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Palaeoecological evidence for the timing and causes of Lake acidification in Galloway, south-west Scotland.

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CONTENTS

ABSTRACT

During the contract period sediment cores from six lakes, three with non-afforested and three with partially afforested catchments, were analysed to enable lake water acidity (pH) to be reconstructed over approximately the past 200 years. The sites are all situated on granitic rocks in the Galloway Hills of S.W. Scotland. The results of diatom analysis show that five of the six lakes have become considerably more acid over this time period and that the acidification process is mainly independent of catchment afforestation effects. ^{210}Pb dating of the sediment shows that there has been little change in sediment accumulation rates at sites with non-afforested catchments, but there has been significant increases in accumulation rate at afforested sites associated with erosion caused by pre-planting catchment ploughing. Pollen analysis indicates that no major vegetation change has occurred at the non-afforested sites whilst trace metal analysis of L. Enoch (not funded by this contract) shows elevated levels of Pb, Cu, and Zn in the upper, post 1800 sediments. It is concluded that acid precipitation is the most likely cause of acidification at these sites.

INTRODUCTION

We have employed diatom analysis, pollen analysis, geochemical analysis, and radiochemical dating of lake sediment cores from Galloway, S.W. Scotland, to consider:

(a) whether these lakes have become more acid within the last 200 years or so,

(b) when acidification began,

(c) how the rate of acidification has changed over the last 200 years,

(d) what factors might have caused acidification at each site.

We have used the results to evaluate four alternative hypotheses, namely that acidification is the result of:

(a) the recent (post-war) conifer afforestation of lake catchments,

(b) a decline in upland sheep grazing and moorland burning,

(c) slow, long-term paludification of catchment soils over the post-glacial period (last 10,000 years),

(d) an increase in the acidity of precipitation since the beginning of the Industrial Revolution (from about 1800).

STUDY SITES AND METHODS OF INVESTIGATION

Six lakes, three with non-afforested catchments (Round Loch of Glenhead, L. Enoch, and L. Valley) and three with partially afforested catchments (L. Dee, L. Grannoch, and L. Skerrow) were chosen for study (Table 1, Figs. 1-7). Short sediment cores, 70-90cm long, were collected from the deepest point of each lake during 1980 and 1981. Sediment chronologies were constructed using ^{210}Pb dating (Appleby & Oldfield 1978), limnological history and past water pH values were inferred using diatom analysis and the Index B method of Renberg & Hellberg (1982), and catchment history is based on local information and pollen analysis. Geochemical analysis, not funded by the contract, was carried out on the L. Enoch core.

In addition background information on contemporary diatom populations, water chemistry and lake bathymetry were obtained. Epilithon and epipsammon diatom samples were collected from all sites eight times during 1981/2 and the material has been used for reference purposes. Water quality samples were also collected from all sites during 1981/2 and these were analysed at CERL (Table 2,3). New bathymetric maps were made of L. Dee and Round L. of Glenhead (Fig. 8,9), using standard field techniques and computer contouring (MAPICS 1984).

The following sections summarise the results of the study and mainly comprise papers published, in press, or submitted for publication.

TABLE 1. LAKE AND CATCHMENT DATA FOR THE SIX SAMPLE SITES.

	Lake Area	Altitude	Maximum Depth	Afforested Area	Unafforested Area	Date of First Planting
	(ha)	(m)	(m)	(ha)	(ha)	
Round Loch of Glenhead	12.6	299	13.5	-	95.1	-
Loch Enoch	50.1	493	36.0	-	185.7	-
Loch Valley	34.7	320	16.5	-	640.1	-
Loch Dee	100.0	225	14.7	291.7	1190.9	1973
Loch Grannoch	114.3	210	21.1	895.1	391.9	1962
Loch Skirrow	50.7	126	10.1	198.0	166.0	1957

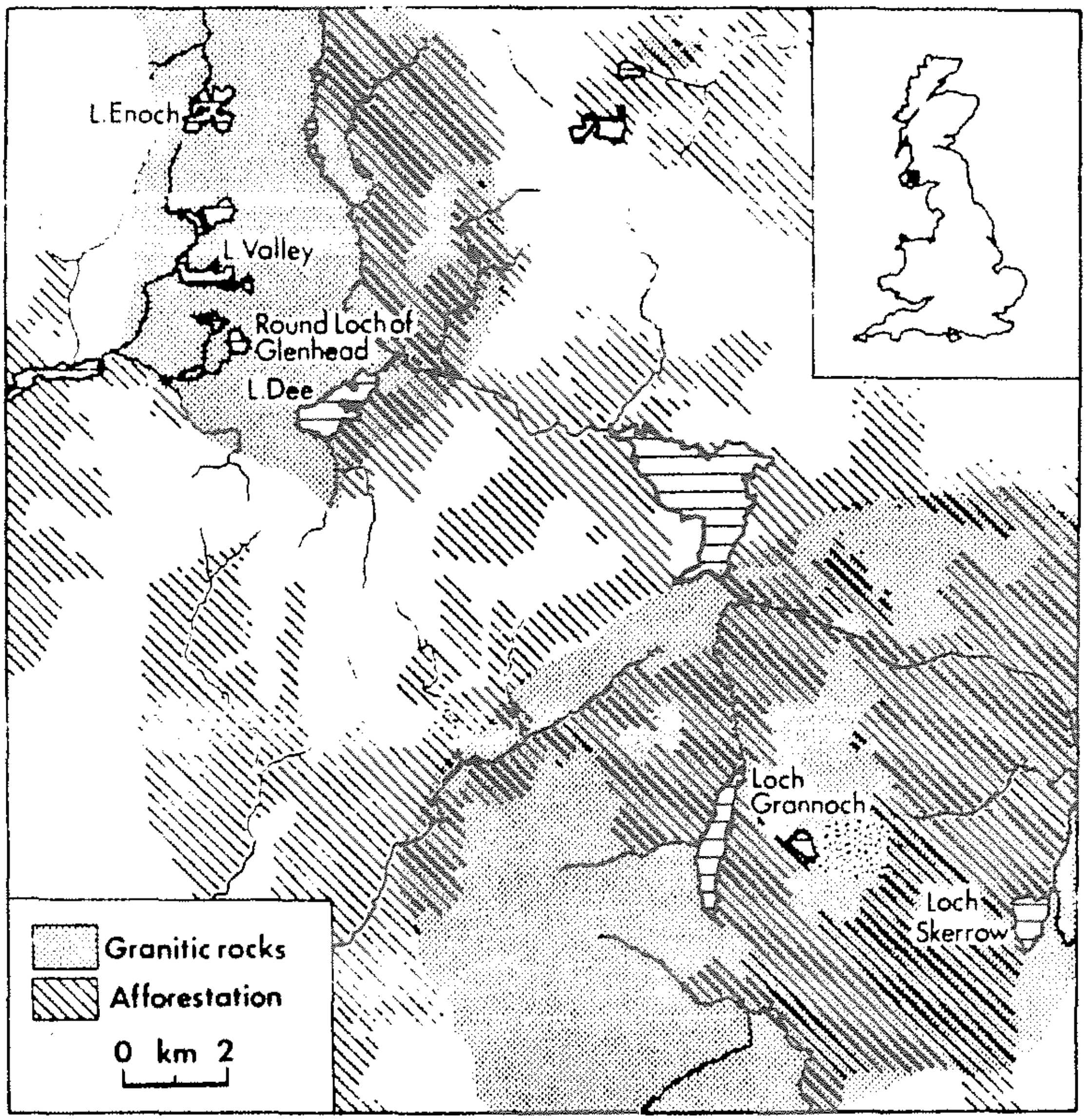


Figure 1. The study area and sample sites.

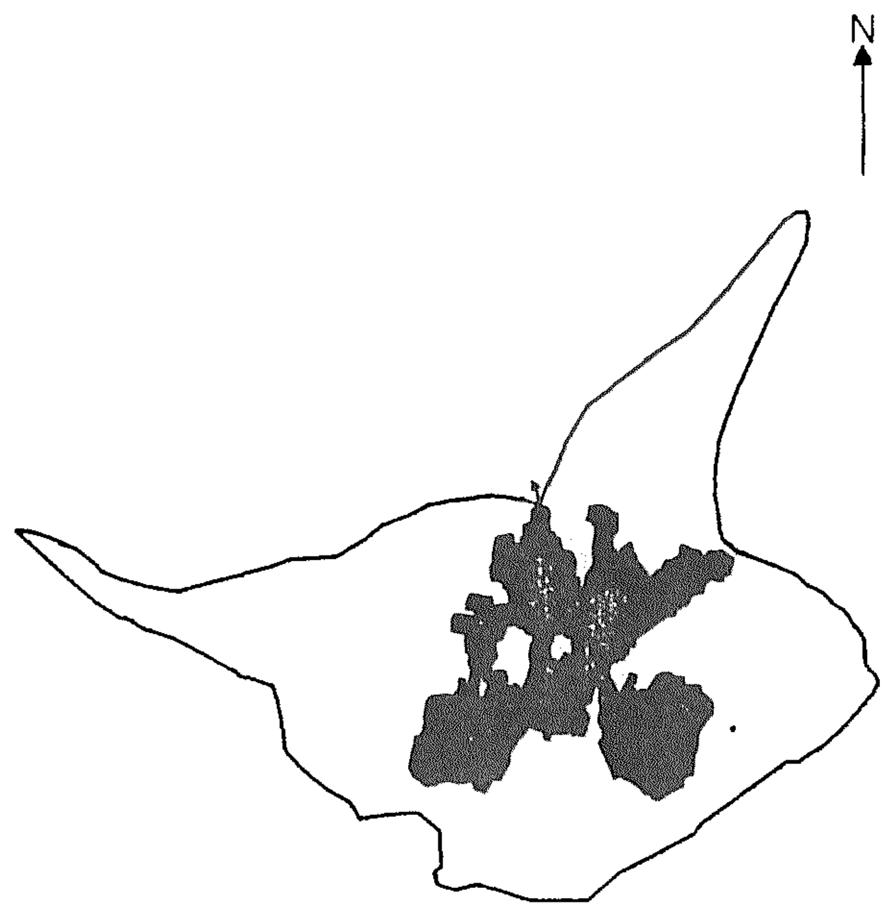


Fig. 2. Loch Enoch Catchment.

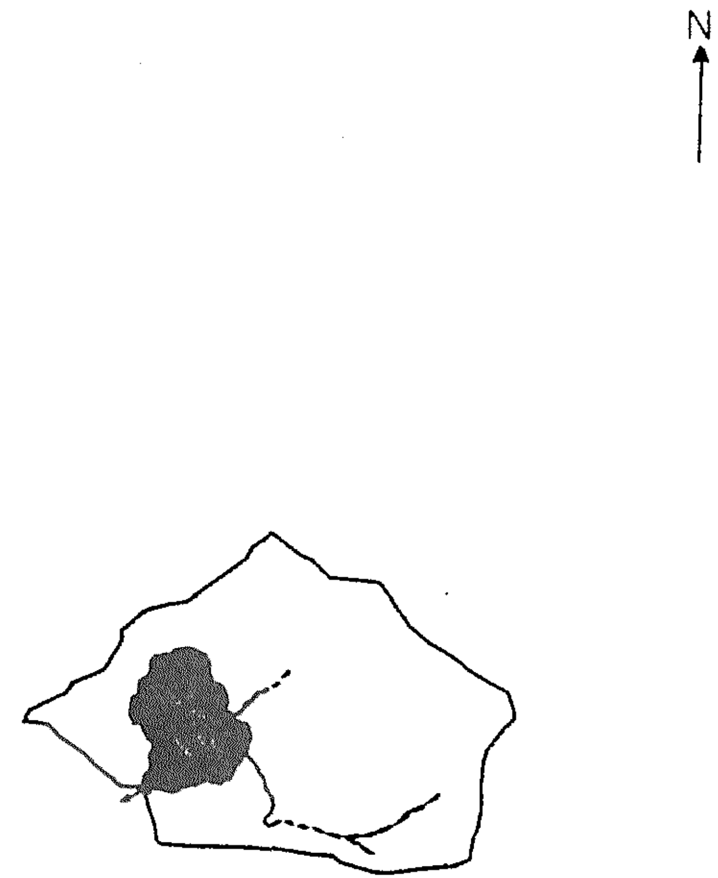


Fig.3 Round Loch of Glenhead Catchment.

1 kilometre.

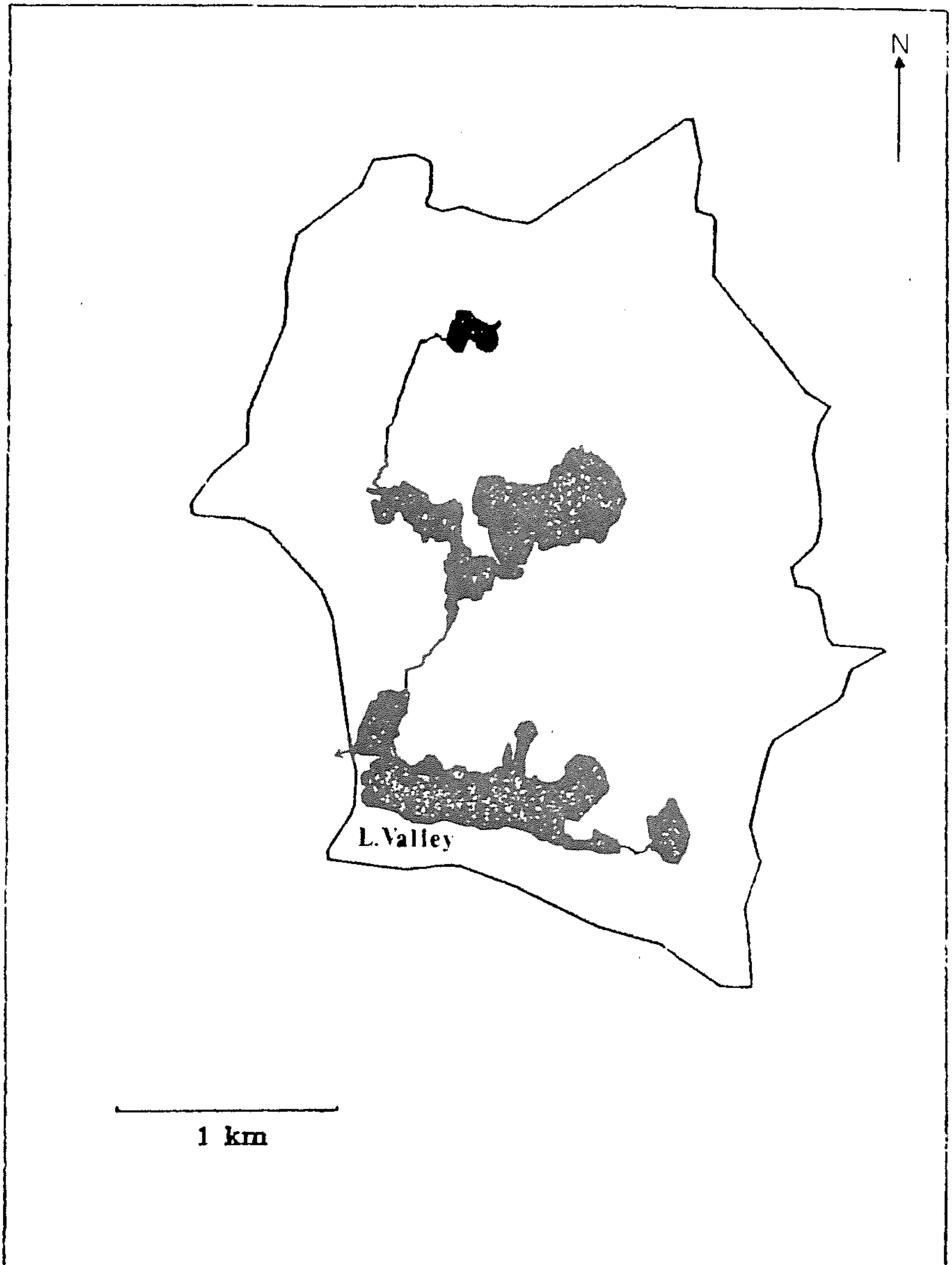
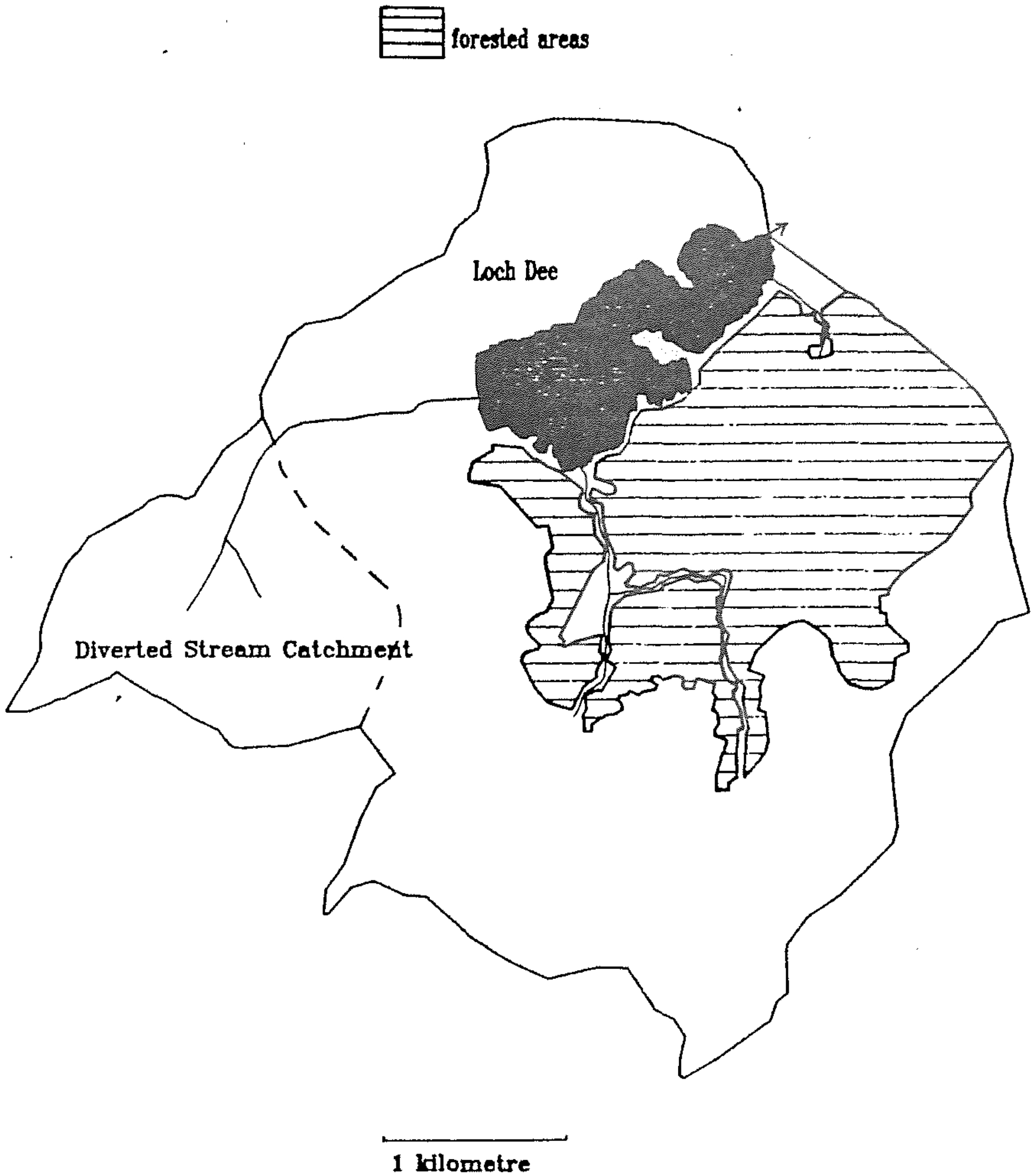


Fig.4 Loch Valley Catchment.

