

**ENVIRONMENTAL CHANGE RESEARCH CENTRE
UNIVERSITY COLLEGE LONDON**

RESEARCH PAPERS

NO. 7

*ACIDIFICATION IN THE CAIRNGORMS AND LOCHNAGAR: A PALAEOECOLOGICAL
ASSESSMENT*

**Jones, V.J., Battarbee, R.W., Appleby, P.G., Richardson, N., Rippey, B., Rose, N.L. &
Stevenson, A.C**

February 1993

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1 INTRODUCTION

Sensitive lakes in areas of the United Kingdom with moderate to high sulphur deposition have been acidified since the middle of the nineteenth century (Battarbee *et al.* 1988). Regions such as Galloway, south west Scotland (eg. Flower and Battarbee 1983, Flower *et al.* 1987), Wales (eg. Battarbee *et al.* 1988, Fritz *et al.* 1990), Cumbria (eg. Battarbee *et al.* 1988, Atkinson and Haworth 1990), and Rannoch Moor in the central Scottish Highlands (eg. Flower *et al.* 1988) have been affected. This study extends the geographical survey of lake acidification to the Cairngorm and Lochnagar regions of north east Scotland (Figure 1). The Cairngorms and Lochnagar are areas of considerable conservation value, forming the largest single area of land over 1000 m in the UK. The Cairngorm mountain plateau is a National Nature Reserve, noted for its alpine flora and fauna, whilst the Lochnagar range is a Scottish Wildlife Trust reserve.

A secondary aim of the study was to evaluate the "land-use" hypothesis (eg. Rosenqvist 1977, 1978, 1981) as a mechanism for lake acidification by examining high altitude sites with no active land-management. Sites selected are all remote, lie above the tree line and have undisturbed catchments.

Lochnagar and the Cairngorms are situated on sensitive granite geology (Kinniburgh and Edmunds 1986, Wells *et al.* 1986) in an area of moderate acid deposition (c. 0.95 g S yr⁻¹). It can be predicted that sensitive lakes in this area (those having Ca²⁺ values of <60 µeq l⁻¹) will have acidified (Battarbee 1989).

2 THE STUDY SITES

Five lakes have been investigated, three from the Lochnagar area (Lochnagar, Loch nan Eun and Dubh Loch - Figure 1) and two from the Cairngorms (Lochan Uaine and Loch Coire an Lochan - Figure 1). Results from Lochnagar have been published elsewhere (Battarbee *et al.* 1988, Patrick *et al.* 1989, Jones *et al.* 1992).

The landscape of the Cairngorms and Lochnagar has been heavily influenced by ice action; the area was a major centre for the accumulation and dispersal of ice during the last glaciation. The highest sites; Lochan Uaine, Loch Coire an Lochan and Loch nan Eun were formed as moraine dammed corrie lakes. Indeed, Loch Coire an Lochan and Lochan Uaine are two of the highest lakes in the British Isles (at 998 m and 950 m respectively). Snow generally occupies the catchments to a varying extent between November and May. The lakes freeze each winter and partial ice cover may last from November to June in many years, with a complete ice cover between January and April which can approach 1 m thickness by early March. Snow and ice melt therefore comprise an important input to the lakes.

The lowest altitude site - Dubh Loch, lies in a deeply incised glacial trough, with a deposit of hummocky drift over 30 m thick lying around the outflow (Sissons and Grant 1972).

Geology

Both the Cairngorms and Lochnagar ranges are situated on granite bedrock (Figure 1). The Lochnagar granitic complex forms a circular area of about 170 km². It is a fine to medium grain granite consisting of oligoclase, quartz and biotite, usually grey in colour with a pink tinge due to the alkali-feldspars (Barrow and Craig 1912). The ring is zoned, from an early diorite unit at the periphery, through a medium-grained adamellite, to a fine-grained adamellite at the centre (Oldenshaw 1974). Dubh Loch lies at the perimeter of the mass, and the catchment includes some diorite (which is richer in MgO, CaO, TiO₂, P₂O₅, Sr and Ba) (Harrison pers comm.). Lochnagar lies on the medium-grained adamellite and Loch nan Eun lies on the second phase of the fine-grained adamellite (Oldenshaw 1974).

The Cairngorm granite covers an area of about 360 km² and consists of two important units, the main granite which is a more or less even grained microcline-oligoclase-biotite granite and the porphyritic granite which has conspicuous feldspar phenocrysts (Harry 1965). Lochan Uaine and Loch Coire an Lochan are situated on the deep red main granite.

In terms of both mineralogy and major element chemistry the Lochnagar and Cairngorm granites are identical. They are both biotite granites with SiO₂ contents between 70-75%, Ca <2%, Al₂O₃ <15% and Na₂O around 4%. Trace element characteristics are however quite different. The Cairngorm granite is markedly richer in Rb, U, Th, Y, Nb, Li, F, Be, and rare earth metals, and poorer in Sr and Ba (Harrison pers. comm.).

Vegetation, land-use and soils

Vegetation description mainly follows vegetation maps produced by the upland survey of the Nature Conservancy Council (Horsfield pers. comm.). The vegetation types described are based on the work of McVean and Ratcliffe (1962), with additions from Birks and Ratcliffe (1980). The nomenclature of the higher plants follows Clapham *et al.* (1981). Soils and land-use descriptions are based on 1:50,000 soil survey maps and Billett (pers. comm.).

Loch nan Eun is characterised by alpine soils, with rankers and lithosols on the steep slopes. The catchment vegetation consists of dwarf shrub heath communities (*Calluna vulgaris* and *Vaccinium myrtillus*) on the flatter areas, with snowbed *Nardus stricta* grassland and species poor *Deschampsia caespitosa* grassland on the steeper slopes.

The catchment of Dubh Loch is dominated by free-draining, predominately alpine soils (>75%), with rankers on the north side of Broad Cairn and Creag an Dubh Loch. Deep peat also occurs in the wetter depressions above and below Dubh Loch and Loch Buidhe. The vegetation is mainly a *Calluna - Eriophorum* blanket bog community, with dry *Calluna* heath in the better drained areas. Species poor *Deschampsia caespitosa* grassland occurs on

the scree slopes on the southern side of the catchment.

Loch Coire an Lochan and Lochan Uaine are characterised by alpine heath and snowbed communities. At Loch Coire an Lochan snowbed *Nardus* grassland and snowbed *Vaccinium myrtillus* heath occur and an *Empetrum nigrum* - *Rhacomitrium* heath community is also important, with *Deschampsia caespitosa* grassland on the scree slopes.

The high altitude and remoteness of these sites means that active land-management has been restricted. Three of the sites (Lochan Uaine, Loch Coire an Lochan, and Loch nan Eun) lie well above the limit of sheep grazing in the region (Scottish Development Department 1967). The lower altitude site, Dubh Loch, is however subject to rather more grazing pressure. Red deer range across the catchments in the Lochnagar area, but stalking in the area is now rare (Clark 1981). The catchments of Lochan Uaine and Loch Coire an Lochan are so high, rugged and remote that it is unlikely that they have ever been used for deer stalking or grouse shooting.

Water and atmospheric quality

Table 1 gives water quality data for the lakes. The Lochnagar sites have lower pHs (about pH 5.0) and lower alkalinities ($0.0-1.37 \mu\text{eq l}^{-1}$) than the Cairngorm lakes (pH above 5.5, alkalinity $>13 \mu\text{eq l}^{-1}$). Lochan Uaine has a rather unusual chemistry with high Ca^{2+} and Mg^{2+} values, perhaps indicating the presence of alkalinity sources in the catchment, it also has high levels of Na^+ and Cl^- . Ca^{2+} levels give a good indication of the susceptibility of waters to acidification (Battarbee 1989, Battarbee *et al.* 1988) and the low levels ($<30 \mu\text{eq l}^{-1}$) at the sites (with the exception of Lochan Uaine) suggest they are very sensitive to acidification. Except for Dubh Loch the lakes are very clear with low total organic carbon (TOC) values and secchi-disc depth values of >6 m (Patrick pers. comm.). The higher TOC level at the Dubh Loch is probably a reflection of the greater proportion of peat in its catchment. Differences in SO_4^{2-} concentrations and pH values between the five lakes probably reflect differences in sulphur retention in the catchments and lake sediments together with differences in alkalinity production in both catchments and sediments (cf. Cook *et al.* 1987). Despite these differences, SO_4^{2-} concentrations are relatively high, reflecting the moderate level of sulphur deposition in the area (see below).

