forms, whereas at Lochan Dubh there is only an increase in acidophilous forms. Diatom based pH reconstruction shows that a greater degree of acidification has occurred at the Round Loch of Glenhead; these results are reinforced by the cladoceran data.

At the Round Loch of Glenhead, the most rapid pH change experienced in the Postglacial period began in the mid-19th century. Concentrations of Zn and Pb also began to rise at this time. After about 1900 a further decrease in pH occurred, and the concentrations of SCP and magnetic minerals increased sharply. At Lochan Dubh, a much weaker acidification occurred with less pronounced increases in Zn, SCP and magnetic minerals.

The onset of lake acidification is better correlated with the start of atmospheric contamination indicated by the trace metal profiles than by either the SCP or magnetic concentration profiles. However, the later 20th century increase in atmospheric contamination is shown by all the three atmospheric contamination techniques. This could be because of differences in resolution between the techniques or, alternatively, the SCP and magnetic mineral trends may reflect a shift in the relative importance of different contamination sources.

Whereas both sites show a record of atmospheric contamination of trace metals, spheroidal carbonaceous particles and magnetic minerals, levels of contamination are consistently lower at Lochan Dubh than the Round Loch of Glenhead.

The observation that Lochan Dubh, a sensitive site situated in an area of low sulphur deposition, is less contaminated and less acidified than the Round Loch of Glenhead, a similarly sensitive site situated in an area of high sulphur deposition, supports the argument that acidification is caused by atmospheric contamination.

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