Fig. 5 Loch Dee Catchment.



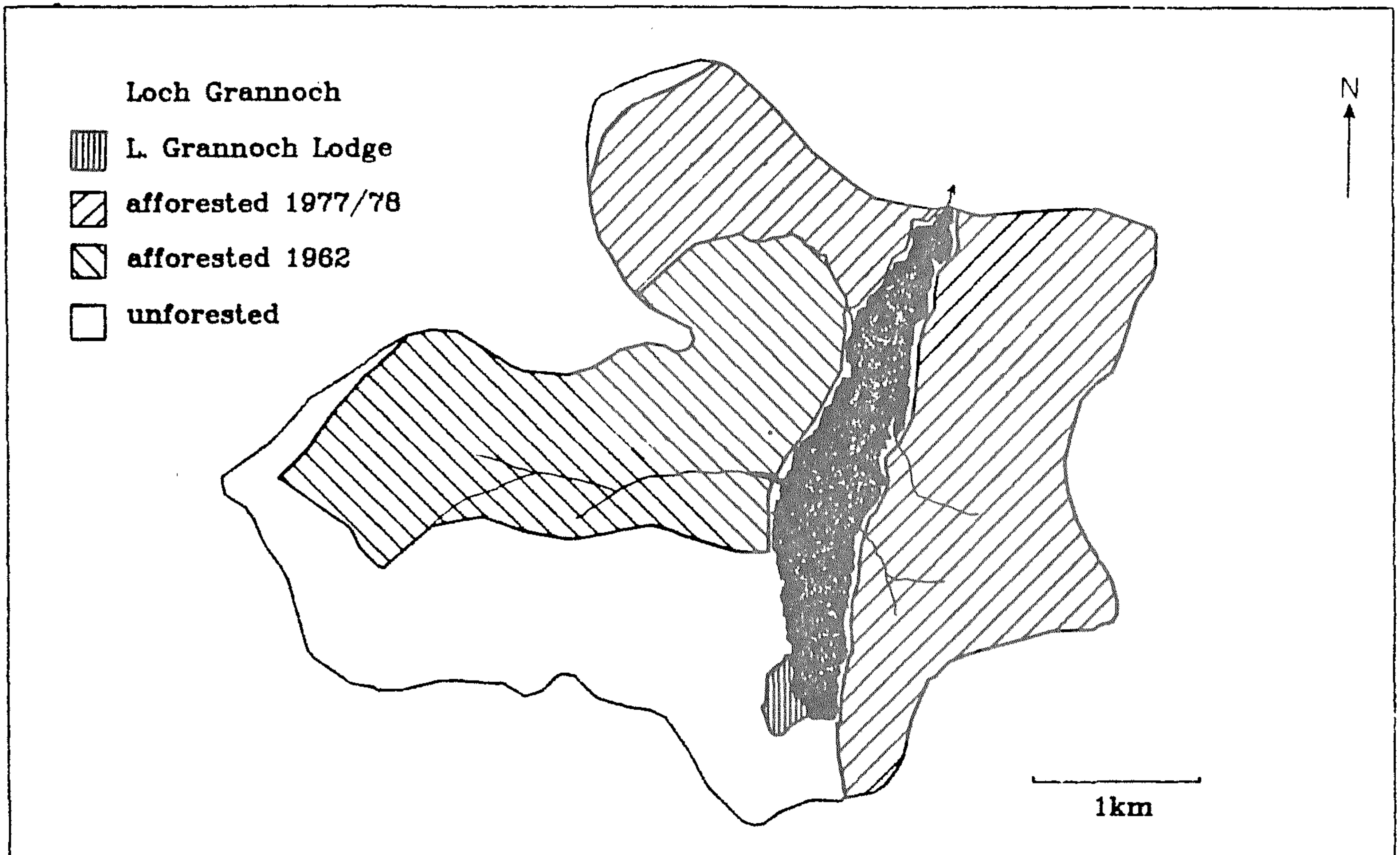


Fig. 6 Loch Grannoch Catchment.

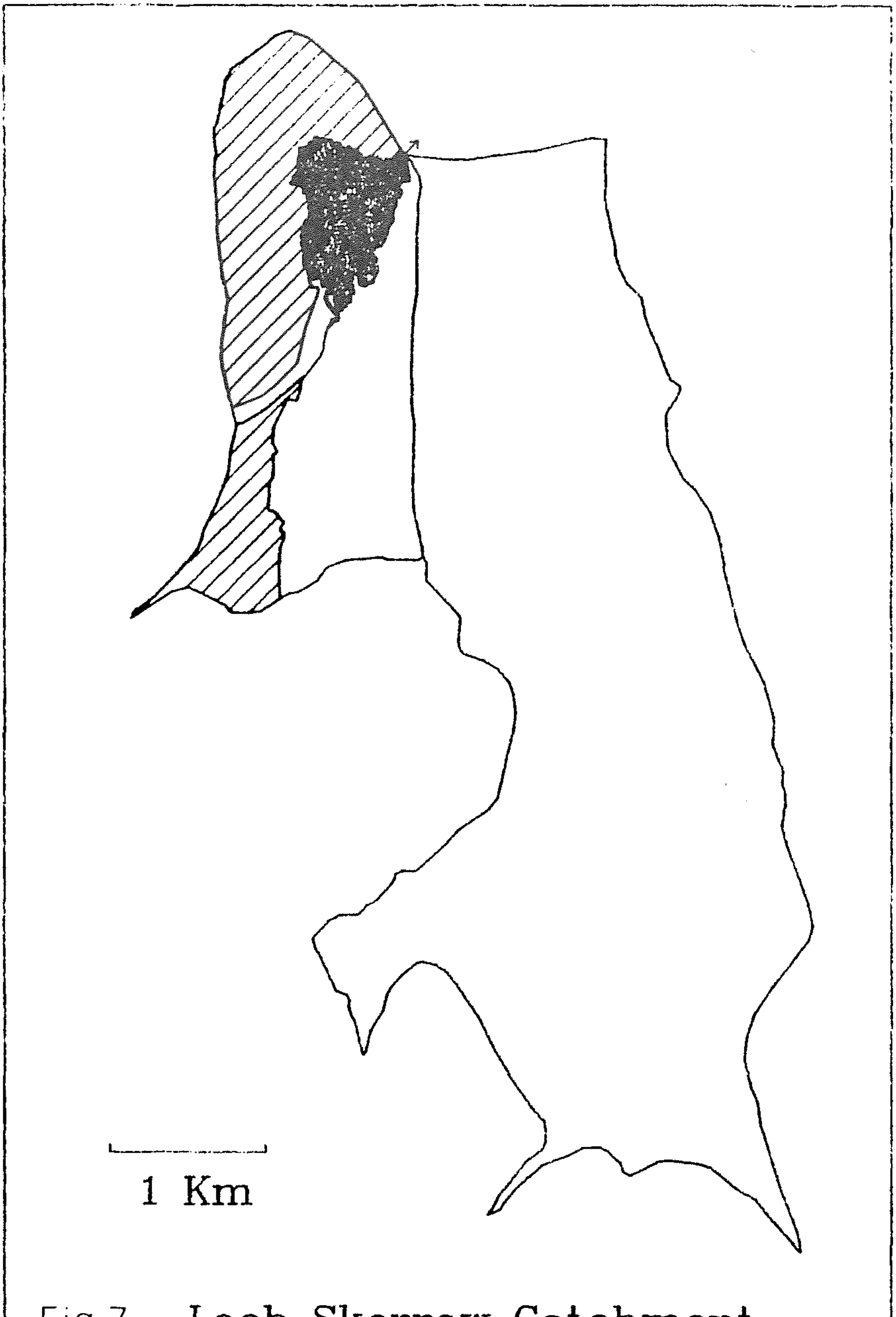


Fig. 7 Loch Skerrow Catchment.

hatching denotes forested areas.

TABLE 2. MEAN VALUES FOR WATER QUALITY DATA.

	Mean Temperature (°C)	Mean pH	Mean $[H^+]$ (μ eq H^+ l^{-1})	Mean Conductivity (μS cm^{-1} at 18°C)
ROUND LOCH OF GLENHEAD	9.0(4.5)	4.7	18.6(5.5)	30(4.6)
LOCH ENOCH	7.0(4.4)	4.5	28.9(6.8)	30(8.0)
LOCH VALLEY	8.5(4.6)	4.7	19.7(3.4)	31(4.1)
LOCH DEE	9.0(4.3)	5.3	5.4(3.6)	37(14.8)
LOCH GRANNOCH	9.1(4.3)	4.6	24.1(9.2)	38(4.9)
Loch Skirrow	--	5.2	8.4(6.1)	64(17.4)

() Standard deviation of the mean values

TABLE 3. MEAN VALUES FOR THE WATER QUALITY DATA. All concentrations in $mg\ l^{-1}$.

	Ca	Mg	Na	K	Fe	Al	SiO_2	SO_4	Cl
ROUND LOCH OF GLENHEAD	0.39(0.17)	0.26(0.05)	1.62(0.32)	0.20(0.08)	0.07(0.03)	0.09(0.04)	0.47(0.28)	2.57(0.85)	4.43(1.35)
LOCH ENOCH	0.21(0.07)	0.21(0.07)	1.34(0.56)	0.26(0.18)	-	0.09(0.04)	0.27(0.16)	2.14(0.62)	3.58(0.63)
LOCH VALLEY	0.25(0.08)	0.22(0.06)	1.45(0.97)	0.24(0.8)	0.03(0.01)	0.10(0.03)	0.25(0.17)	1.30(0.18)	4.43(3.72)
LOCH DEE	0.52(0.28)	0.34(0.16)	1.47(0.76)	0.30(0.20)	0.04(0.01)	0.07(0.01)	0.32(0.31)	2.74(1.45)	5.00(2.31)
LOCH GRANNOCH	0.44(0.18)	0.25(0.10)	1.47(0.65)	0.27(0.07)	0.05(0.01)	0.13(0.05)	0.55(0.39)	3.04(1.36)	3.93(1.13)
LOCH SKIRROW[1]	2.01 --	0.92 --	3.60 --	0.43 --	-- --	0.22 --	-- --	7.30 --	5.20 --

() Standard deviation of the means

[1]From Wright and Henriksen =979

Fig.8 **Round Loch of Glenhead** : Bathymetry

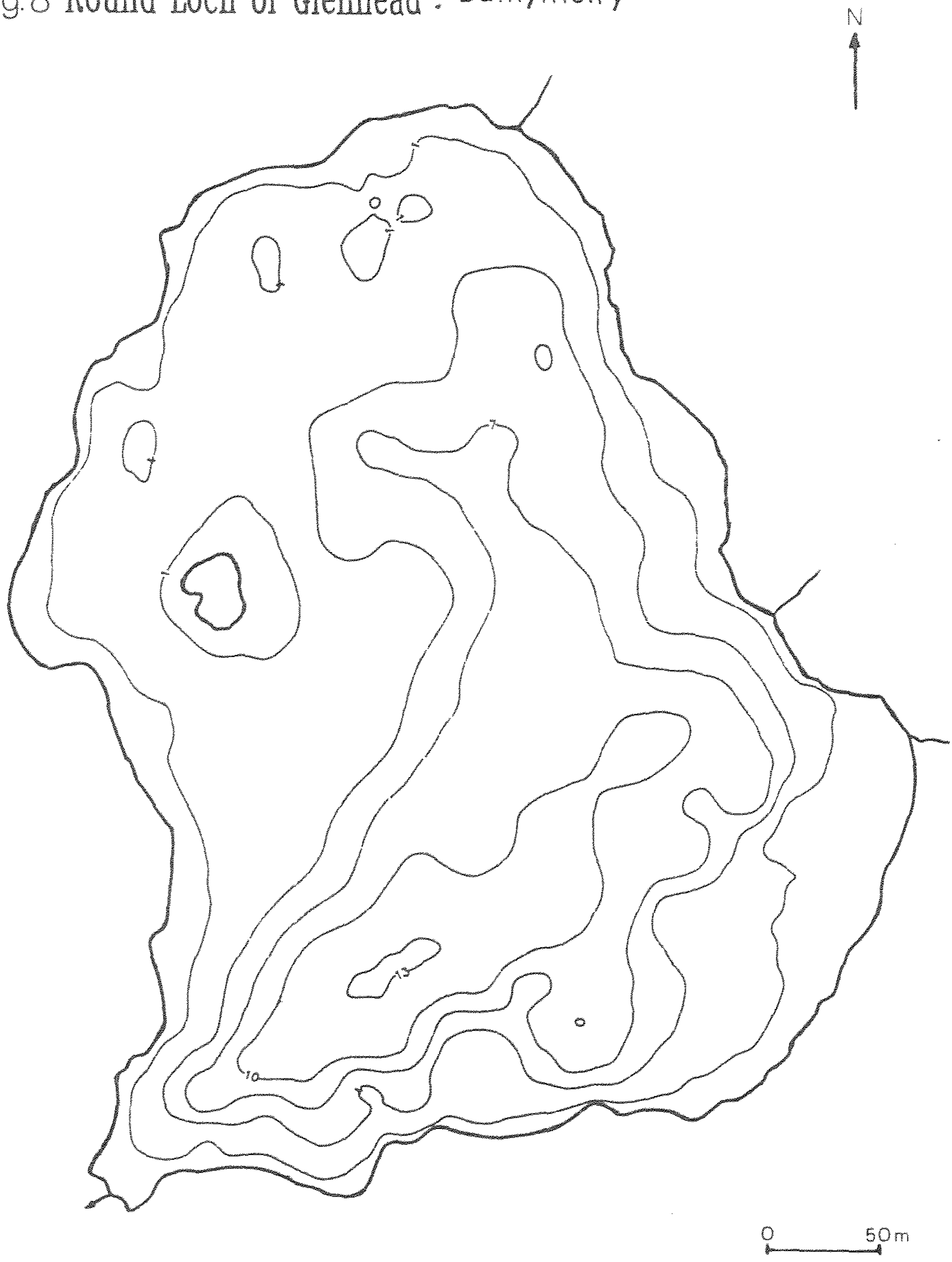
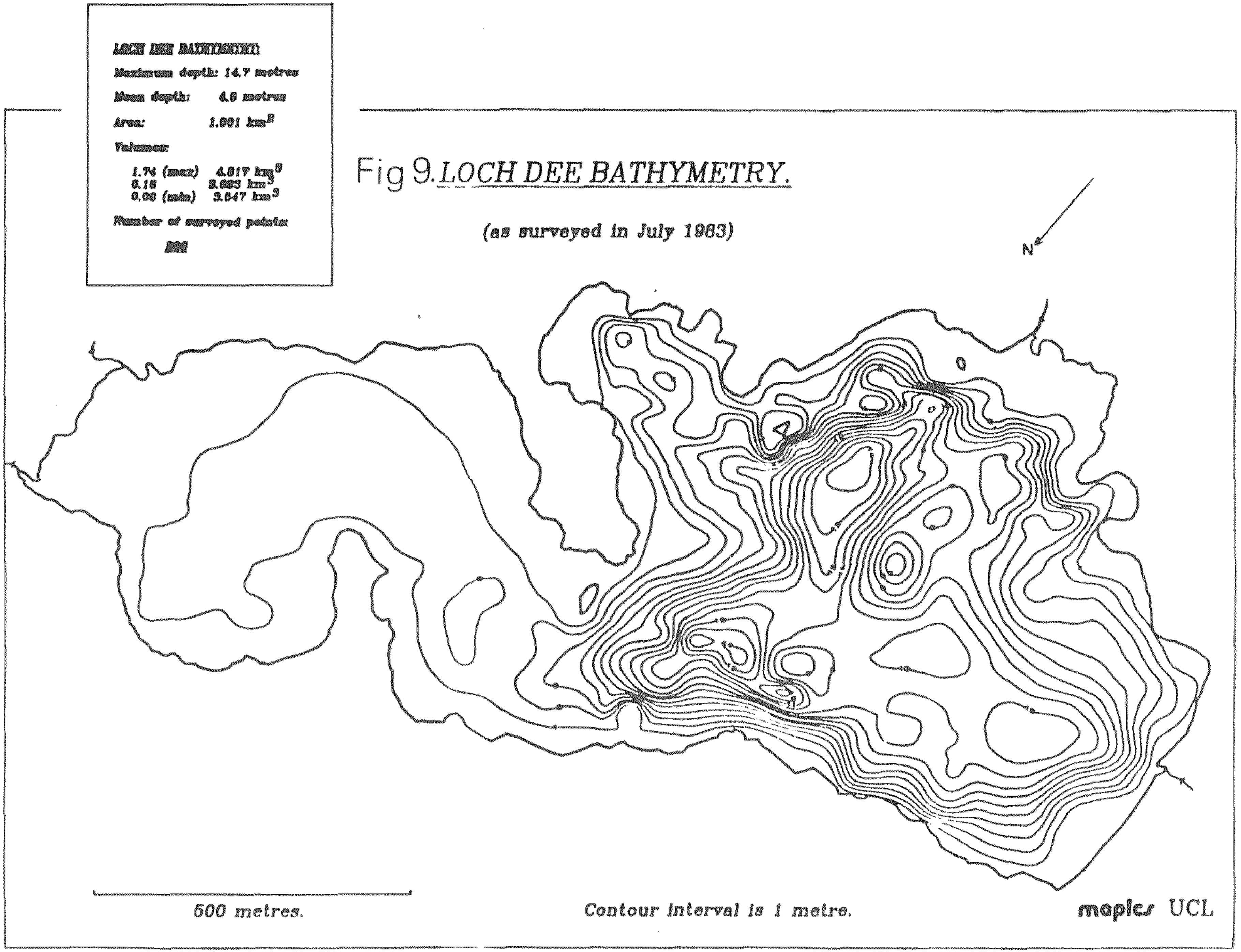


Fig 9. LOCH DEE BATHYMETRY.