Regional surveys of the fishery status of lakes in the area have been made. At Lochnagar trout were only found in the 5-7 year classes, which may indicate recruitment problems (Harriman and Morrison 1989). Loch Coire an Lochan and Loch Uaine were found to be fishless. Dubh Loch seems to have a relatively healthy fishery with trout of a range of ages (Morrison pers. comm.). Loch nan Eun has not been sampled. The absence of fish and the occurrence of recruitment problems could be due to the high levels of acidity at these sites, but it is also likely that physical conditions such as the long period of ice cover, the lack of suitable spawning sites, and the absence of clearly defined outflow streams play an important part in determining their fishery status.

Data concerning atmospheric quality (Table 2) are available from an Institute of Terrestrial Ecology (ITE) collector situated about 3.5 km to the east of Lochnagar. Modelled sulphur deposition levels (Table 3) have also been calculated from the Harwell trajectory model (Derwent pers. comm.). These range from 0.84-1.01 g S m⁻² yr⁻¹ reflecting a moderate level of atmospheric contamination. The more highly polluted Galloway area has by contrast sulphur deposition levels of >1.2 g m⁻² yr⁻¹.

Black snowfalls, with a low pH (c. 3.0) and a large particulate component (fly ash from fossil fuel combustion), have also been reported from the area. For example in February 1984 an area of about 200 km² was affected. Episodes such as these could make an important contribution to the annual input of industrially derived atmospheric material (Davies *et al.* 1984). At high altitude sites such as these, hill cloud deposition is also likely to be important. This can increase deposition twofold by seeder-feeder scavenging (Fowler *et al.* 1988).

Table 1 Water quality Lochnagar and the Cairngorms (mean values 1986-1987)

	Lochnagar sites			Cairngorm sites	
	Lochnagar	Dubh Loch	L. nan Eun	Lochan Uaine	L. Coire an Lochan
mean pH	5.02	5.28	4.96	5.83	5.66
Conductivity μS cm ⁻¹	21.4	17.3	22.3	40.5	17.8
Ca ²⁺ μeq l ⁻¹	30.2	27.3	29.7	69.0	28.8
Mg ²⁺ μeq l ⁻¹	33.7	21.3	26.3	59.5	19.8
K ⁺ μeq l ⁻¹	7.5	5.3	5.7	10.0	7.8
Na ⁺ μeq l ⁻¹	89.0	72.3	82.3	191.0	67.0
Cl ⁻ μeq l ⁻¹	76.2	67.7	55.0	215.0	81.7
SO ₄ ²⁻ μeq l ⁻¹	66.0	54.0	62.3	82.0	47.3
Alkalinity (Alk _d) μeq l ⁻¹	0.00	1.37	0.00	14.79	13.73
TOC mg l ⁻¹	0.8	1.6	0.5	0.4	0.2
Labile Al μg l ⁻¹	57.0	55.0	128.0	56.0	75.0

Table 2 Annual average data from the ITE bulk monthly collector at Lochnagar ($\mu\text{eq l}^{-1}$).

	1978-1987 mean	1987
Acid H ⁺	37	25
Ammonium NH ₄ ⁺	18	15
Na ⁺	37	44
K ⁺	3	4
Ca ²⁺	17	10
Mg ²⁺	14	8
Cl ⁻	49	42
NO ₃ ⁻	19	19
SO ₄ ²⁻	48	34

Source; N. Cape (pers. comm.)

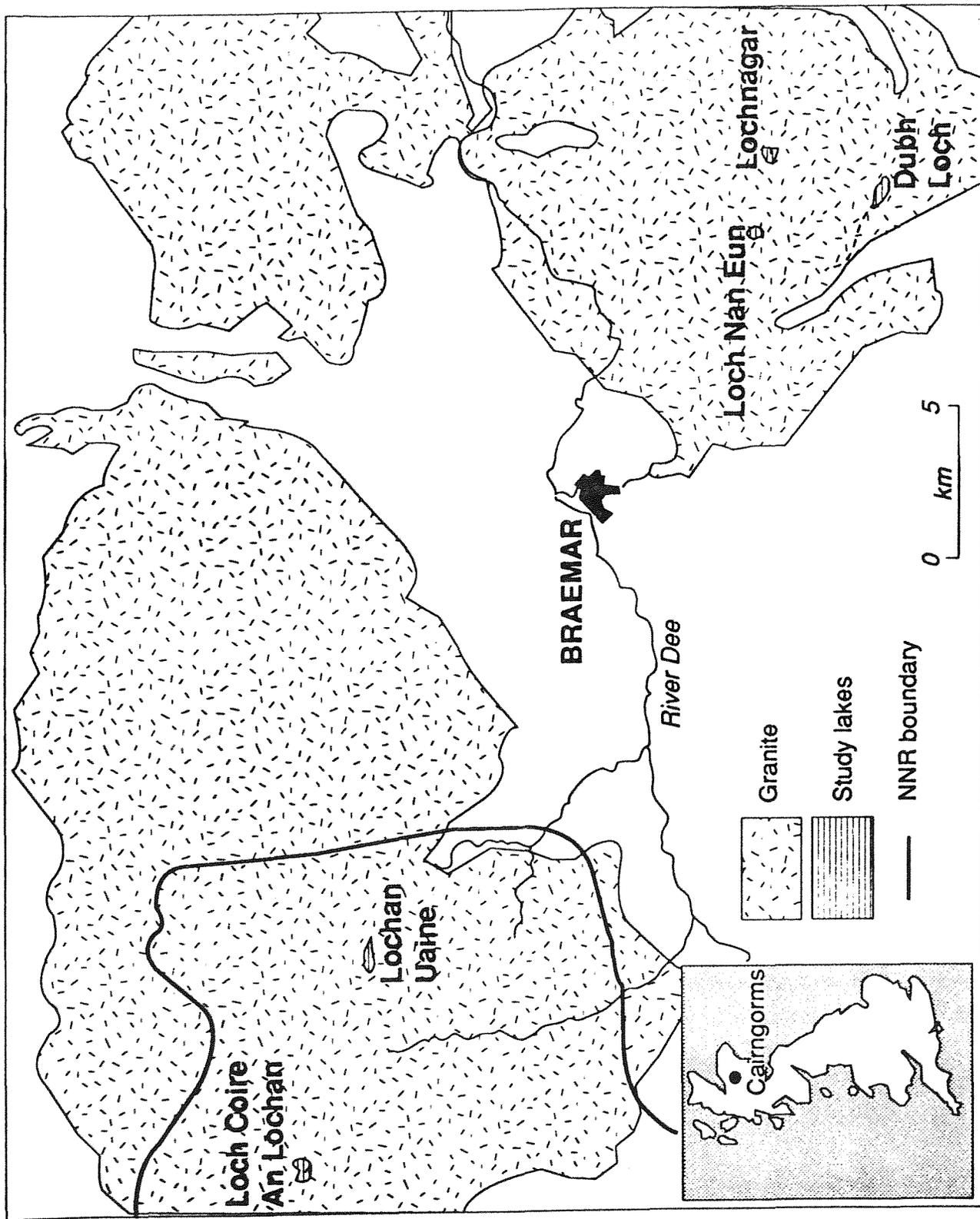
Table 3 Sulphur deposition Lochnagar and the Cairngorms

	Lochnagar	Dubh Loch	L. nan Eun	Lochan Uaine	L. Coire an Lochan
Dry deposited S (g m ⁻² yr ⁻¹)	0.589	0.599	0.585	0.490	*
Wet deposited S (g m ⁻² yr ⁻¹)	0.404	0.410	0.401	0.354	*
Total deposited S (g m ⁻² yr ⁻¹)	0.993	1.008	0.986	0.844	*

* no data available

Source; Harwell trajectory model (Derwent pers. comm.)