(as surveyed in July 1983)



^{210}Pb DATING OF SCOTTISH LAKE SEDIMENTS AFFORESTATION AND ACCELERATED SOIL EROSION

INTRODUCTION

Whilst accelerated soil erosion following forest clearance has been widely reported (e.g. Blackie *et al.* 1980) the erosive effects of pre-afforestation ploughing are less well known (Painter *et al.* 1974, Robinson & Blyth 1982). In recent decades extensive areas of the upland regions of Britain have been afforested. Much of the afforestation has occurred in areas covered by blanket peats where deep ploughing for drainage purposes is necessary before planting. Where lake catchments have been afforested acceleration in the rates of erosion can be identified from changes in the rates of sediment accumulation. In this study of lakes in Galloway, S.W. Scotland we evaluate the importance of this process by comparing accumulation rates of lakes that have partially afforested catchments with those of lakes with non-afforested catchments.

SITES

The seven lakes for which data are available are all situated on granite intrusions (Figure 1) but have differing sizes, altitudes, catchment relief, and catchment to lake area ratios (Table 4). Round Loch of Glenhead, L. Valley and L. Enoch are situated on the Merrick intrusion and have non-afforested catchments. L. Dee is also situated on the Merrick granite but approximately 20% of its catchment area has recently (1976) been planted. Lochs Grannoch, Fleet and Skerrow are situated on the Cairnsmore of Fleet intrusion and have partially afforested catchments (Table 4). All the lakes are acid and some have been recently acidified (Flower & Battarbee 1983, Battarbee 1984).

METHODS

One metre long sediment cores were taken from the deepest point in the seven lochs using a Mackereth mini-corer (Mackereth 1969). The cores were

Table 4 Selected characteristics of the lakes and their catchments

Lake	Altitude (m)	Area (ha)	[1]Catchment Area: Lake Area	[2]Maximum Relief* (m)	% Afforestation	Date of Ploughing
L. Enoch	493	50.1	3.7:1	350	-	-
L. Valley	320	34.7	1.9:1	334	-	-
Round Loch of Glenhead	299	12.6	8.6:1	232	-	-
L. Dee	225	100.0	14.8:1	306	20.8	1975
L. Grannoch	210	114.3	12.3:1	391	69.5	1961,1976
L. Fleet	344	17.3	6.2:1	127	21.8	1961
L. Skerrow	126	50.7	30.1:1	163	14.8	1955

[1]Catchment area excludes lake area.

[2]Difference between lake altitude and highest altitude in catchment.

extruded vertically. sliced into 1 cm or ½ cm slices and routine measurements of dry weight, wet density and loss on ignition were made. ^{210}Pb measurements, following Häsänen (1977), were made at regular intervals down each core until background levels were reached. ^{226}Ra determinations were also carried out at more widely spaced intervals to allow unsupported ^{210}Pb values to be calculated. Dates were calculated according to both the 'cic' and 'crs' models (Appleby & Oldfield 1978, 1983). Where profiles were clearly non-monotonic the crs model is preferred. In other cases the mean of crs and cic derived dates have been used.

RESULTS

1. Unsupported ^{210}Pb concentration

Figure 2 shows the unsupported ^{210}Pb concentration versus depth profiles for all seven lakes. The data are divided into sites with non-afforested catchments (Figure 10a) and those with afforested catchments (Figure 10b). Where relevant, the dates of catchment ploughing are indicated. It is clear that L. Enoch, L. Valley and Round Loch of Glenhead have monotonic exponential declines in ^{210}Pb whilst all others show various degrees of non-linearity. For L. Skerrow, planted in 1955, the profile is non-linear between 4.5 and 15.5 cm but there is a progressive downcore decline in values. For Loch Grannoch, L. Fleet and L. Dee inversions of ^{210}Pb occur. In L. Dee it is confined to the upper 10 cm, in L. Grannoch the upper 30 cm show inverted values whilst in L. Fleet the inversions occur between 30 cm and approximately 10 cm. The upper 10 cm in this core show a linear profile. Inversions and non-monotonic ^{210}Pb profiles have been noted before (Oldfield et al 1978) and indicate ^{210}Pb dilution by the increased accumulation rate of sediment, normally as a result of catchment disturbances and the increased input of allochthonous material. In Loch Skerrow, L. Dee and L. Grannoch the disturbances in ^{210}Pb concentration can be seen to be

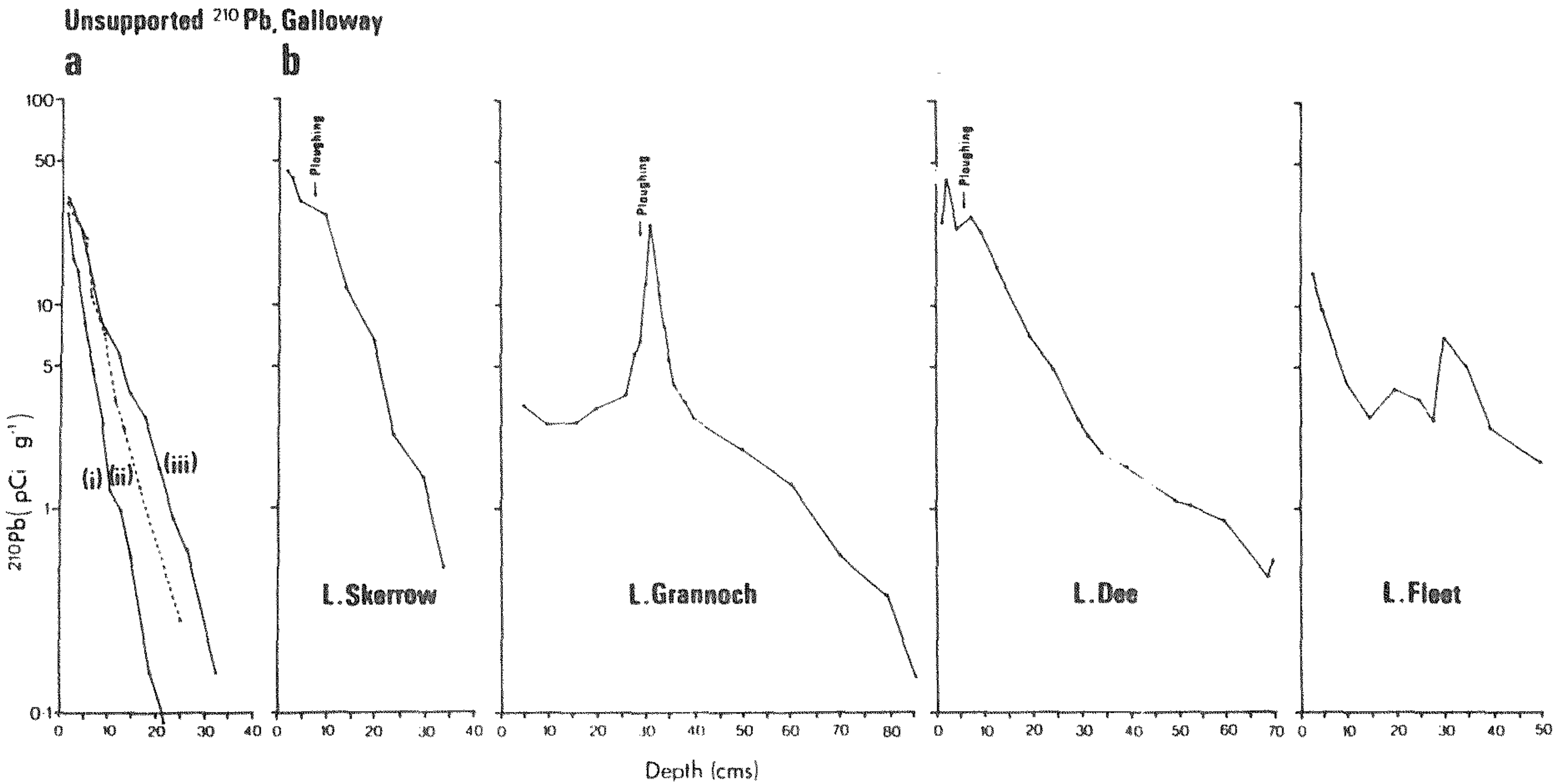


Figure 10 Unsupported ^{210}Pb vs depth for lakes with (a) unafforested and (b) afforested or partially afforested catchments.

(i) = Round Loch of Glenhead; (ii) = Loch Valley; (iii) = Loch Enoch.

Standard errors of the counts are omitted for clarity.

approximately coincident with the first date of ploughing. In Loch Fleet there is clear evidence of the same effects but so far it has been impossible to work out a chronology for this site because of the variability of both the ^{210}Pb and the ^{226}Ra data.

2. Sediment accumulation rates

Figure 11 shows variations in sediment accumulation rate through time for all lakes excluding Loch Fleet. The non-afforested sites (Figure 11a) display little or no change over the last 200 years with mean rates varying from 0.1 cm a^{-1} for Round Loch of Glenhead to 0.3 cm a^{-1} for Loch Enoch. Afforested sites all show variations in sediment accumulation although at Loch Dee and L. Grannoch there are significant changes in accumulation rate that pre-date any afforestation effects. At both these sites decreases in accumulation rate, at 1900 and 1930 respectively, are probably the result of a reduced flux of diatoms to the sediment associated with acidification processes, since in both cases the onset of these changes coincide with the first diatom microfossil evidence for acidification (Flower & Battarbee 1983). On the other hand, more recent increases in sediment accumulation rate all coincide with dates for pre-afforestation ploughing. For Loch Skerrow the data indicate a temporary increase in accumulation rate from 0.3 cm a^{-1} to 0.8 cm a^{-1} starting at the time of ploughing (c. 1955). The subsequent fall (Figure 11b) to 0.25 cm a^{-1} may be related to the re-stabilisation of the catchment soils after canopy closure. The low relief of the Loch Skerrow catchment also probably helps to minimize continued erosion.

The Loch Grannoch site shows the most dramatic evidence of accelerated erosion. The date for the rapid rise in accumulation rate agrees almost exactly with the date for the beginning of ploughing in the catchment of 1961. The rate rises from a low of 0.2 cm a^{-1} in 1958 to a contemporary value of 2.2 cm a^{-1} (Figure 11b). Unlike Loch Skerrow there is no evidence

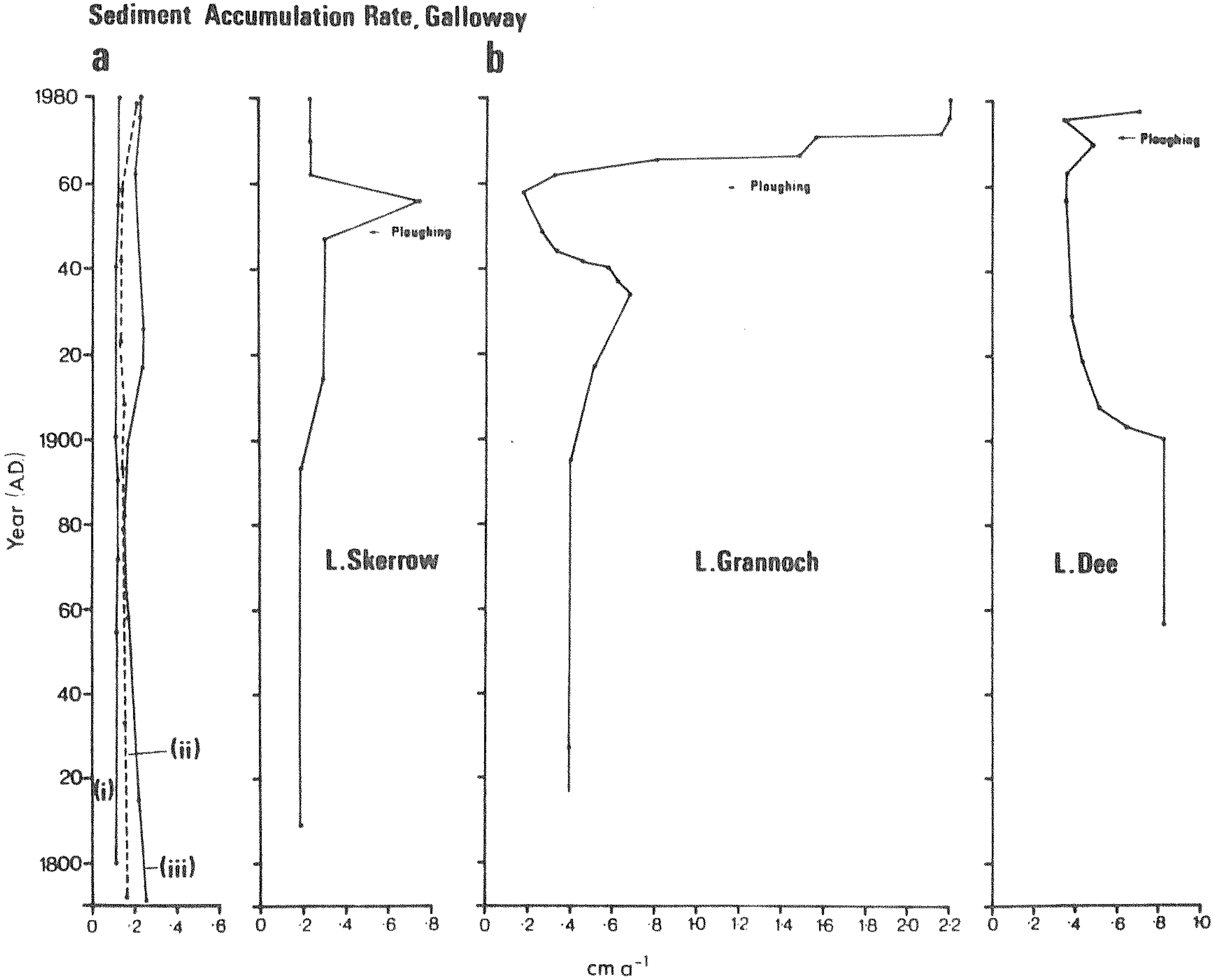


Figure 11 Sediment accumulation rate vs time (as for Fig. 10).

of a clear decline in values towards the sediment surface and this may be due to the further ploughing and planting of additional parts of the L. Grannoch catchment in 1976. However, the levelling off in the sediment accumulation rate over the top 15 cm may be indicative of the beginning of a reduction in erosional intensity as the canopy has now closed in all areas planted in 1962. The Loch Dee catchment has only recently been afforested and the effect on sediment accumulation is consequently only apparent in the uppermost part of the core. The accumulation rate increases from a pre-planting rate of 0.35 cm a^{-1} to a contemporary rate of 0.7 cm a^{-1}. Recent management practices introduced by the Forestry Commission at this site aimed at reducing the movement of sediment into the streams and lake may prevent the rate from increasing. ^{210}Pb dates from future lake sediment cores should allow the effectiveness of these management practices to be evaluated.

DISCUSSION

An assessment of the data presented here needs to take into account the limitations of results based only on single cores and the errors associated with radiometric dating techniques. In each case the rates calculated apply only to the central part of the lake basin and as such probably reflect the rates of the sites of fastest accumulation. Moreover, this rate itself is influenced by the morphometry of the lake basin as well as by the quantity of particulate material received by and produced by the lake. Between-lake comparisons of sediment accumulation rate can therefore only be made with caution and small differences between sites are not significant. In addition, it is not possible to use these data to calculate mean loss rates from the catchment since the relationship between the central core rate and the mean rate for the lake is not known (cf Dearing 1983) and the sediment core data are biased towards organic material and fine (silt/clay) minerogenic fractions.

The significance of the data however lies in the change in accumulation rate in each basin, the dates of these changes and the comparison between non-afforested and afforested sites. All the lakes considered are acid lakes, situated on similar geological substrates with similar climate regimes. They differ essentially in altitude, size, relief and history of catchment disturbance. Altitude, size and relief are approximately constant over time, at least in the short and medium term. It can be seen that the recent changes in sediment accumulation are closely related to afforestation history. Round Loch of Glenhead, L. Valley and L. Enoch show little change over the last 200 years whereas all the afforested lakes show varying degrees of change. The most extreme case is Loch Grannoch where a combination of high relief and successive ploughing episodes have maintained high accumulation rates approximately 10 times higher than pre-planting rates. Data for Loch Skerrow, on the other hand, show that where only a single episode of ploughing has taken place a higher accumulation rate is maintained for approximately 10 years before a decline to base rates occur. The Loch Dee disturbance has so far been minor possibly because of efforts to prevent drainage ditch waters discharging directly into streams and the lakes.

Whilst these data indicate that high erosion rates associated with ploughing eventually decrease it is clear that the catchments of these lakes are highly susceptible to erosion and that further pulses of increased erosion are likely to be caused when current forests are felled and a second crop planted. These conclusions are likely to apply to a large area of the British uplands wherever routine deep ploughing is practised prior to afforestation. Moreover, many of these lakes have impoverished fisheries probably as a result of acidification and it is likely that the fisheries problems are exacerbated by the effect of soil erosion stemming from such drainage practices.

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DIATOM EVIDENCE FOR THE RECENT ACIDIFICATION OF TWO SCOTTISH LOCHS

There have been many claims that highly acid (pH <5.0) often fishless lakes have become so as a result of recent (mainly 20th century) acidification processes. Many of these claims have not been fully validated since there are few acid lakes that have been the subject of long-term continuous monitoring. However, diatom analysis of ^{210}Pb-dated sediment cores can provide the detailed historical perspective required. Here we use these techniques to show that some lakes in the United Kingdom have become strongly acidified within the last 130 years. Analysis of cores from two lakes in Galloway, south west Scotland, indicate that both lakes, one with an unafforested catchment and one with a recently afforested catchment, have experienced pH declines of approximately 1 pH unit (from c. pH 5.5 to 4.5) during this time.

Following early reports of recent lake acidification in Norway,[1] Sweden,[2] and Canada[3] lakes with pH values <5 have been noted in several other countries.[4-8] In the United Kingdom the high acidity of precipitation and the occurrence of acid lakes and streams has been documented.[9-15] There is dispute, however, as to whether these acid lakes and streams have become increasingly acid over recent decades. According to Sutcliffe *et al*[15] chemical records for some Lake District lakes show no evidence of acidification over the period 1928-1980. On the other hand, lakes draining granitic catchments in Galloway, south west Scotland, that currently have pH values ranging from c. 4.5 to 6.0,[16] no longer support a salmonid fishery while others have declining fish populations.[17]

The increased acidity of lakes reported from different parts of the world has been ascribed mainly to acid precipitation,[1-5,7,16,17] but catchment land-use changes, including afforestation,[18,19] and the longer-term continuous effects of soil leaching associated with early forest clearance and climatic change,[20] have also been implicated. Since historical data are mainly inadequate or lacking, evidence for acidity changes in lakes can best be found in sediments by diatom analysis. Diatoms are siliceous algae and are usually found in great abundance and diversity in lakes and rivers. They are sensitive to changes in water quality and floristic analysis of their fossil remains in lake sediments can be used to reconstruct former pH values of lake water.[21-25]

We are carrying out diatom analysis and ^{210}Pb dating of sediment cores from a range of lakes in the Galloway region not only to assess the history of acidification in the area but also to evaluate the role of afforestation as a cause of acidification. Here we report on a comparison between a core from a loch with an unafforested catchment (Round Loch of Glenhead) and one from a loch with a partially afforested catchment (Loch Grannoch, afforested in 1962-4 and 1977-8) (Fig. 12). Lake and catchment characteristics are given in Table 5. Both lakes are situated entirely on granite, soils are mainly blanket peats and both lakes have similar modern pH ranges.

Cores approximately 80 cm long were taken at the deepest point in each lake. The sediment was extruded at 0.5 or 1 cm intervals. ^{210}Pb measurement follows Häsänen[26] and dates were calculated according to the crs model.[27,28] The species frequency composition of the diatom assemblages was analysed using standard techniques.[29] Over 140 taxa were identified at the two sites. A selection of those that show significant stratigraphic changes through the core sequence at each site are shown in Figs. 13 and 14. ^{210}Pb dates and pH curves calculated to an accuracy of ca. ±0.3 units using Index B of Renberg[24] are also shown.

For Round Loch of Glenhead the ^{210}Pb chronology indicates that the rate of sediment accumulation at this site is 1 mm a^{-1} and has been approximately constant over the last 200 years with unsupported ^{210}Pb concentrations reaching background levels at about 25 cm. If this mean rate were extrapolated the 80 cm core covers approximately the last 700 years of accumulation. It can be seen for the period spanning approximately 1300 AD to 1600 AD (80-40 cm) that the diatom flora of the lake was dominated by Anomoeoneis vitrea (Grun.) Ross, Eunotia veneris (Kütz). O. Mull., Fragilaria virescens Ralfs, Cyclotella kützingiana Thwaites and Melosira distans (Ehr.) Kutz. There was little significant change within the period and the pH reconstruction gives a value of about 5.7. Somewhat before about 1600 AD (40 cm) there was a strong change in the composition of the flora as Cyclotella kützingiana and C. arentii Kolbe declined rapidly. The reason for this change is not clear but Cyclotella kützingiana is a planktonic diatom found mainly in water with circumneutral pH. Planktonic diatoms seem to be rare in waters below about 5.5[25] and the reconstructed pH curve suggests that there was a slight increase in acidity at this time. Since the change predates the beginning of significant fossil fuel combustion it is likely to be related either to a climatic, or more probably, to a land use change at that time.

Between c. 1600 and c. 1850 (40-15 cm), fairly stable conditions are indicated but a second floristic change begins from about 1850 (15 cm). The following changes are sequential and indicate a progressive decline in pH from the mid-nineteenth century to the present. The first species to change were Fragilaria virescens and Achnanthes microcephala (Kütz.) Grun., both of which declined. Anomoeoneis vitrea also declined, but more gradually and there were reductions in Nitzschia perminuta Grun. (at 13 cm), Navicula cocconeiformis Greg. (at 10 cm), and Cymbella gracilis (Rabh.) Cleve (at 5 cm), all taxa associated with less acid waters. These taxa were replaced by the increase of species typical of more acidic waters especially Tabellaria quadriseptata Knudsen, T. binalis (Ehr.) Grun., Eunotia veneris, E. alpina (Naeg.) Hust, E. arcus Ehr., E. bactriana Ehr.,

E. tenella (Grun.) Hust, and Navicula höfleri Cholnoky. The reconstructed pH curve shows a continuous but variable rate of decline in pH from about 1850 to the present day from about 5.5 to about 4.6. The data indicate that the rate has been more rapid since 1900 (about 0.01 pH unit a^{-1}). The calculated pH value of 4.8 ±0.3 from the surface sediment sample at this site agrees well with the present measured pH of the lake water (cf. Table 5).

Although the cause of the recent acidification at this site is not known, it is possible to eliminate ploughing and afforestation as factors. Our unpublished historical, palynological and lithostratigraphical data contain no evidence for significant alteration in land-use practice that might have promoted acidification in this lake and therefore the most likely cause is an increase in acidity deposited from atmospheric sources, a supposition that is consistent with the post-Industrial Revolution timing of the changes.

The core from Loch Grannoch covers a much shorter time period than that from Round Loch of Glenhead since the sediment accumulation rate is faster. This is especially so for the top 30 cm where the accumulation rate increases to 1 cm a^{-1} as a result of accelerated soil erosion associated with pre-afforestation ploughing. Consequently, the whole 80 cm falls within the ^{210}Pb dating range and includes sediment accumulated from about 1840 to the present. Some of the floristic changes are similar to those at Round Loch of Glenhead but the recent history of Loch Grannoch is complicated by the additional effects over the last 20 years of soil erosion and afforestation.

Figure 14 shows that there was little floristic change in Loch Grannoch between 1840 and about 1930. Except for the presence of two additional taxa, Eunotia pectinalis v. minor (Kütz.) Rabh. and Achnanthes marginulata

Grun., that probably reflect the greater importance of epipsammic habitats in Loch Grannoch, the flora of this period (Fig. 14, 80-40 cm) is similar to that at Round Loch of Glenhead during the previous 300 years (Fig. 13, 40-20 cm), and represents a lake pH of 5.5 ±0.3. The first evidence of acidification occurs within this period at about 1910 (50 cm) and is indicated by a slight shift in the composition of the assemblage. Nitzschia perminuta and Achnanthes microcephala decline, and Frustulia rhomboides v. saxonica (Rabh.) de Toni, an acidophilous form, increases. An acceleration in the process occurs in the 1920s and 1930s as Anomoeoneis vitrea, Fragilaria virescens, Nitzschia perminuta and Cymbella gracilis decline, paralleling identical but earlier changes in the Round Loch of Glenhead core (cf. Fig. 13, 10-15 cm) and indicating a decline in pH to about 5.0 by 1940. The most marked change in the flora, however, occurs between 1940 and 1960 (35-30 cm) as the two acidobiontic species Tabellaria binalis and T. quadriseptata appear and rapidly expand, representing a further and more rapid decline in pH values to levels similar to those of today. Overall, the floristic changes from 1910 to 1960 suggests that there was a drop of approximately 1 pH unit from about 5.5 to about 4.5 during this period. Since the change pre-dates the beginning of afforestation (1962) in the Loch Grannoch catchment this acidification cannot be ascribed to afforestation processes, and as there is no historical evidence for a significant change in other land use practices during this period it is most likely, as at Round Loch of Glenhead, that the increased acidity was derived from atmospheric sources.

In 1961 the eastern part of the Loch Grannoch catchment was ploughed prior to planting in 1962 (Fig. 12). The effect is clearly registered in the sediment by a sudden increase in sediment accumulation derived from a sharp decrease in ^{210}Pb concentration at the 29-30 cm level. It can be seen (Fig. 14) that immediately after the onset of accelerated erosion there is a strong decrease in *Tabellaria binalis* and *T. quadriseptata*, followed by a recovery to higher values in the upper 8 cm (from 1977). In the top few centimetres *Anomoeoneis vitrea* declines to trace levels and *Pinnularia hilseana* (Janish) Müll. and *A. marginulata* expand. Index B suggests that these diatom changes reflect a reversal in the trend of pH values from <4.5 in 1960 to about 5.0 in the mid-1970s followed by a second decline to modern values (4.4 - 5.0, Table 1) over the last 5-6 years. So far we have no firm explanations for this observation. It is likely, however, that the temporary rise of pH is an artefact caused by the addition of a significant quantity of diatoms from inwashed catchment peats depressing the percentages of *Tabellaria binalis* and *T. quadriseptata*. These are acidobiontic taxa that are not characteristic of catchment diatom floras.

Despite some uncertainty concerning the effects of afforestation a number of firm conclusions can be made from the results obtained at the two sites. First, the contemporary level of acidity in the lakes studied so far is a result of acidification during the late nineteenth and twentieth centuries. Second, the lakes were already acid (pH <6.0) before the recent changes took place and a change in water quality involving a slight acidification is apparent at Round Loch of Glenhead about 400 years ago. Third, the recent acidification has occurred in lakes with unafforested as well as afforested catchments. And fourth, although the diatom changes were similar at both sites, the timing and rates of acidification were different, suggesting that the acidification process is at least in part controlled by catchment characteristics.

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Table 5   Selected characteristics of the catchments.

	Alt (m)	Loch area (ha)	Maximum depth (m)	Catchment[1] area (ha)	Catchment:[1] loch	% afforested	pH range[3]	[4]Na^+	Ca^{2+}	Mg^{2+}	Al^{3+}	Cl^-	SO_4^{2-}	NO_3^-
Round Loch of Glenhead	299	12.6	13.5	95	8.6:1	0.0	4.5-5.0	113	30	41	13	136	111	6
Loch Grannoch		114.3[2]	21.1	1287	12.3:1	69.5[2]	4.4-4.9	152	50	50	30	165	152	8

1 - excludes loch area.

2 - includes 510.8 ha (39.7%) afforested in 1962-64, and 376.4 ha (29.8%) afforested in 1977-78.

3 - range of 8 values from samples taken at intervals from November 1981 to November 1982.

4 - Water chemistry values are taken from Wright and Henricksen.[16] Concentrations in $\mu eq\ l^{-1}$.

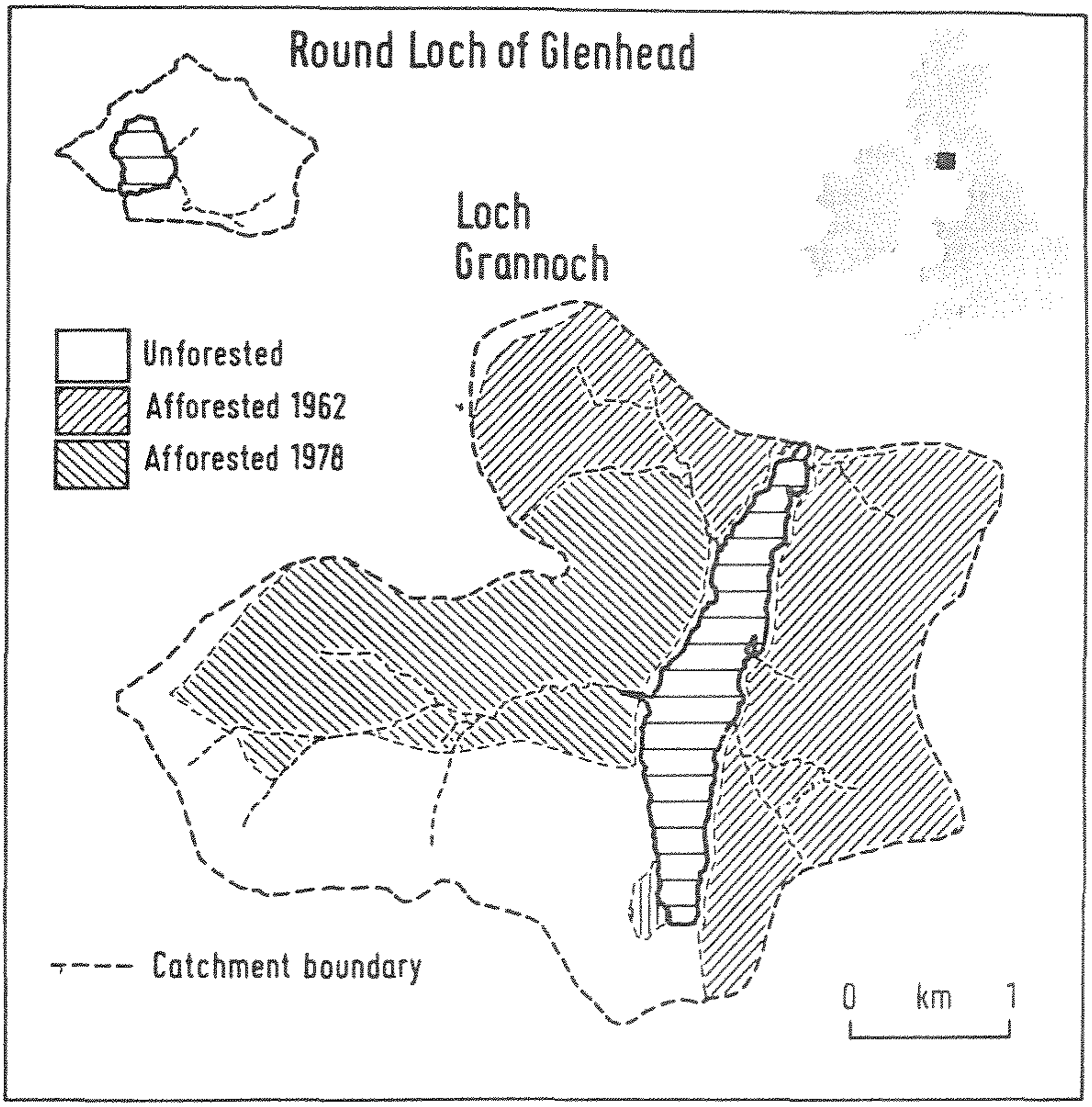

Figure 12 Site location and lake catchment maps for Round Loch of Glenhead and L. Grannoch showing areas afforested.

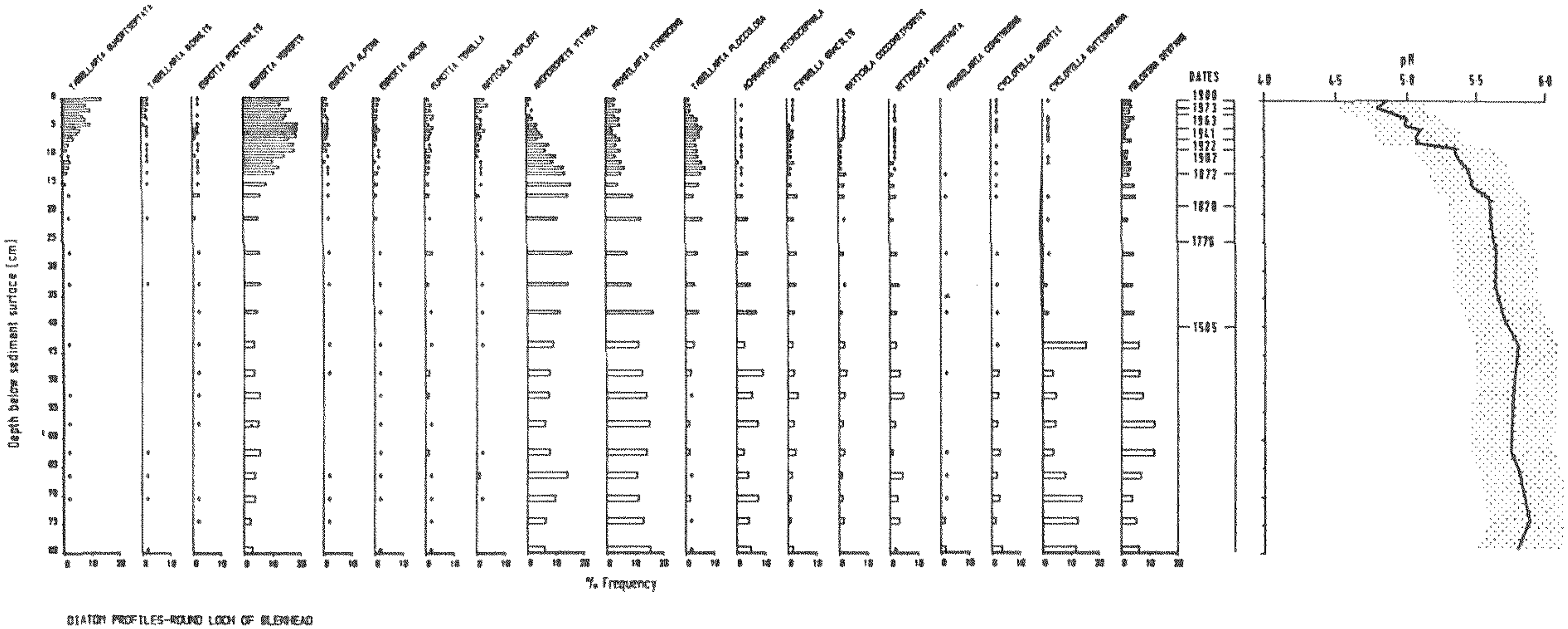

Figure 13 Diatom diagram for Round Loch of Glenhead showing percentage frequency of selected taxa, ^{210}Pb dates and pH values using Index B of Renberg and Hellberg.[17] Stipple on either side of pH curve indicates ±0.3 unit boundary * = extrapolated date.