Figure 1 Cairngorms and Lochnagar study sites



3 METHODS

Sediment cores were taken from the sites in June 1986 using a mini-Mackereth corer (Mackereth 1969). Methods for lithostratigraphic, biostratigraphic, radiometric and carbonaceous particle analysis follow the Royal Society Surface Water Acidification Project (SWAP) protocol (Stevenson *et al.* 1987). Magnetic properties of the core were analysed according to Oldfield and Richardson (1990). Reconstruction of pH from the sediment diatom record utilises the statistical methods of multiple regression (Flower 1986) using the pH categories of Hustedt (1937-1939) and weighted averaging (ter Braak and van Dam 1989, Birks *et al.* 1990) using the SWAP calibration data set (Munro *et al.* 1990).

4 RESULTS

4.1 Loch Coire an Lochan

Bathymetry

A full bathymetric survey of this site was precluded by partial ice cover. It was apparent that the loch comprised a concentric basin with a deepest measured water depth of 20 m. Sediment cores were taken in 18 m water depth (Figure 2).

Lithostratigraphy

From 6-60 cm the sediment is a light grey diatomaceous mud with a low (c. 10% loss-on-ignition (LOI)) organic content (Figure 3). Above 5 cm the sediment is a greyish brown colour and LOI increases to c. 18% with concomitant declines in wet density and dry weight measurements.

Dating

^{210}Pb and ^{226}Ra results are given in Table 4. ^{137}Cs results are given in Table 5. The short-lived Chernobyl fallout isotope ^{134}Cs was not detected. ^{226}Ra activities in this core were unusually high, with a mean value of 14.4 pCi g^{-1} . Unsupported ^{210}Pb activities were also very high, suggesting low accumulation rates, and ^{210}Pb equilibrium was reached at a depth of less than 5 cm. The unsupported ^{210}Pb activity declines more or less exponentially with depth, indicating a reasonably constant sediment accumulation rate. Mean accumulation rates have been calculated using both the CRS and CIC ^{210}Pb dating models (Figure 4) (Appleby and Oldfield 1978). The dates given in Table 6 have been calculated using the average value of $0.0093 \text{ g cm}^{-2} \text{ y}^{-1}$ given by these two methods and extrapolated down to a depth of 5 cm.

The ^{137}Cs data from this core (Table 5) are of little chronological value. The maximum ^{137}Cs activity occurred in the topmost sediments, although it should be noted that the ^{210}Pb dates put the 1963 level at a depth of only 0.75 cm. The presence of significant activities at depths which clearly predate the onset of ^{137}Cs fallout in 1954 indicates the extent of ^{137}Cs mobility within the core.

Table 4 *Loch Coire an Lochan : ^{210}Pb and ^{226}Ra data*

Depth cm	Dry mass g cm ⁻²	^{210}Pb Conc.				^{226}Ra Conc.	
		Total		Unsupp.		pCi g ⁻¹	±
		pCi g ⁻¹	±	pCi g ⁻¹	±		
0.25	0.0521	60.27	1.35	45.74	1.39	14.53	0.33
0.75	0.2070	40.75	1.01	28.81	1.05	11.94	0.30
1.75	0.5671	27.08	0.73	12.72	0.77	14.36	0.26
2.25	0.7722	14.33	1.16	1.83	1.21	12.50	0.33
2.75	0.9715	16.85	0.81	1.35	0.85	15.50	0.27
5.50	2.0089	12.42	0.71	0.33	0.75	12.09	0.24
9.50	3.4865	15.44	0.52	0.59	0.56	14.85	0.21
12.50	4.3692	14.63	0.60	-1.66	0.65	16.29	0.24
16.00	5.3807	10.34	0.47	0.65	0.50	9.69	0.18

Table 5 *Loch Coire an Lochan : ^{137}Cs data*

Depth cm	^{137}Cs Conc.	
	pCi g ⁻¹	±
0.25	6.36	0.22
0.75	3.31	0.18
1.75	1.57	0.13
2.25	0.77	0.17
2.75	0.94	0.11
5.50	0.03	0.09
9.50	0.00	0.00
12.50	0.00	0.00
16.00	0.00	0.00

Table 6 *Loch Coire an Lochan : ²¹⁰Pb chronology*

Depth cm	Cum. dry mass g cm ⁻²	Chronology			Sedimentation rate		
		Date AD	Age yr	±	g cm ⁻² yr ⁻¹	cm yr ⁻¹	± (%)
0.00	0.0000	1986	0				
0.25	0.0521	1980	6	1			
0.50	0.1295	1972	14	2			
0.75	0.2070	1964	22	4			
1.00	0.2970	1954	32	5			
1.25	0.3871	1944	42	7			
1.50	0.4771	1935	51	9			
1.75	0.5671	1925	61	10			
2.00	0.6696	1914	72	12			
2.25	0.7722	1903	83	14			
2.50	0.8718	1892	94	16	0.0094	0.026	16.9%
2.75	0.9715	1881	105	18			
3.00	1.0658	1871	115	19			
3.25	1.1601	1861	125	21			
3.50	1.2544	1851	135	23			
3.75	1.3487	1841	145	24			
4.00	1.4430	1831	155	26			
4.25	1.5374	1820	166	28			
4.50	1.6317	1810	176	30			
4.75	1.7260	1800	186	31			
5.00	1.8203	1790	196	33			

Diatom analysis and pH reconstruction

A summary diatom diagram is presented in Figure 5. Floristic changes occur throughout the core. In the top 3 cm the values of *Fragilaria virescens*, *F. lata*, *F. cf. oldenburgiana* and *Brachysira vitrea* fall and there are increased abundances of *Achnanthes austriaca* f. *minor* and *A. marginulata*. Below 3 cm the composition of the flora is not stable; *F. lata* is only found in levels above 18 cm and *B. vitrea* starts to decline above 20 cm. Acidobiontic forms (eg. *Aulacoseira distans* v. *nivalis*) account for >10% of species at the top and bottom of the profile.

Weighted averaging and multiple regression methods were used to reconstruct pH at this site, and results are shown in Figure 5. The multiple regression method gives consistently higher pH values, and a closer correspondence with the modern measured pH. Weighted averaging suggests a slight trend to a lower pH above 24 cm from values around pH 5.3 to a value of 5.0 at the top of the core. The multiple regression method also suggests a lower pH above about 24 cm, although values fluctuate. Neither of the methods show a decline in pH from the mid-nineteenth century (3.5 cm). The weighted averaging method gives a pH of 4.9-5.0 throughout the period from the mid-nineteenth century to the present day (0-3.5 cm). There is therefore some evidence of a very slight drop in pH at an early date, but there is no evidence of any additional acidification in the recent past (nineteenth or twentieth century).

Diatom concentrations are shown in Figure 6. Concentrations are high throughout, but fall towards the top of the core.

Pollen analysis

There is little vegetation in the catchment at present and it is unlikely that a dense vegetation cover has ever existed at such a high altitude site. As a result much of the pollen shown in Figure 7 has been blown into the lake from lower altitudes. For example, it is extremely unlikely that pine ever grew in the catchment (the mid-post-glacial upper limit of pine in the Cairngorms is estimated at about 800 m (Pears 1970)), and it is certainly not present today, yet it contributes about 40% of the total pollen record. Similarly *Calluna* is not present in the catchment today, but makes up about 10% of the total pollen in the surface sediments.

Because of the influence of long-distance pollen transport at this site the pollen diagram is difficult to interpret and conclusions about changes in catchment vegetation cannot be made.

Carbonaceous particle analysis

The results of carbonaceous particle analysis are shown in Figure 8. Carbonaceous particles are only found in high concentrations in the surface sediment sample (0-0.5 cm). Because of the low sediment accumulation rate this sediment slice represents a period from about 1972 to 1986. Concentrations from above 3 cm (extrapolated date 1871) are much

lower but still probably reflect a significant amount of atmospheric contamination. Below this level counts are low and are also rather unreliable since they approach the limit of detection (c. 100 particles gDM^{-1}).

Sediment chemistry

The high dry weight and low LOI content of the sediment and the high concentrations of the major cations sodium and potassium (Figure 9), indicate that the sediments of Loch Coire an Lochan are coarse and minerogenic. These features are a result of the semi-arctic conditions in the catchment of the lake with harsh meteorological conditions and the absence of vegetation. The abundance of sodium and potassium feldspars in the granite of the area results in the high concentrations in the lake sediment.

All four cations and the four trace metal concentrations (Figure 10) generally increase up the core. As the increase in trace metal concentration is not large compared to that of the major cations, it is not possible to establish if there is any trace metal contamination of the Loch Coire an Lochan sediments. When the major cation concentrations vary in a core as a result of changes in the sedimentary regime, the variation of trace metal concentration with a major cation in the lower part of the core, before contamination starts, can usually be used to estimate the trace metal concentrations in the absence of contamination in the upper part of the core. The point where the measured trace metal concentration rises above the estimated values calculated to be appropriate for conditions in the absence of contamination can then be estimated. However, in this case it is impossible to detect any departures from the relationship between zinc or lead and sodium in the upper 5 cm of the core (Figure 11). If there is any contamination of the sediments of Loch Coire an Lochan by material deposited from the atmosphere, it is small.