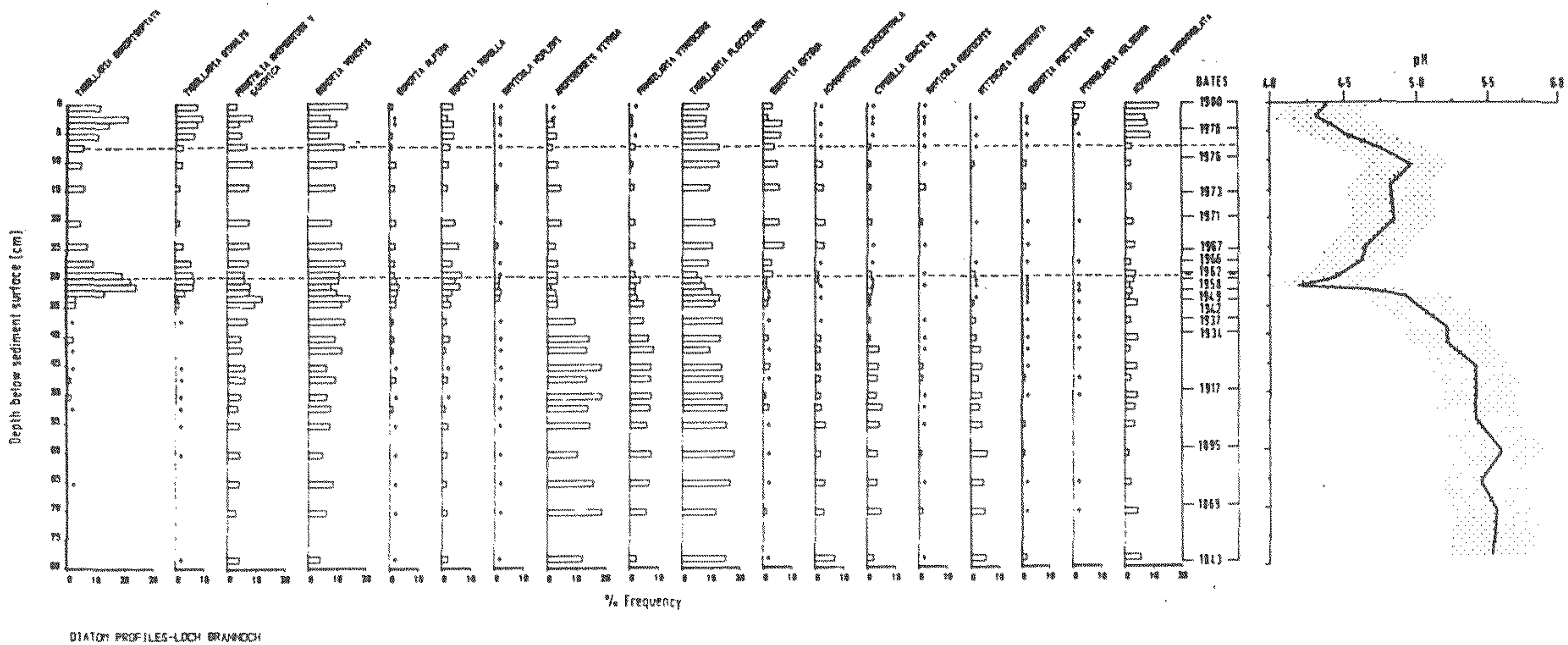

Figure 14 Diatom diagram for Loch Grannoch, as for Fig. 2 but including dates of pre-afforestation ploughing in 1961 and 1977 indicated by the dashed lines (---).

THE INWASH OF CATCHMENT DIATOMS AS A SOURCE OF ERROR IN THE SEDIMENT BASED RECONSTRUCTION OF pH IN AN ACID LAKE[1]

Diatom analysis has been used to identify recently acidified lakes in many areas (Berge 1979, Renberg and Hellberg 1982, Davis *et al*. 1983, Flower and Battarbee 1983, Battarbee 1984). In all cases some method of pH reconstruction has been employed, either based on the Index system of Nygaard (1956) or on multiple regression analysis (Davis and Berge 1980, Davis and Anderson in press, Charles 1982). Percentage analyses only are required to generate pH reconstructions but calculations of diatom concentration and diatom accumulation rates can provide important additional palaeoecological information (Battarbee 1973, 1978a, b). Here we show how calculations of diatom accumulation rate have helped us to differentiate between allochthonous and autochthonous diatom input to the sediment of an acidified lake in Scotland. The results have allowed us to revise and re-interpret the recent pH history of the lake.

Loch Grannoch is an acid loch in Galloway, S.W. Scotland. It has an area of 114.3 ha and a maximum depth of 21.1 m. It is situated at an altitude of 210 m on granitic rocks that are mainly covered by till and blanket peat. Although the loch formerly sustained an important recreational trout fishery it now has a pH of 4.4-4.9 and fish stocks have reportedly declined rapidly in recent years (Harriman and Morrison in press). In 1961 the eastern half of the catchment was deep ploughed prior to afforestation in 1962. A further portion of the catchment was ploughed and planted in 1976-7. In order to assess whether the present acidity levels of the lake could be ascribed to acid precipitation, afforestation, or a combination of both, we carried out an analysis of a sediment core from the centre of the lake. The results (Flower and Battarbee 1983, and

Fig. 15) show that the lake had already become acidic (pH <5.0) before ploughing and afforestation. However, after afforestation the pH, as inferred from Renberg and Hellberg's index B (1982) unexpectedly increased (Fig. 15) as the percentages of the two most important acidobiontic taxa in the flora, Tabellaria quadriseptata Knudsen and T. binalis (Ehr.) Grun. decreased. There are three main alternative hypotheses that might explain the inferred pH change. First, the eroding soils had a temporary neutralising effect on the loch water and caused a decrease in the growth of acidobiontic diatoms; second, the erosion brought a significant quantity of diatoms into the lake from the catchment and depressed the percentages of acidobiontic Tabellaria recorded in the sediment, but had little effect on the water quality of the lake and the growth of these taxa within the lake; and third, the observations are a result of some combined effect of both these processes. These hypotheses can be evaluated using the diatom accumulation rate data and the fact that T. binalis and T. quadriseptata, the two main acidobiontic taxa found in the lake, have not been found growing in the catchment whereas almost all other taxa found in the catchment are also found in the lake and are mainly acidophilous (cf. Table 6). The predominance of acidophilous taxa over acidobiontic taxa in the more acid blanket peat environment of the catchment seems paradoxical. It may be that tolerance of desiccation is more important than pH in these situations. Hypothesis 1 predicts a substantial decrease in the accumulation rate of Tabellaria binalis and T. quadriseptata in the sediment during this period of erosion, hypothesis 2 predicts no significant decline in the accumulation rate of these taxa, but an increase in the accumulation rate of the total excluding them, and hypothesis 3 predicts something of both.

The sediment accumulation rate of the core was calculated from ^{210}Pb analysis using the constant rate of supply model of Oldfield et al. (1978).

Results are shown in Fig. 16. A sudden decline in ^{210}Pb concentration occurs at a level of 29-30 cm and is dated to about 1961, the date of catchment ploughing as determined from forestry records. Figure 16 also shows variations in the rate of sediment accumulation. The rise above 30 cm is clearly associated with ploughing and subsequent erosion. However, prior to the afforestation period there is a sharp decline in sediment accumulation rate from about 45 cm to 30 cm, from 1920-1960.

Diatom concentration analysis followed Battarbee and Kneen (1982) and results are shown in Fig. 17. The values for total diatom concentration in the lower half of the core fluctuate from $7-17 \times 10^7$ cells g dry wt^{-1}. A very sharp decline in concentration occurs at 29-30 cm at the same level as the decline in ^{210}Pb concentration (Fig. 16), almost certainly the result of dilution by allochthonous material. Above this level diatom concentrations are consistently low until the top 8 cm where there is a substantial increase.

Fig. 17 shows the diatom accumulation rate. High values occur between 50 and 40 cm. From 40-30 cm there is a rapid decline to a minimum at 30 cm. This decline parallels the decline in sediment accumulation rates (cf. Fig. 15) and occurs coincidentally with the floristic evidence for the beginning of acidification (the decline of <u>Anomoeoneis vitrea</u>, Fig. 15). Although analyses of biogenic silica have not been carried out to estimate the diatom silica fraction of the sediment, the concentration of diatoms ($>7 \times 10^7$ g dry wt^{-1}) is sufficiently large to suggest that the decline in sediment accumulation rate is in large measure due to the decline in the accumulation of diatoms, possibly as a consequence of acidification (cf. Almer et al. 1974). The minimum value of 0.15×10^7 cm^{-2} yr^{-1} occurs at 29 cm at the point where catchment ploughing began. Above this there is a substantial increase to values of about 0.5×10^7 cm^{-2} yr^{-1} between 25 cm and 8 cm and the uppermost three levels show further increase.

The higher levels in diatom accumulation after the first ploughing episode from 30 to 10 cm indicate either that diatom production was higher during this period, consistent with hypothesis 1, or that up to three times as many diatoms were being introduced from the catchment in eroding peats, consistent with hypothesis 2, or that more diatoms were being derived from both sources (hypothesis 3).

If the accumulation rates of Tabellaria binalis and T. quadriseptata are compared to the total diatom accumulation rate and to their percentage contributions to the total (Fig. 17) these hypotheses can be evaluated. First, the acidification process responsible for the large increase in the percentages of T. binalis and T. quadriseptata (from 35-30 cm) caused only a small increase in the accumulation rate (growth) of these species, indicating that the reason why they become dominant is related more to the decline in other taxa and in total diatom production than in the competitive expansion of acidobiontic Tabellaria spp. Second, after the inwash of eroded material from the catchment, that causes a sudden decline in the diatom concentration at 30 cm (Fig. 17) but an increase in diatom accumulation rate (Fig. 17), the percentages of T. binalis and T. quadriseptata decline rapidly. However, their accumulation rates remain constant implying no decline in their productivity in the lake and suggesting strongly that the decline in percentages is due to the addition of diatoms from the catchment, an observation inconsistent with hypotheses 1 and 3.

The change in diatom concentration, accumulation rate and Tabellaria percentages between 5 and 8 cm below the sediment surface appears to be associated with the second ploughing and planting event in the catchment in 1976-7. However, in this second event, the relationship between the percentage and accumulation rate data differs from that recorded in the first. Rather than a further depression in concentration (Fig. 17) there is a large increase in diatom accumulation rate and both percentages and accumulation rates of T. quadriseptata and T. binalis increase strongly. The continued high level of post-1977 sediment accumulation rate (Fig. 16) indicated that this period of ploughing caused erosion but, despite the lower value of T. quadriseptata at the 0-1 cm level, did not cause a significant depression of the two Tabellaria taxa. Indeed, there is a very marked expansion in the accumulation rate and presumably production of Tabellaria in the lake. Since the total diatom accumulation rate also expands strongly over the last six years there is a suggestion that diatom production as a whole has increased. The reason for this is not clear but one possibility is that fertilisers applied to the newly planted trees and to the more mature eastern forest in 1974 have carried into the lake causing nutrient enrichment.

Although the explanation for the changes associated with the second phase of planting have not been fully established, it is clear that the probable reason for the decline in acidobiontic Tabellaria percentages in the early 1960s is caused by the in-wash of diatoms from the catchment (hypothesis 2) rather than a significant change in the water quality of the lake (hypotheses 1 and 3). The upturn in inferred pH (Fig. 15) from about 30 cm is consequently an artifact and the lake has probably had a pH of <5.0 since 1962.

These data indicate that in oligotrophic acid lakes that have extensive peatlands in their catchments a considerable quantity of diatoms found in the lake sediment may be derived from the catchment if the peatlands have been disturbed by ploughing. Errors in pH inference of lake-water are likely to ensue. Moreover, since many species of periphyton in acid lakes can also be found in terrestrial peatlands, inwashed species from the catchment can pass unrecognized in routine sediment analysis. In some situations diatoms from allochthonous sources might be readily identified. The occurrence of fragments of the Cretaceous marine diatom *Stephanopyxis* in lake sediments in Minnesota and South Dakota (e.g. Haworth 1972) cannot be confused with the autochthonous modern flora in that region. Also eu-terrestrial diatoms can sometimes be recognised in late-glacial sediments, possibly as evidence for solifluction (Haworth 1976). However, problems arise when the species inwashed from the catchment can also be found growing in the lake. It may be possible, as in this example, to identify these by comparing percentage and accumulation rate data. In other cases it might be necessary to monitor stream inputs to assess the quantitative significance of such sources. In all cases this study underlines the importance of calculating diatom concentration and diatom accumulation rates to aid the interpretation of percentage data.

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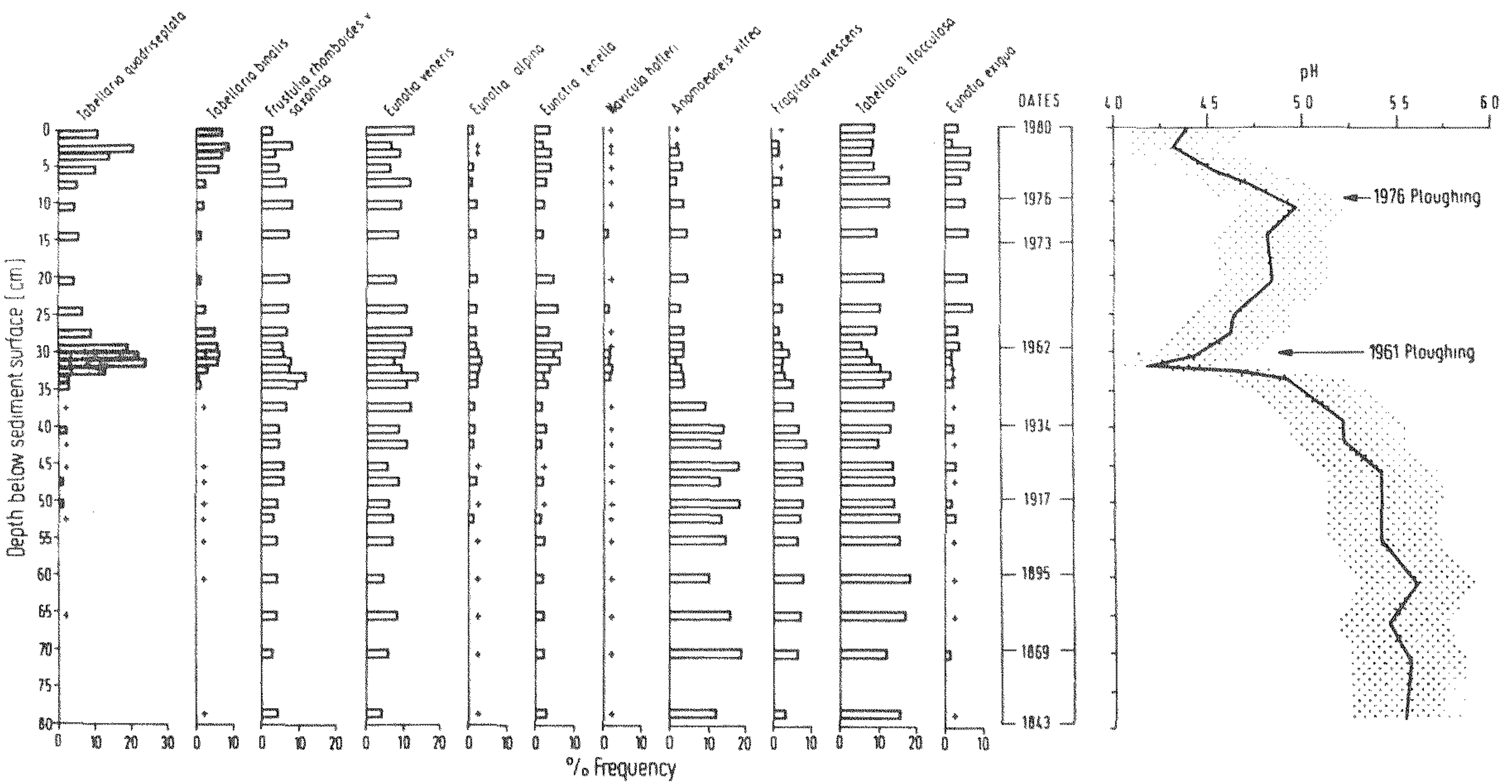
Table 6 Comparison of catchment, lake and sediment diatom flora.