Figure 2 L. Coire an Lochan: coring location

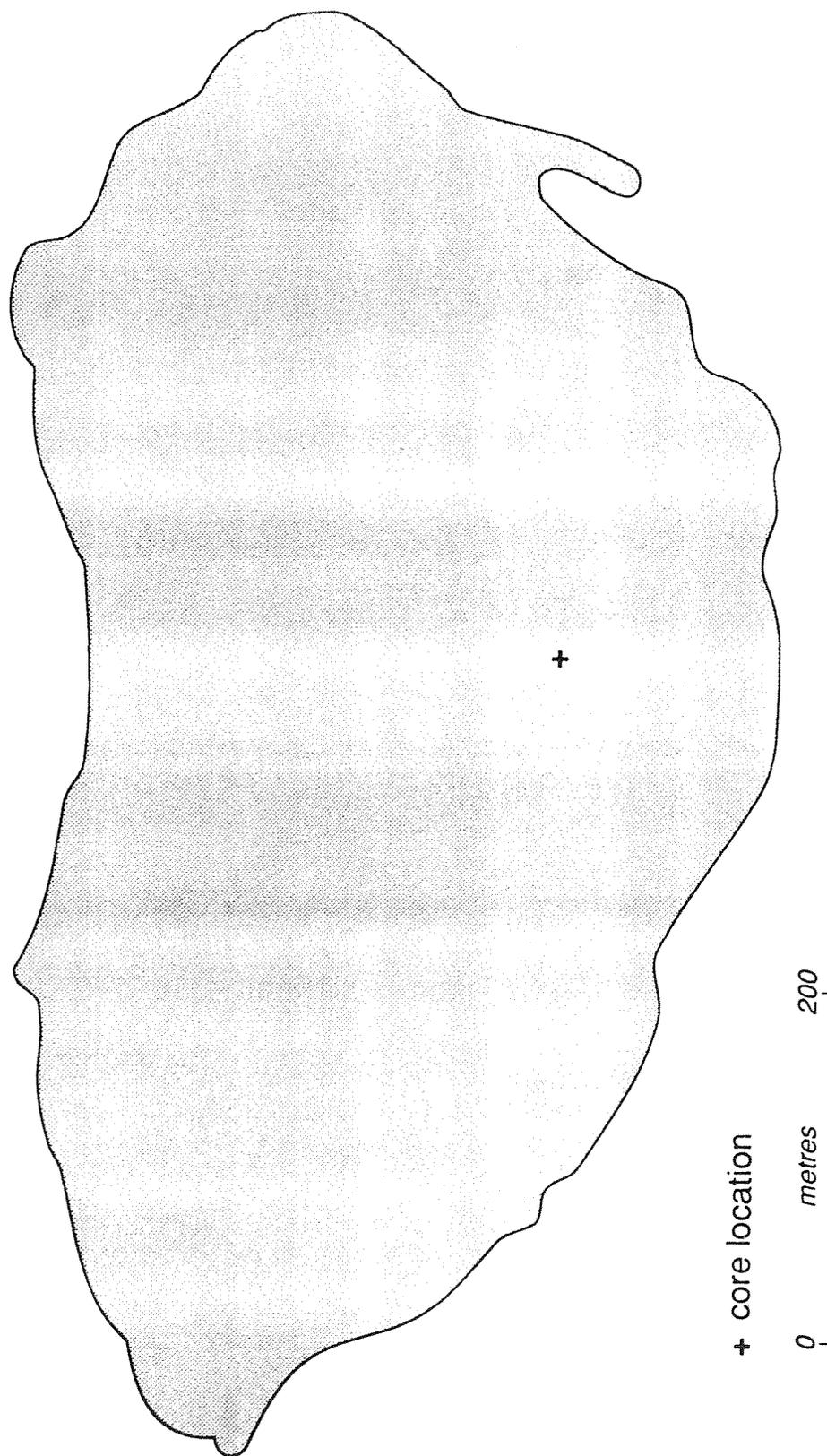


Figure 3 L. Coire an Lochan: lithostratigraphy

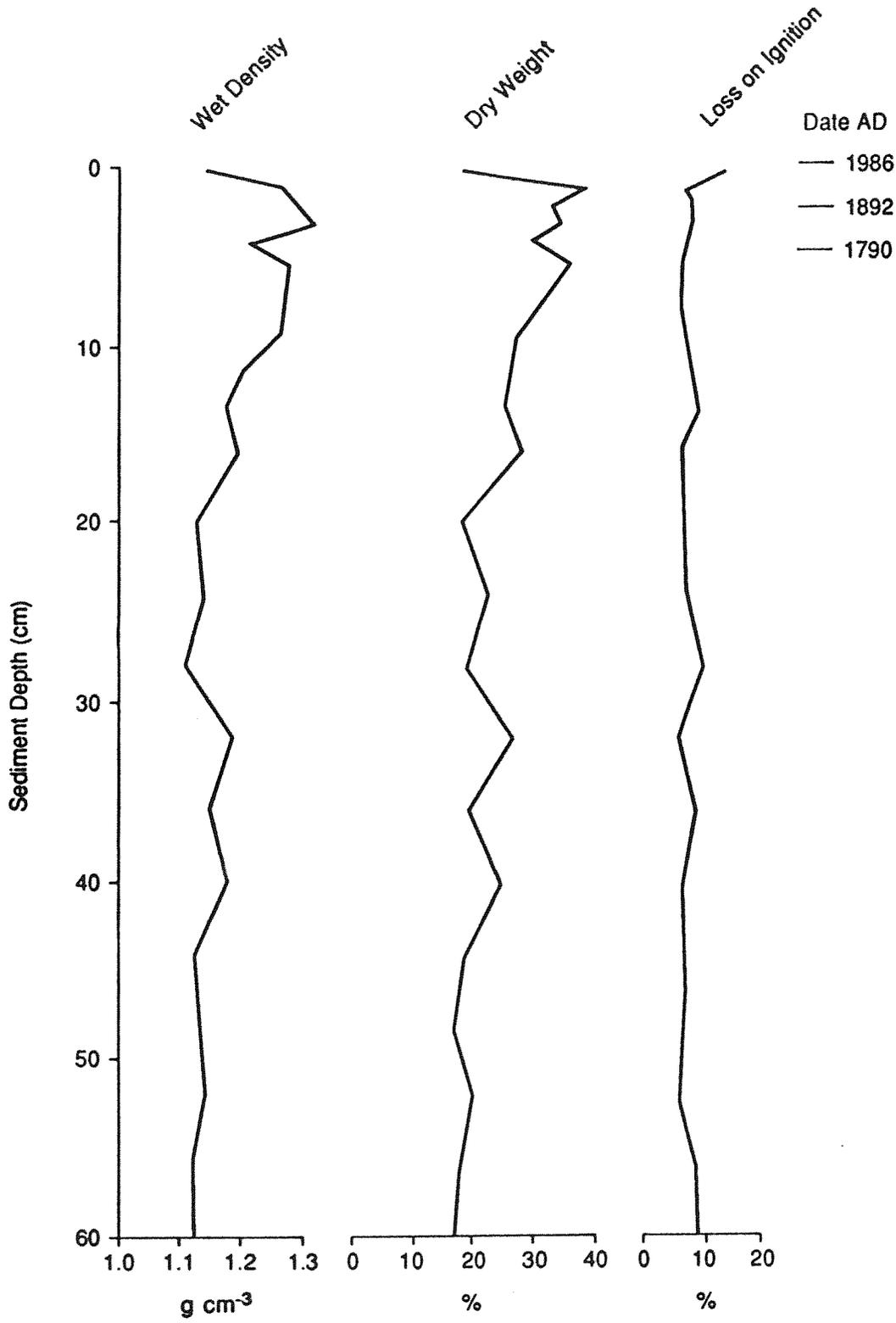


Figure 4 L. Coire an Lochan: ^{210}Pb Chronology

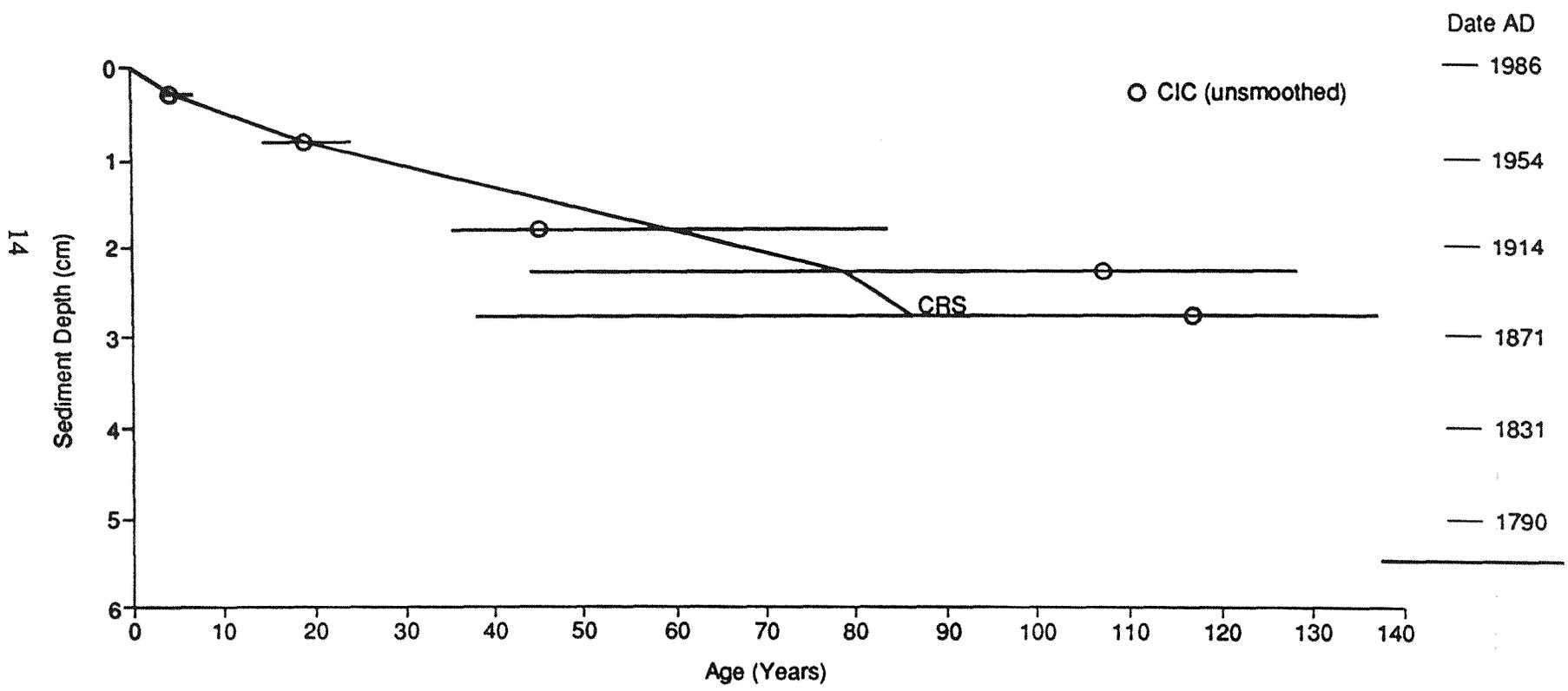


Figure 6 L. Coire an Lochan: diatom concentration ($\times 10^8 \text{ g}^{-1}$), showing 95% confidence limits

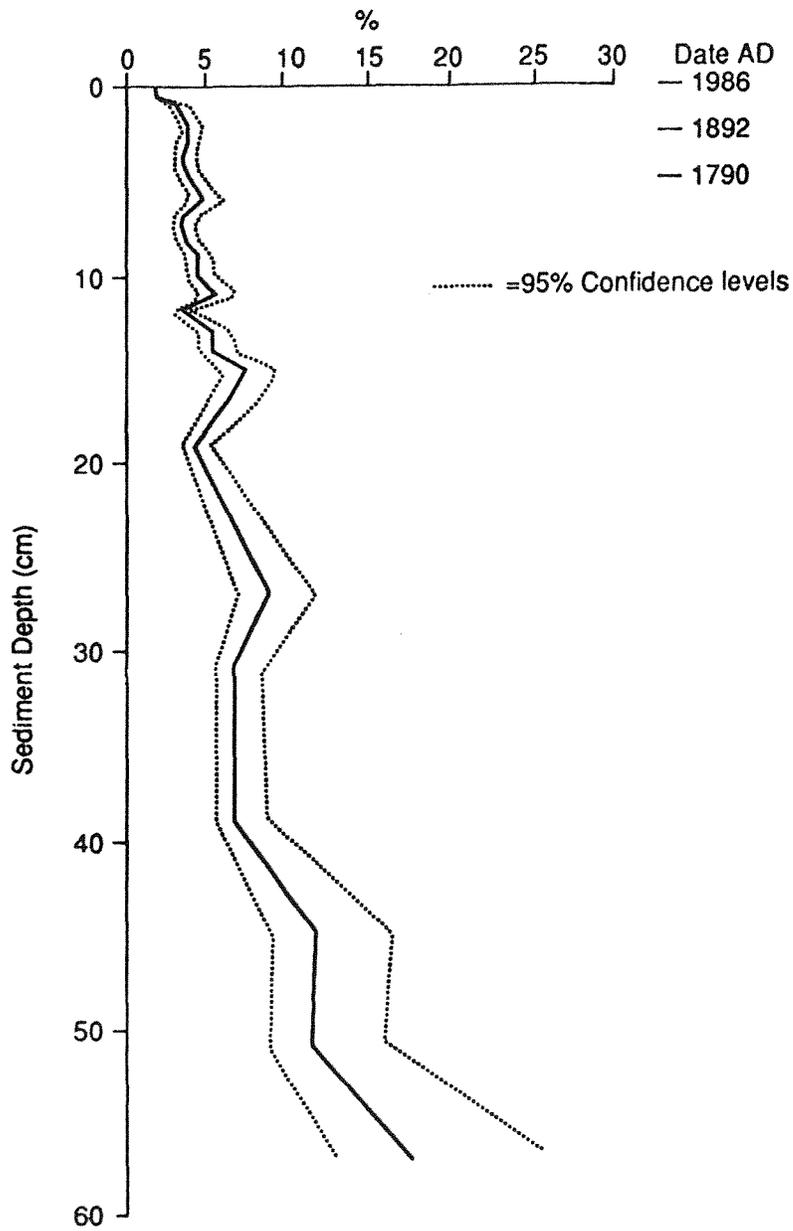


Figure 7 L. Coire an Lochan: summary pollen profile

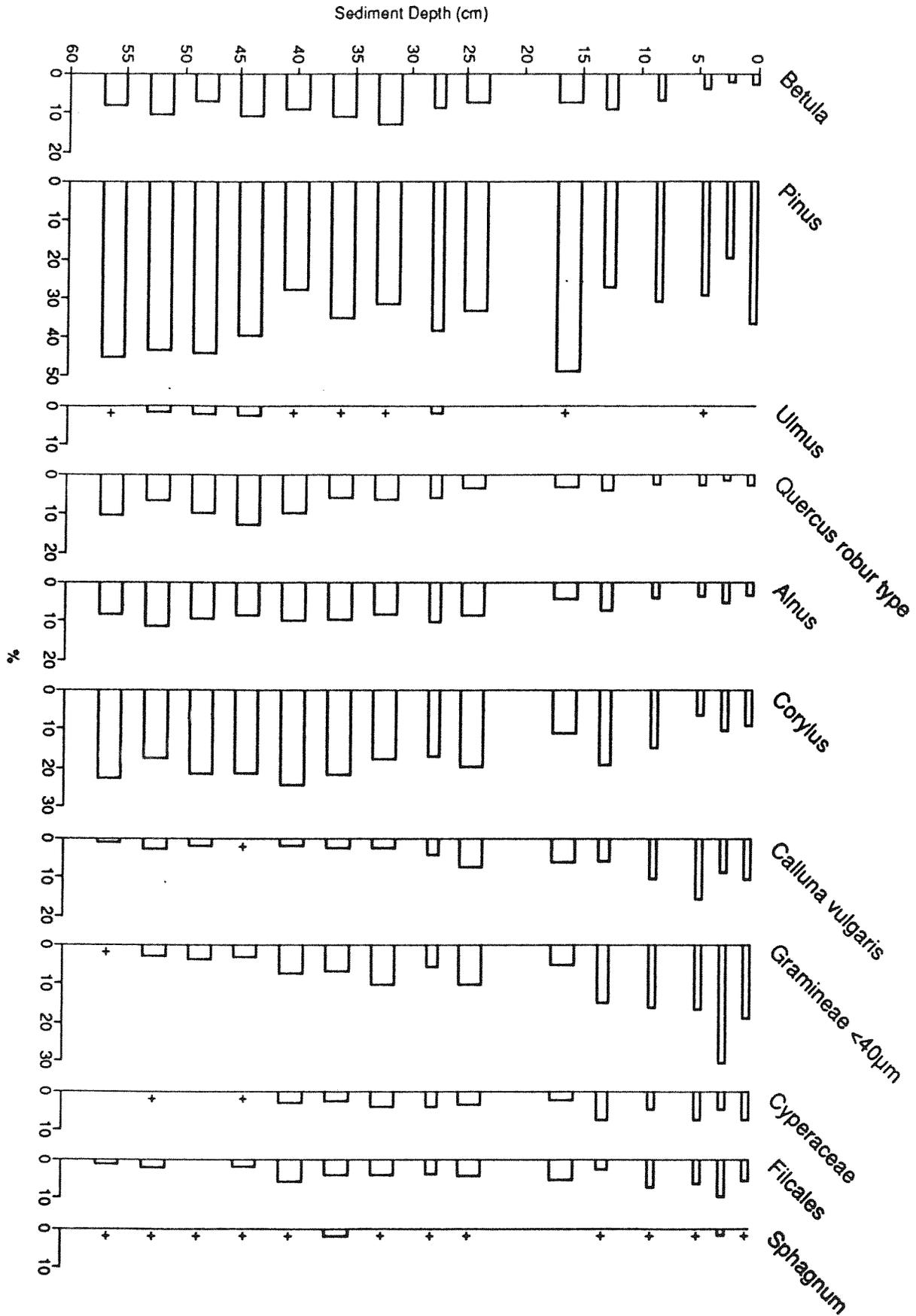


Figure 8 L. Coire an Lochan: carbonaceous particle profile ($\times 10^3 \text{ gDM}^{-1}$)