	Catchment		Lake		Sediment
	Epilithon	epiphyton	Epilithon	Epipsammon	Surface Sediment
	%	%	%	%	%
Tabellaria quadriseptata Knudson	-	-	10.3	-	11.7
T. binalis (Ehr.) Grun.	-	-	2.6	6.9	7.6
T. flocculosa (Roth) Kütz.	6.2	16.5	9.9	+	5.3
Eunotia veneris (Kütz.) O. Müll.	71.1	39.0	9.9	+	13.8
E. exigua (Breb.) Rabh.	5.8	2.7	-	-	3.3
E. lunaris v. *subarcuata* (Naegeli) Grun.	-	6.4	-	-	+
E. denticulata (Breb.) Rabh.	- -	3.7	-	+	+
E. pectinalis (Kütz.) Rabh.	+	2.8	+	-	+
E. pectinalis v. *minor* (Kütz.) Rabh.	2.2	+	7.6	16.2	3.5
E. tenella (Grun.) Hust.	-	-	2.3	22.3	3.8
Peronia fibula (Breb. ex Kütz.) Ross	2.7	14.2	-	-	+
Achnanthes austriaca Hust.	-	-	2.3	3.1	+
A. recurvata Hust.	-	-	3.6	26.8	+
A. marginulata Grun.	-	-	20.5	21.3	11.5
Anomoeoneis brachysira v. *thermalis* Nov. comb.	+	3.2	13.2	-	2.9
Frustulia rhumboides v. *saxonica* de Toni	+	+	7.0	+	3.2
Navicula subtilissima Cleve	-	-	+	-	2.7
Pinnularia appendiculata (Agardh) Cleve	+	-	2.0	-	2.9
P. subcapitata Gregory + *P. hilseana* (Janisch) Müll.	6.2	-	3.3	-	4.0



LOCH GRANNOCH - SELECTED DIATOM PROFILES

Figure 15 Diatom diagram for Loch Grannoch showing percentage frequency of selected taxa, selected ^{210}Pb dates and inferred pH values using Index B of Renberg and Hellberg (1982). Stipple on either side of pH curve indicates ±0.3 pH unit standard error for the method (based on Flower and Battarbee 1983).

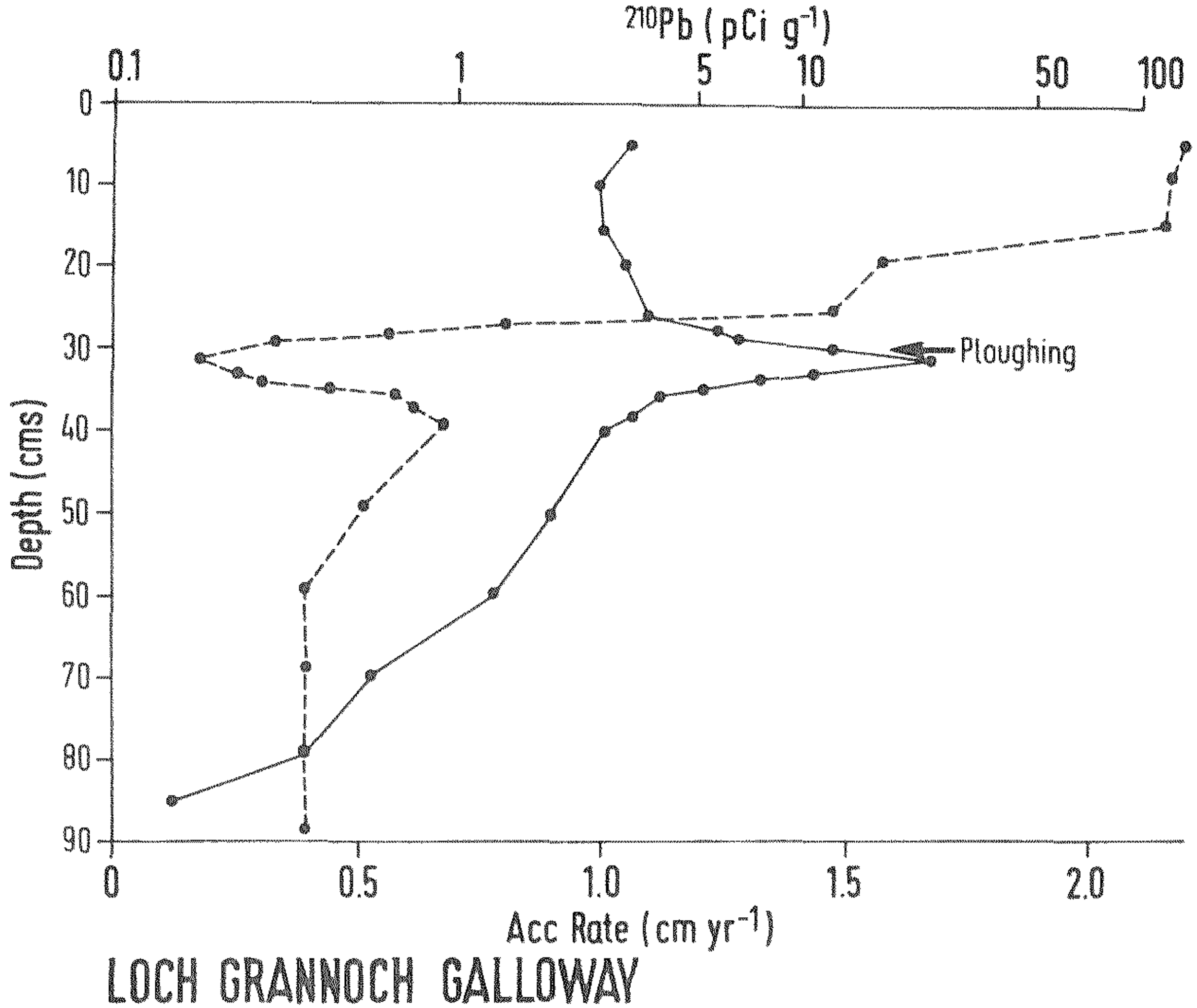


Figure 16   Unsupported ^{210}Pb concentration and sediment accumulation rate for a Loch Grannoch core. ——o—— = Unsupported ^{210}Pb, - - - o - - - = Accumulation rate.

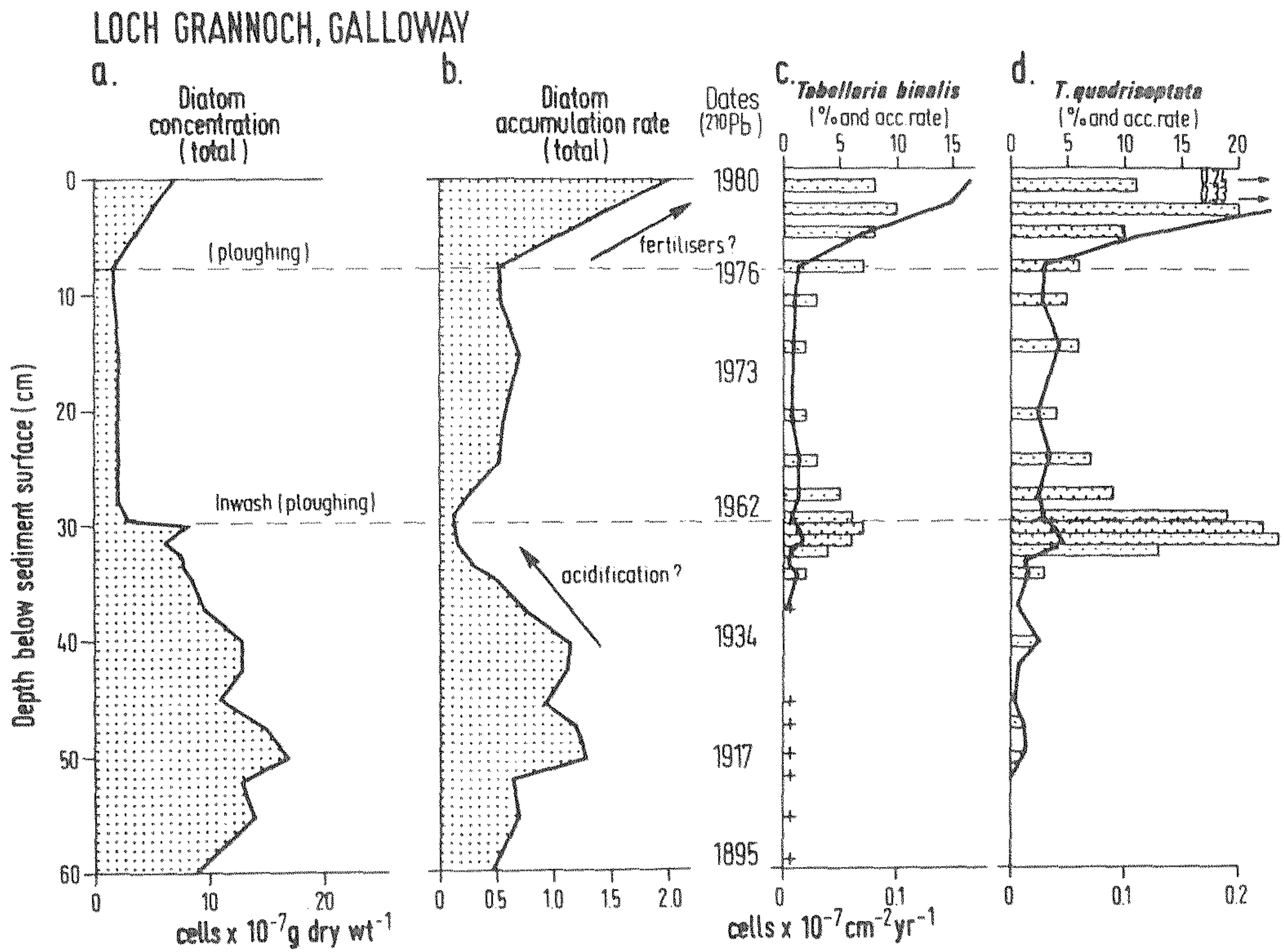


Figure 17 Diatom data for a Loch Grannoch core (a) total diatom concentration (b) total diatom accumulation rate (c) Tabellaria binalis % frequency (histogram) and accumulation rate (curve) (d) T. quadriseptata % frequency (histogram) and accumulation rate (curve).

DIATOM DIAGRAMS AND pH RECONSTRUCTION FOR L. ENOCH,

L. VALLEY, L. DEE and L. SKERROW

Diatom diagrams for Loch Enoch, Loch Valley, Loch Dee and Loch Skerrow are shown in Figs. 18-21. These diagrams have been constructed showing taxa that increase in relative frequency at the left through to those that decrease on the right. ^{210}Pb dates and a reconstructed pH curve using the Index B method are also shown.

Loch Enoch and L. Valley have non-afforested catchments (Figs. 1,2 & 4), are situated at high elevation (Table 1) almost entirely on granitic rocks in the Merrick area and have present pH values in the range 4.5-4.7 (Table 2). L. Dee and L. Skerrow have partially afforested catchments that are situated on the margin of non-granitic rocks, and they have low altitudes and higher pHs (5.2-5.3) than lochs Enoch and Valley. They differ from each other in that afforestation is older and more extensive round L. Skerrow (e.g. Table 1), L. Skerrow is situated on the Cairnsmore granite, and the eastern shore inflows of this loch have been disturbed by railway construction works in the mid-nineteenth century.

1. Loch Enoch (Fig. 18)

The core from L. Enoch was taken from the deepest part of the S.E. basin at a depth of 21 m. The core was 70 cm long and the ^{210}Pb dating shows that the 1800 AD level is about 35 cm below the sediment surface suggesting that the diagram (Fig. 18) covers a time period from about 1600 AD to the present.

The diagram shows that little change occurred until about 1840 when the relative frequency of the acidophilous taxa *Eunotia veneris* and *E. aplina* began to increase and the circumneutral taxon *Anomoeoneis vitrea* began to decline. A progressive acidification took place and an acceleration in this process started about 1930 - indicated by the expansion of the acidobiontic taxa *Tabellaria quadriseptata*, *T. binalis* and *Navicula hőfleri*.

The pH reconstruction summarises these floristic changes indicating a drop in pH of about 1 unit from c. 5.3 in 1800 to c. 4.3 at present. Since the measured pH of the water is usually about 4.5 the model predicts the present pH at about 0.2 pH unit too low.

It is interesting to note that the pre-acidification pH of c. 5.2 indicates that L. Enoch had strongly acidic waters before the recent change and this is consistent with the presence of T. quadriseptata in substantial quantities (3-5%) and the complete lack of planktonic diatoms throughout the time-period of the core.

2. Loch Valley (Fig. 19)

The L. Valley core was taken close to the S. shore of the lake in 15 m. The 70 cm core covers a longer period of time than the L. Enoch core owing to the slower accumulation of sediment. 1800 AD occurs at c. 20 cm suggesting, on the basis of extrapolation that the bottom of the core at 70 cm is dated to about 1350 AD.

The diatom diagram (Fig. 19) shows a situation very similar to L. Enoch with in excess of 500 years prior to 1860 of stable conditions at a pH of about 5.5. This is somewhat higher than the pH of L. Enoch at the same time. It is indicated by the diatom spectra by the considerable importance of Fragilaria virescens, circumvental species, and absence of T. quadriseptata during the period.

The species changes that indicate acidification from about 1860 are almost identical to those for L. Enoch with E. veneris expanding slightly and A. vitrea decreasing. The more rapid acidification over the last 50 years is likewise indicated by the expansion of T. quadriseptata, T. binalis and loss of A. vitrea. The estimation of water pH for the uppermost sediment of 4.6 is almost the same at the mean measured pH of 4.7 and the data indicate a drop of pH of about 1 unit since c. 1860.

The results from this site should be related not only to L. Enoch but also to Round Loch of Glenhead (Fig. 13) where a similar drop in pH of about one unit was observed from about 5.7 to about 4.7. The pH of L. Valley both before and after acidification falls between that for L. Enoch and Round Loch. The diatoms also show that L. Valley is in an intermediate position having no plankton over the last 400 years, as L. Enoch but unlike Round L., and having considerable quantities of *Fragilaria virescens* as Round L. but unlike L. Enoch.

The three lakes are very close together on identical bedrock but are not hydrologically linked. The close correlation between pH/diatom flora and altitude is thus not related to surface water connections but to some environmental parameter correlated with altitude.

3. *Loch Dee* (Fig. 20)

The L. Dee core has a much faster sediment accumulation rate than L. Valley and L. Enoch with unsupported ^{210}Pb present at the base of the core. The core covers approximately the last 120 years. L. Dee is much less acid than L. Enoch and L. Valley and this is reflected by the diatom data especially the absence of acidobiontic species. The lake pH ranges (1981-82) from 5.0 to 6.0 with a mean of 5.3 (Table 2) and Index B predicts 5.6 from the surface sediment diatom assemblage. The diatom assemblage lacks plankton and includes many acidophilous *Eunotia* taxa, but the diagram shows that these taxa were far less important 100 years ago and that prior to 1900 AD there was a significant diatom plankton. Acidification has certainly occurred at this site and the Index B reconstruction suggests that pH values in the 19th century were about 6.1 and that a decline of 0.5 pH units has taken place since then. Part of the catchment was afforested in 1976 but it is clear that the main acidification pre-dates this disturbance.

It is interesting to relate the species changes shown in Fig. 20 to those from the adjacent more acid sites discussed above. There is a continuum in species change from site to site with altitude and with pH. The surface spectra of L. Dee correlate closely with the c. 1900 AD flora of Round Loch of Glenhead, and the surface spectra of the Round L. is very similar to the 1940 AD flora of L. Enoch. These overlapping successions indicate that species changes could be predicted with confidence at these sites were acidification to continue, (and liming at L. Dee to stop). At L. Dee a further slight acidification would lead to a decline in Anomoeoneis vitrea presently the dominant diatom in the sediment assemblage and the acidobiontic diatoms T. quadriseptata and T. binalis would appear. At the Round Loch of Glenhead E. veneris would begin to decline, F. virescens would continue to decline, and T. quadriseptata and T. binalis would continue to increase.

4. Loch Skerrow (Fig. 21)

It was our original intention not to work on L. Skerrow because of possible difficulties in separating the influence of hydrological changes and other changes associated with railway construction works from the influence of acid precipitation and afforestation. However, since the L. Fleet sediment core proved difficult to date we decided to abandon L. Fleet in favour of L. Skerrow. This decision was made towards the end of the contract period so less background information on the contemporary water quality and diatom flora of the lake has been collected, and uncertainty still exists as to the extent of drainage diversions during the 19th century. At present the Grobdale Lane (Fig. 7) draining mainly non-granitic rocks and with relatively high pH values of 5.5-6.5 joins the outflow stream from L. Skerrow a short distance below the outflow and a leat from the river, originally constructed to supply L. Skerrow Halt with water for the railway, runs into the lake at the N.E. corner. All inflows from the afforested, granitic parts of the catchment on the south, west and north (cf. Fig. 7) bring more acidic water to the lake (pH 4.0-5.0).

Fewer chemical measurements are available for this site but they indicate that the pH is sbout 5.2 (Table 2). This is surprising since conductivity and major ion concentrations are considerably higher than at other sites including L. Dee (cf. Tables 2 & 3).

The diatom data are shown in Fig. 21. The core covers a period of approximately 300 years and the data indicate that few changes have occurred during this period. The most important change is the decline in the planktonic diatom Cyclotella kutzingiana at the end of the nineteenth century. This is similar to the decline of C. comensis in L. Dee at about the same time and the earlier decline, about 1700 AD, of C. kutzingiana at the Round Loch. As at the other sites this plankton decline probably represents an acidification of the lake, but unlike the other sites, only a very slight further acidification is indicated as Achnanthes microcephala and Anomoeoneis vitrea decrease gradually. These changes are hardly reflected in the Index B pH reconstruction. Moreover there appears to be a slight increase in pH over the last 10 years represented by the emergence of Asterionella formosa a planktonic, alkaliphilous diatom.

These data are not easy to interpret at this stage but a number of comments can be made. First, there is no evidence whatsoever of any acidification caused by the planting and growth of conifers in the catchment. Second, the decline in plankton in the nineteenth century could be a small change in water quality associated with the diversion of more alkaline inflows from the lake, to acid precipitation, or to a so far unknown factor. Third, the increase in A. formosa may be related to recent liming by anglers. Fourth, the very slight acidification indicated here for a lake partially on the granite suggests that lakes with catchments completely off the granite are unlikely to be biologically acidified. A fifth issue is the lack of agreement between the measured pH of c. 5.2 and the predicted pH of c. 5.9. This is the only site where a significant difference between measured and predicted values has been found. It is difficult to explain although it is clear the diatom data are more in agreement with the conductivity and general chemistry of the lake and with the observation that there are no problems with fisheries. The difficulty, hence, is in explaining why the pH readings are so low when all other chemical and biological data indicate a largely non-acidified system. Further studies that take into account spatial and temporal variations in water quality are needed before further speculation is warranted on this site.