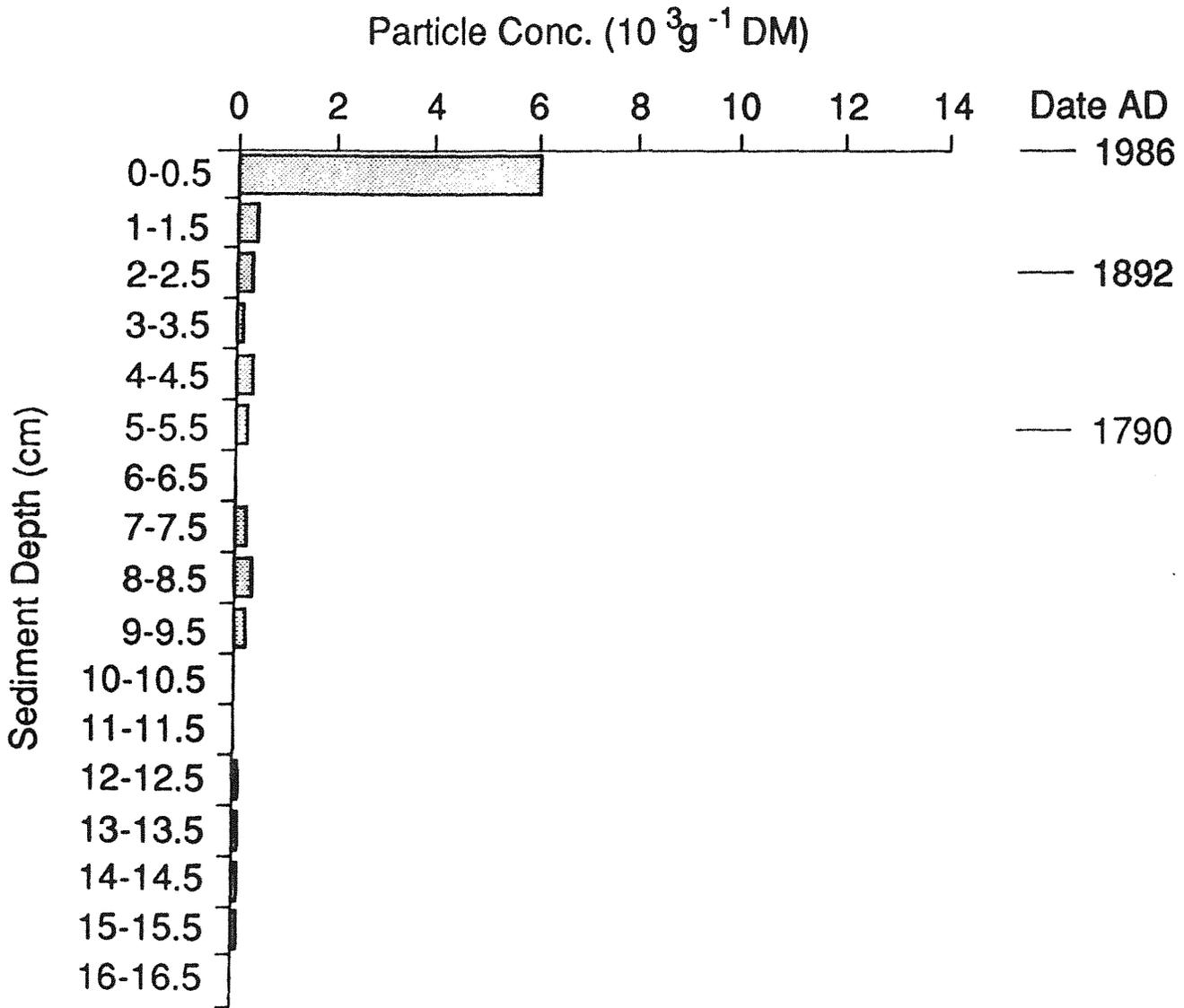


Figure 9

L. Coire an Lochan: the variation of major ion concentrations with depth

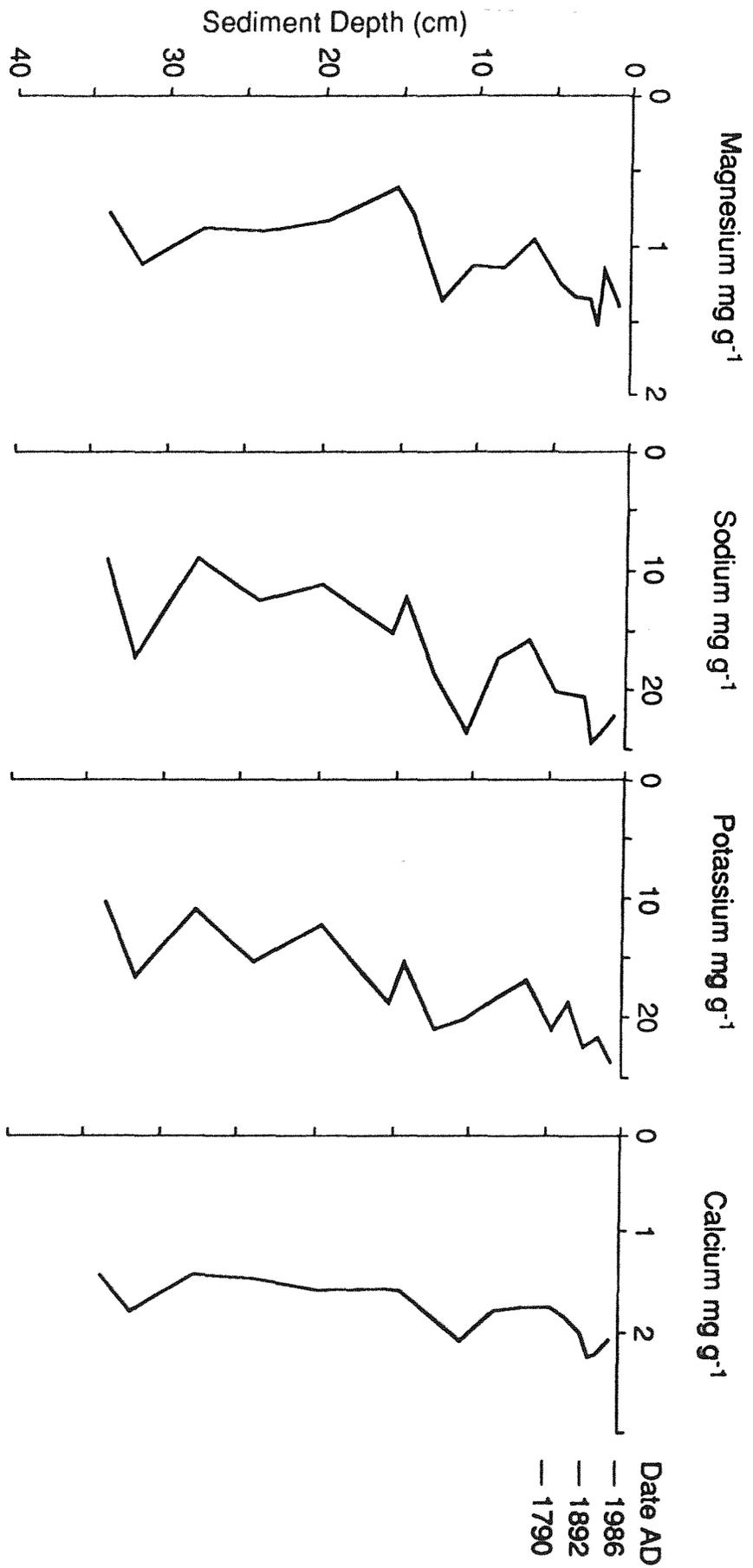


Figure 10

L. Coire an Lochan: the variation of trace metal concentrations with depth

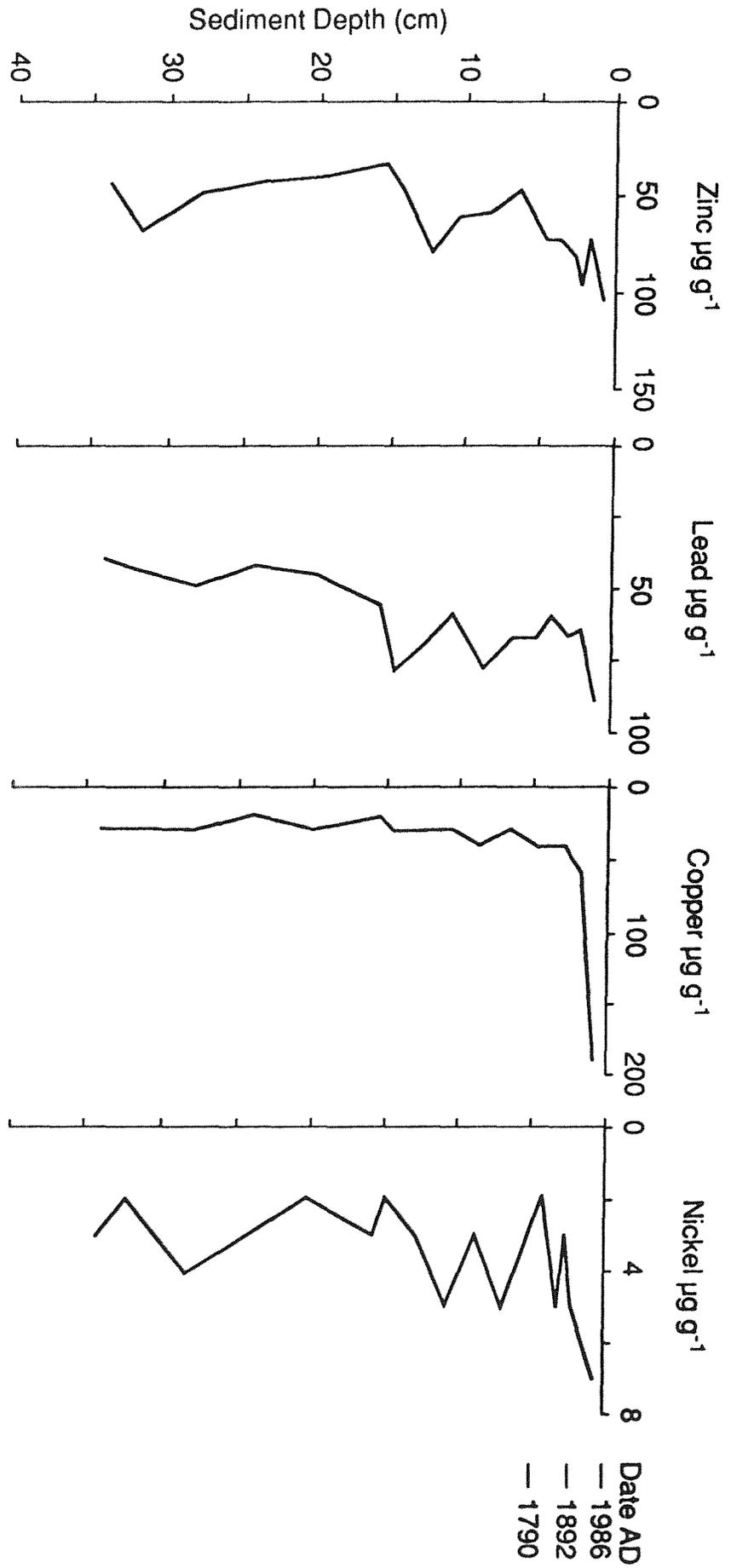
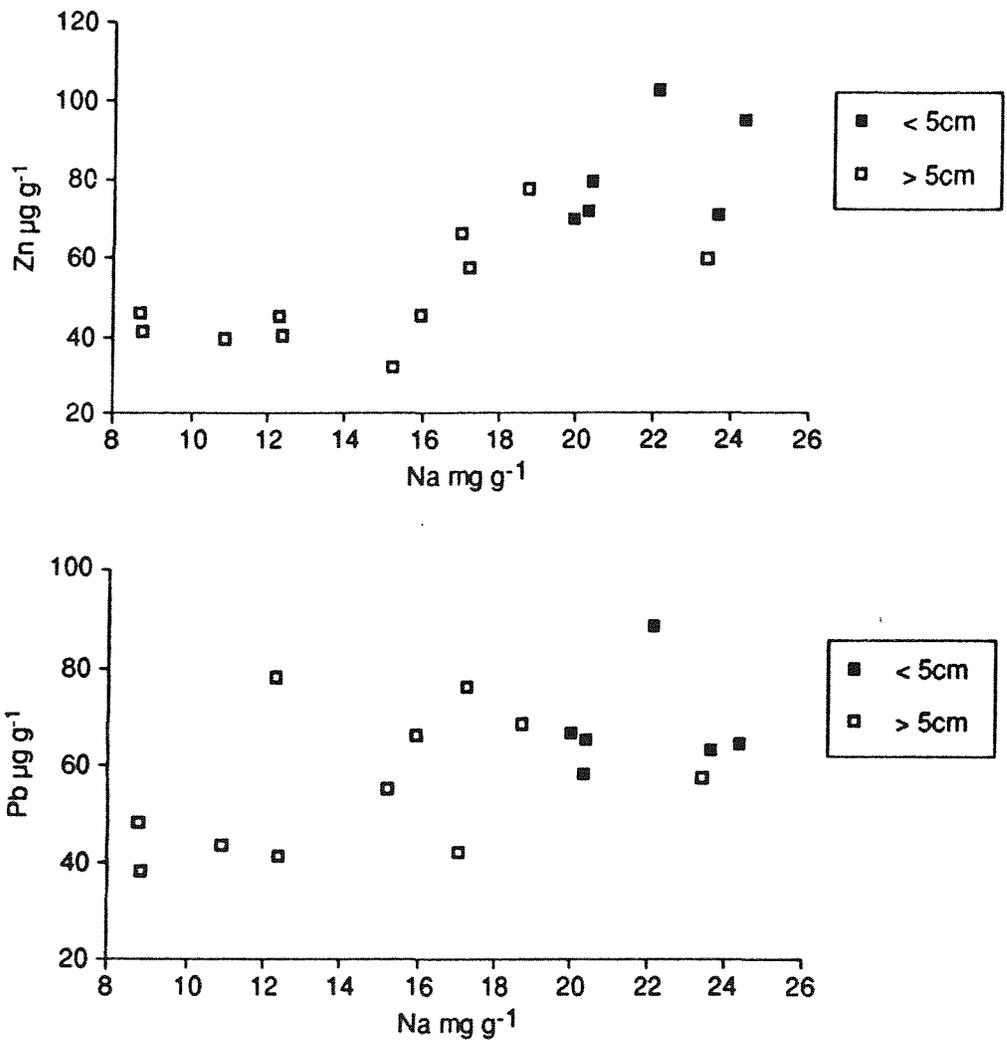