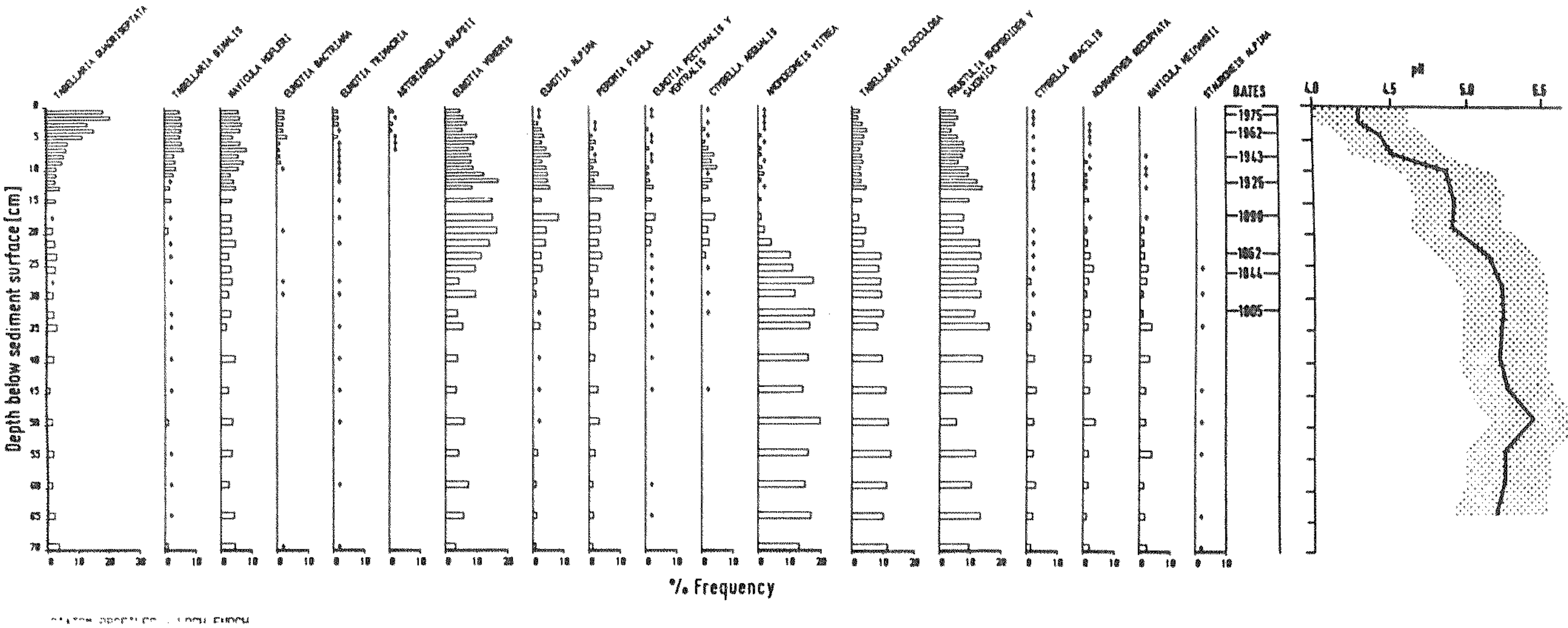


Figure 18 Diatom changes for Loch Enoch.

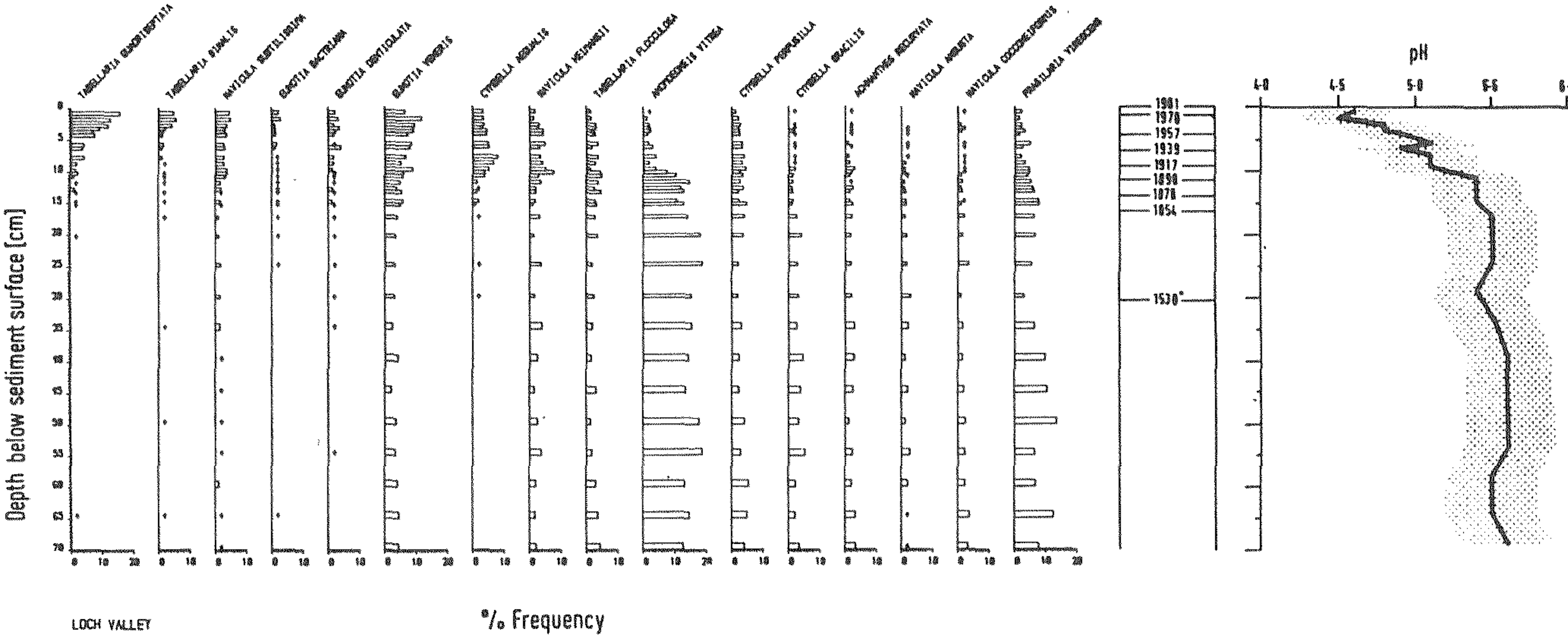


Figure 19 Diatom diagram for Loch Valley.

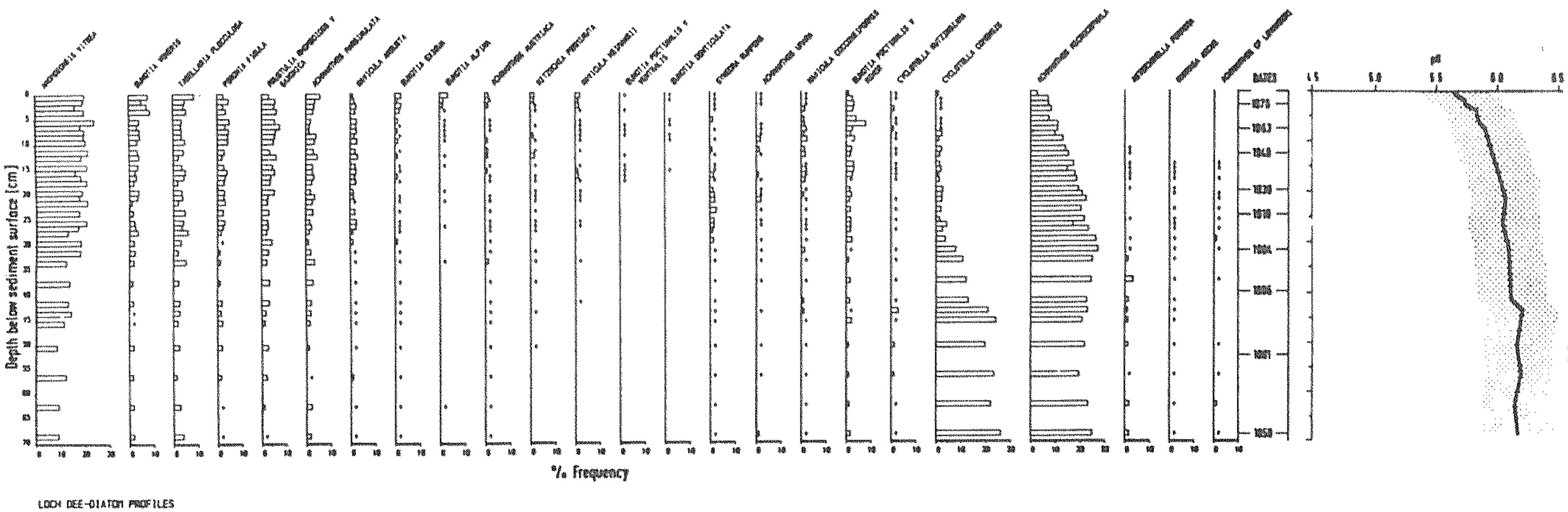


Figure 20 Diatom diagram for Loch Dee.

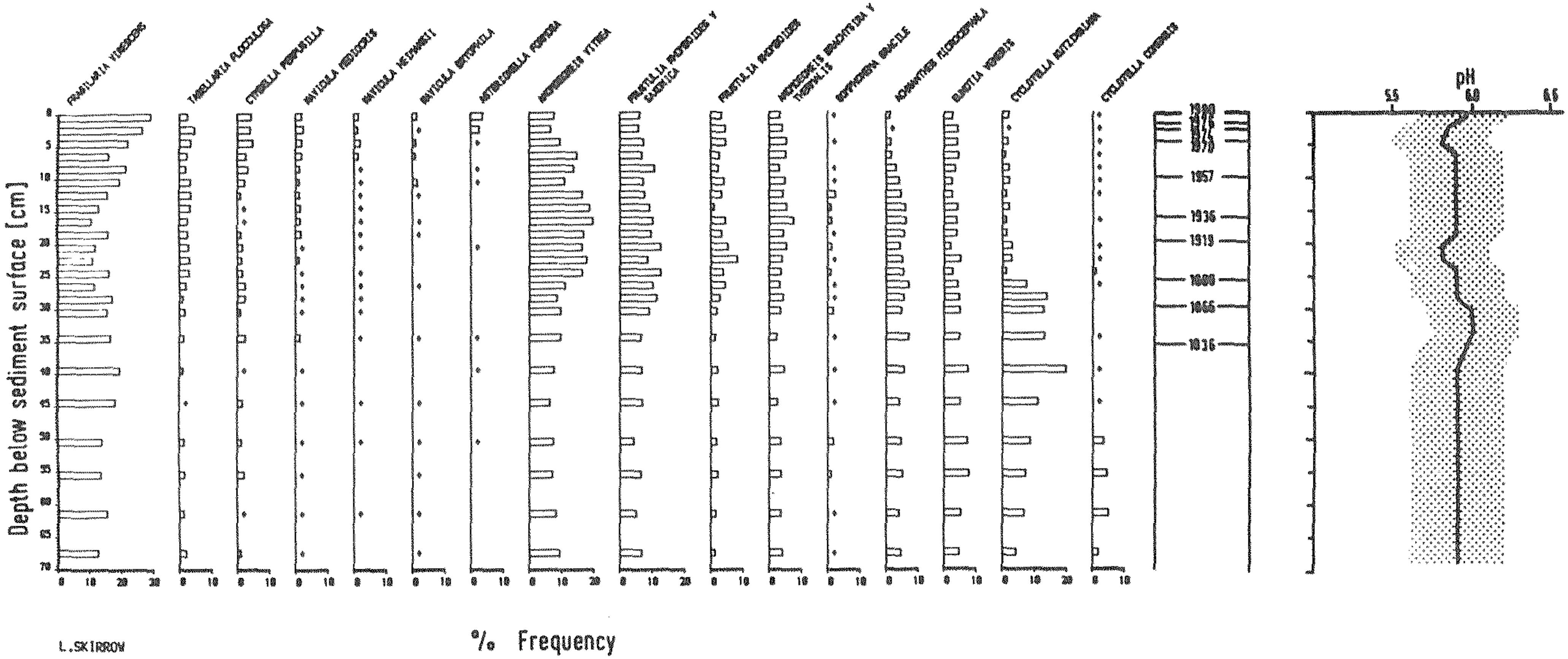


Figure 21 Diatom diagram for Loch Skerrow.

LAKE ACIDIFICATION IN GALLOWAY:
A PALAEOECOLOGICAL TEST OF COMPETING HYPOTHESES

Possible causes of lake acidification in Britain include acid precipitation, heathland regeneration, afforestation, and post-glacial paludification (long-term change). In Galloway, S.W. Scotland, several lakes with non-afforested catchments have acidified by approximately one pH unit since about 1840.[1,2] These observations clearly discount long term change and afforestation as causes of the present acidity. They do not, however, distriminate between the possible effects of heathland regeneration and acid precipitation, since it could be argued that a demise of upland farming in this area leads to an increase in acid heathland communities capable of promoting lake acidification over this time period. Here we use pollen analysis to show that there is no evidence for an increase in Calluna vulgaris, the most important heathland species, over the last 200 years or so and we substantiate the acid precipitation hypothesis by demonstrating substantial increases in the heavy metals Pb, Cu, and Zn in the sediment since about 1800 AD.

Surface water acidification has been variously ascribed to the effects of acid precipitation,[3] land-use change,[4] afforestation,[5,6] and long-term change.[7] The importance of these processes can be assessed using palaeoecological techniques. The last two of these are considered first.

The process of long-term acidification is well known and has been recognised in Scandinavia,[8-11] Britain,[12-15] and North America.[16,17] In Britain this process has been suggested as the cause of contemporary acidity in upland tarns of the English Lake District and that certain lakes were so acidified by 1800 that no further change has occurred since then.[7] Although no examination of this supposition has yet been carried out in the English Lake District it is clear that the present acidity of lakes with granitic catchments in Galloway, S.W. Scotland, cannot be explained in this way where diatom analysis indicates that rapid acidification of these lakes has occurred since about 1840 after long periods of stability.[1,2]

Recent afforestation, although responsible for causing accelerated soil erosion,[18] and other undesirable ecological changes, is also of little significance since lakes with non-afforested catchments have been acidified and lakes with afforested catchments began to be acidified in the late nineteenth and early twentieth century, well before the beginning of afforestation.[1]

A land-use change hypothesis to explain recent lake acidification in Norwegian conditions has been put forward by Rosenquist[4] and has been supported by Krug & Frink.[19] It maintains that acidity generated by soil processes is substantially greater than that received from atmospheric deposition and that an increase in the formation of acid humus in soils as a result of a decline in upland agriculture is likely to be of more significance than an increase in the acidity of precipitation.

In Galloway it might be expected that a similar decline in grazing would lead to the greater dominance of Calluna vulgaris in particular and to a regeneration of heathland communities in general.[20] This hypothesis could be rejected if it could be shown that such a land-use change was

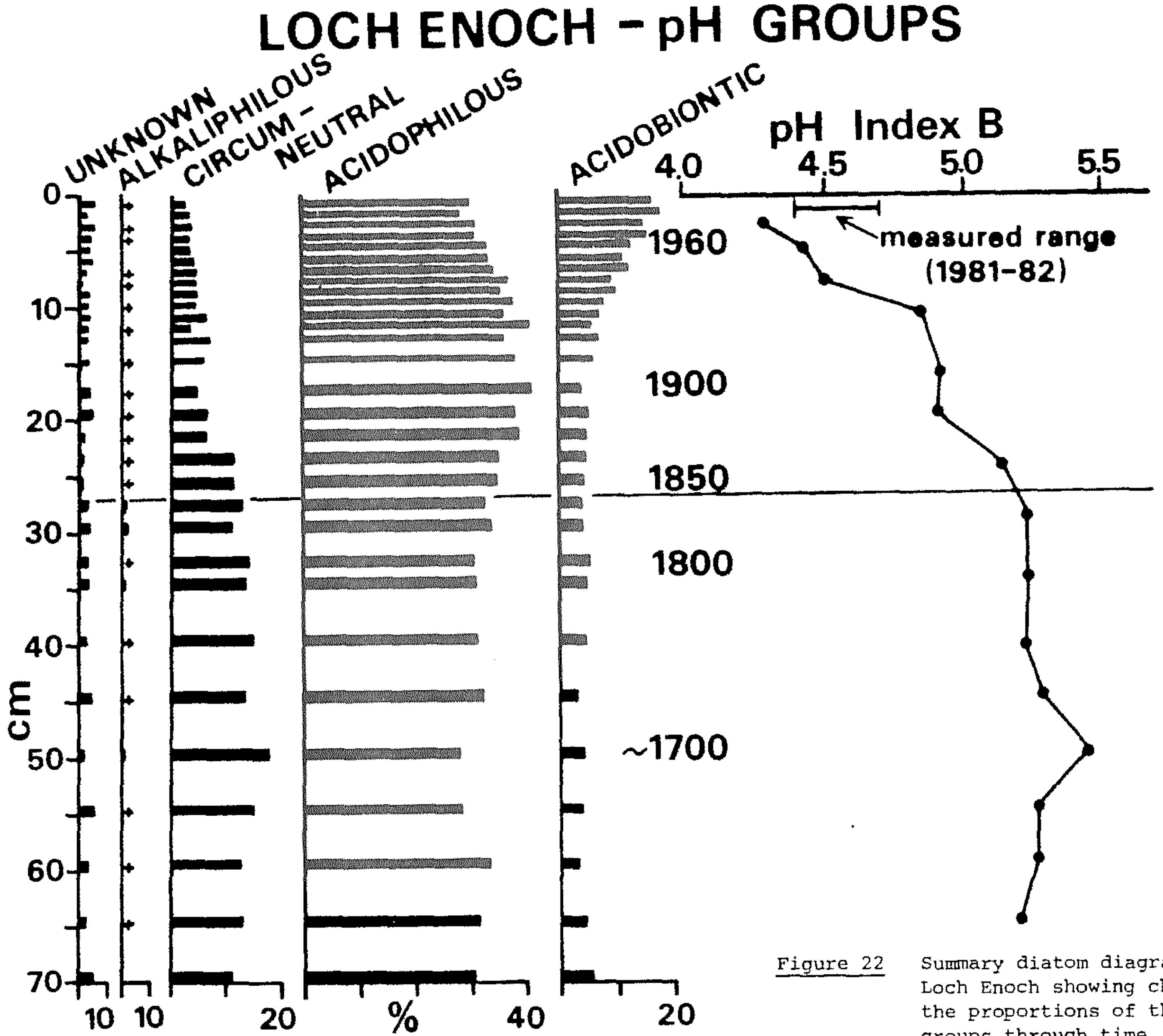


Figure 22 Summary diatom diagram from Loch Enoch showing changes in the proportions of the various pH groups through time.

Table 7 Physical and chemical characteristics for Loch Enoch.

Altitude (m)	493	
Lake area (ha)	50	
Catchment area[a] (ha)	186	
Catchment: lake area	3.7	
Maximum depth (m)	c.36	
Temperature (^{o}C)	7.0 (4.4)	
pH (range)	4.4-4.7	4.5[b]
H^{+} ($\mu g\ \ell^{-1}$)	28.9 (6.8)	32[b]
Na^{+} ($mg\ \ell^{-1}$)	1.3 (0.6)	2.9[b]
Ca^{++} ($mg\ \ell^{-1}$)	0.2 (0.1)	0.4[b]
$A\ell$ ($\mu g\ \ell^{-1}$)	90 (40)	150[b]
Cl^{-} ($mg\ \ell^{-1}$)	3.6 (0.6)	5.2[b]
$SO_4^{=}$ ($mg\ \ell^{-1}$)	2.1 (0.6)	4.3[b]
Conductivity $\mu S (18^{o}C)\ cm^{-1}$	30 (8.0)	36[b]

Numbers in parenthesis indicate standard deviations for the mean values calculated from 8 measurements made between November 1981 and November 1982. (a) catchment area excluding lake area. (b) data from Wright and Henricksen.[34]

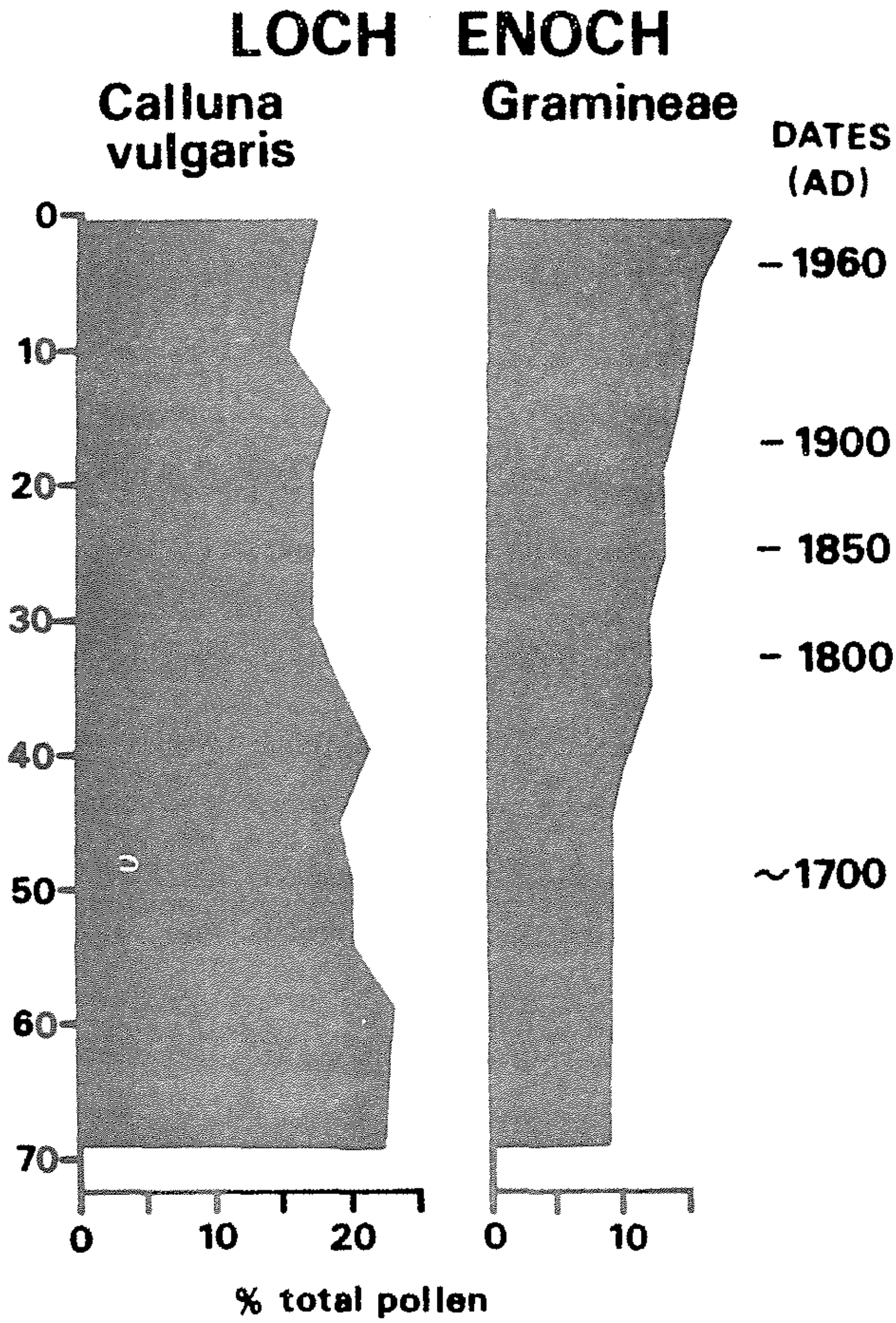


Figure 23. Proportions of Calluna vulgaris and Gramineae pollen from the Loch Enoch sediment core.

absent or was inadequate to explain the observed acidification of the lakes concerned. Rejection would lead to the need to evaluate the acid precipitation hypothesis.

To carry out these tests we have used diatom analysis, pollen analysis, and trace metal analysis of a dated lake sediment core from Loch Enoch. L. Enoch is a large (50 ha), deep (36 m), multi-basin lake situated at an altitude of 493 m on the Merrick in the Rhinns of Kells region of Galloway, S.W. Scotland. The catchment is non-afforested and the vegetation is dominated by Molinia caerulea and Calluna vulgaris.

We obtained a sediment core from the centre of the south-east basin of the lake and determined a linear sediment accumulation rate of about 1 mm yr^{-1} using ^{210}Pb analysis.[18] Diatom analysis shows the onset of recent acidification at about 30 cm, dated to approximately 1840 (Fig. 22). The Index B pH reconstruction[11] gives a ca. 1840 pH of 5.2 and a present pH of 4.3, slightly more acid than the measured range of pH values (cf Table 7). The strong increase in acidobiontic diatoms registered in the diagram (Fig. 22) include Tabellaria binalis (Ehr.) Grun. and T. quadriseptata Knudson, species that typically increase in a similar way at other sites in the region.[1,21]

If acidification were to have been caused by a land-use change in the catchment of the lake according to the hypothesis advanced above we should expect historical records to show a decline in grazing and consequent regeneration and expansion of heathland over roughly the same timescale. Since records of sheep numbers over the last 150 years are available only on the basis of large administrative regions,[22] it is more appropriate to test this prediction using pollen analysis of the lake sediment core itself. Such analysis was carried out using standard procedures[23] on samples selected at 5 cm intervals from the top of the core. The full pollen diagram shows very little change over the last 200-300 years except in the relative

Table 8 Background sedimentary concentrations, fluxes and recent fluxes of Zn, Cu and Pb in a sediment core from Loch Enoch.

	Zn	Cu	Pb
Background concentration, $\mu g\ g^{-1}$	25.4	6.2	74.6
Standard deviation Less than 32 cm depth, n = 12	7.4	4.6	23.3
Background sedimentary flux, $mg\ m^{-2}\ a^{-1}$ Average value for 32 to 61 cm	6.1	1.5	17.8
Recent sedimentary flux, $mg\ m^{-2}\ a^{-1}$ Average value for 0 to 10 cm	35.5	8.3	67.0
Anthropogenic sedimentary flux, $mg\ m^{-2}\ a^{-1}$ Recent - background flux	29.4(83)	6.8(82)	49.2(73)

Calculations made using trace metal concentrations (Fig. 24) and sediment wet density, dry weight and accumulation rate (Flower & Battarbee[24]). Numbers in parenthesis indicate % anthropogenic flux of total metal influx.

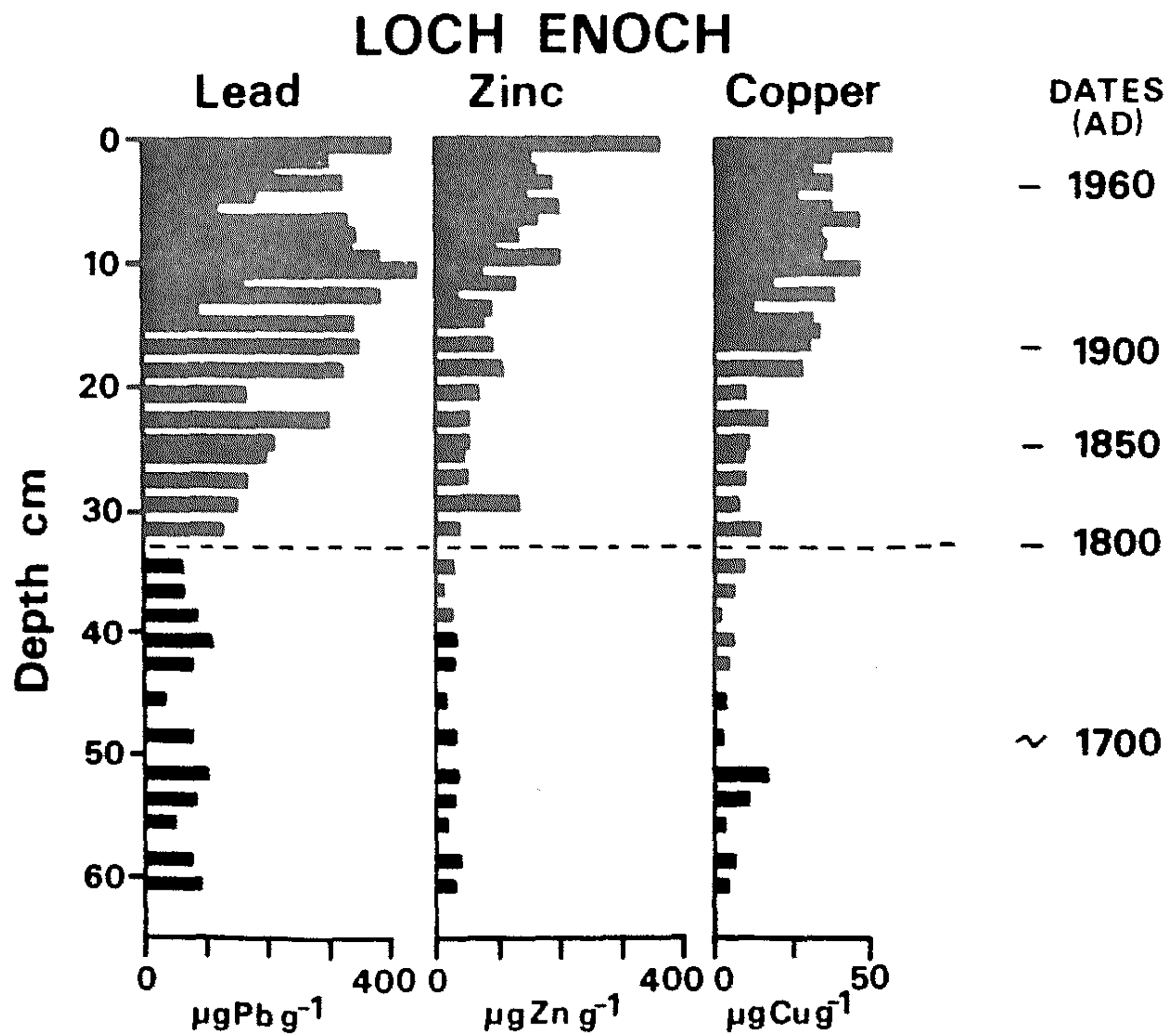


Figure 24 Concentrations of Pb, Zn and Cu from the Loch Enoch sediment core.

proportions of Calluna vulgaris and Gramineae, the taxa of greatest interest (Fig. 23). However, rather than an increase in C. vulgaris, as would be expected were the land-use hypothesis valid at these sites, there is a gradual decrease of this species and a reciprocal expansion of Gramineae. Similar results have been obtained from the analysis of cores from other lakes in the area. These results are not unexpected since they are in accord with contemporary observations showing that all the non-afforested lake catchments in this area are grazed and regularly burnt over, a process that in the west of Scotland favours Molinia caerulea and keeps Calluna vulgaris young and palatable for both cattle and sheep.[24]

We have been able to reject three alternative hypotheses for the recent acidification of L. Enoch. The acid precipitation hypothesis remains. If not disproved this could be seriously questioned were we not able to show that L. Enoch and other similar Galloway lakes had received atmospheric pollutants that might be associated with industrial emissions. To do this we carried out trace metal analysis of the sediments (Fig. 24).

Total sedimentary Zn, Cu and Pb concentrations were determined by flame atomic absorption after $HF/HNO_3/HClO_4$ digestion. The precision of the results are 4.7 μg Zn g^{-1}, 0.9 μg Cu g^{-1} and 3.8 μg Pb g^{-1}, n=14. The background concentrations (Table 8) show that there are no geochemical anomalies in the catchment. The slightly lower Cu and Zn and somewhat higher Pb concentrations, compared to the mean for freshwater sediments,[25] are due to the granites in the catchment.[26] The increased concentrations (Fig.24) and sedimentary fluxes (Table 7) can be accounted for by increased deposition from the atmosphere,[27] a situation common in remote and rural lakes.[28-30] It has been shown that atmospheric contamination of lake waters by trace metals is correlated with acidification,[31,32] and atmospheric contamination of sediments by trace metals is correlated with sedimentary evidence of acidification.[33,27,28] This latter relationship holds true for

L. Enoch and all these relationships are consistent with expectations derived from the acid precipitation hypothesis.

We cannot prove that an increase in acid deposition was responsible for the acidification of Loch Enoch and similar lakes in Galloway but we have shown that alternative hypotheses, as presently formulated, are inadequate. Land-use change may have a role to play in other areas but at the present time it is clearly not applicable to the Galloway sites. For this hypothesis to be tested by palaeoecological means examination of an acidified site where heathland regeneration has occurred concurrently in an area of low acid deposition is required.

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CONCLUSIONS

Our results show that:

1. 5 of the 6 lakes have become significantly more acid, by between approximately 0.5 and 1.2 pH units, within the last 130 years since 1850.

2. One lake, L. Skerrow, shows no significant change in pH but there has been a change in the diatom flora that indicates slight acidification since about 1880.

3. The beginning of recent acidification varies from lake to lake and from about 1850 to about 1925.

4. Acidification in the nineteenth century was slow, but there was an acceleration in the twentieth century between approximately 1930 and 1960.

5. Ploughing prior to afforestation caused substantial soil erosion judging from accelerations in the rate of sediment accumulation following the time of ploughing, but

6. Evidence for any acidification associated either with the effects of ploughing or the effects of a growing forest is obscured at these sites because of sedimentological problems caused by soil erosion.

7. Afforestation, however, cannot be the most important factor causing acidification since we compared three unafforested sites with three afforested sites and the most acidified lakes are those without afforestation Moreover acidification of two of the afforested sites, L. Grannoch, and L. Dee, occurred well before the afforestation of their catchments.

8. Upland land-use changes involving a decline in sheep grazing and a cessation of moorland burning cannot be the explation of acidification at these sites since pollen analysis carried out at each site shows an increase, rather than a decrease, in the Gramineae :Calluna pollen ratio. It would be expected that an increase in Calluna would be found were there to have been a significant decrease in sheep grazing and heather burning.

9. Long term leaching cannot be the cause of the observed acidification since adification has occurred rapidly and in very recent (in a geological sense) times, and after long periods of stable conditions.

10. Heavy metal analysis (Pb, Zn, Cu) of the sediments from L. Enoch, the most isolated and the most acid lake of the six, shows increases through time in the sediment profile that parallel the diatom evidence for acidification. Such increases are usually thought to be evidence of increases in atmospheric pollution.

11. Since our data sustain none of the alternative hypotheses we infer that the acidification of these lakes is most likely the result of an increase in the acidity of precipitation since 1850. At least this is the one hypothesis that we have so far been unable to disprove.

12. It is likely that our conclusions apply to the other acid lakes situated on the upland Galloway granites. On the other hand it cannot be assumed at this stage that lakes in Galloway not on the granite and lakes elsewhere in the United Kingdom have been or will become acidified.

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PALAEOECOLOGY RESEARCH UNIT

WORKING PAPERS

No 1 Patrick, S. & Battarbee, R.W. 1981 The influence of sanitary and other social changes on the eutrophication of Lough Erne since 1850: Project introduction and a consideration of the potential role of metabolic wastes. 43pp.

No 2 Battarbee, R.W. 1983 Diatom analysis of River Thames foreshore deposits exposed during the excavation of a Roman waterfront site at Pudding Lane, London. 18pp.

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