Figure 11 L. Coire an Lochan: the variation of zinc and sodium and lead and sodium concentrations in the sediments



4.2 Lochan Uaine

Lochan Uaine was included jointly in the SWAP programme (Battarbee and Renberg 1990) and DoE projects. Within SWAP it was selected as a lake site to complement the Allt a'Mharcaidh experimental stream catchment. Here it is used to extend the regional coverage of sites in this area.

Bathymetry

A bathymetric survey revealed a single deep basin with a maximum water depth of 20 m (Figure 12). Sediment cores were taken from 19 m.

Lithostratigraphy

The sediment is an inorganic diatomaceous mud with percentage LOI values of around 10% throughout the core (Figure 13). The inorganic nature of the sediment is also reflected in the high dry weight and wet density values. The peaks in dry weight and wet density tend to coincide, for example at 12-13 cm and 28-29 cm. There is evidence of an inwash of catchment material at 28 cm, since coarse sand and unhumified roots were also found at this level. A possible mechanism for the incorporation of coarse material into the sediment at the deepest part of the lake is ice-rafting. Material from the steep back wall of the corrie falling on to the frozen lake in winter could subsequently be transported by ice-rafts and deposited in the centre of the lake when ice melt occurs.

Dating

The ^{210}Pb and ^{226}Ra results are given in Table 7. The ^{137}Cs and ^{134}Cs results (corrected for decay) are given in Table 8. The ^{134}Cs data can be used to partition the ^{137}Cs activity into its two components, deriving respectively from weapons testing and Chernobyl fallout. Concentrations of the other weapons test fallout radionuclide, ^{241}Am , were below the limits of detection.

The site is unusual since it has very high ^{226}Ra activity in the sediments, the mean concentration of 18.9 pCi g^{-1} is about 10 times the value typical of other UK sites. Activities were also quite varied, with values significantly above the average between 14 cm and 19 cm. The high unsupported ^{210}Pb activity of the near-surface sediments and high ^{210}Pb inventory of the core (137 pCi cm^{-2} representing a constant ^{210}Pb supply of $4.27 \text{ pCi cm}^{-2} \text{ yr}^{-1}$) may reflect an enhanced local ^{210}Pb flux due to higher ^{222}Rn diffusion rates from the soils of the region.

^{210}Pb chronologies have been calculated using both the CRS and CIC ^{210}Pb dating models (Appleby and Oldfield 1978) and the results are given in Figure 14. Below 1.75 cm, dated 1973, the two models are in good agreement. The unsupported ^{210}Pb activity (plotted on a logarithmic scale) declines more or less linearly with depth and both models indicate a reasonably constant sedimentation of $c. 0.028 \pm 0.004 \text{ g cm}^{-2} \text{ yr}^{-1}$. The decline in ^{210}Pb activity above 1.75 cm appears to indicate a recent acceleration in sediment accumulation rates. Since the CIC model can not date non-monotonic features the chronology has made

use of the CRS dates for this part of the core. The divergence at lower levels is probably due to errors in the CRS dates arising from the very high ^{226}Ra activity. Dates given by the CRS model are shown in Table 9.

Although the presence of Chernobyl ^{137}Cs down to 1.75 cm and weapons test ^{137}Cs down to the ^{210}Pb equilibrium depth indicates significant downward diffusion of this isotope (cf. Davis *et al.* 1984), the weapons fallout ^{137}Cs still has a well defined peak at 1.75-2.75 cm which would appear to record the 1963 fallout maximum. The definition of this peak would appear to exclude the possibility of mixing as an explanation for the reduced ^{210}Pb activities in the near-surface sediments. Figure 14 shows that the ^{137}Cs date is in good agreement with the ^{210}Pb chronology, which dates the ^{137}Cs weapons fallout peak to the period 1963-1973.

Table 7 *Lochan Uaine: ²¹⁰Pb and ²²⁶Ra data*

Depth cm	Dry mass g cm ⁻²	²¹⁰ Pb Conc.				²²⁶ Ra Conc.	
		Total		Unsupp.		pCi g ⁻¹	±
		pCi g ⁻¹	±	pCi g ⁻¹	±		
0.25	0.0742	88.97	2.01	78.37	2.05	10.60	0.39
0.75	0.2233	107.85	1.54	97.44	1.56	10.41	0.26
1.75	0.4744	127.16	1.85	113.04	1.88	14.12	0.34
2.75	0.7243	106.66	1.40	92.35	1.42	14.31	0.26
3.75	1.0449	65.14	1.16	47.68	1.19	17.46	0.26
4.75	1.3715	50.13	1.04	30.82	1.07	19.31	0.26
5.75	1.6881	42.39	1.01	22.49	1.05	19.90	0.28
6.75	2.0543	34.77	0.80	20.86	0.83	13.91	0.22
7.25	2.2600	37.13	0.96	14.79	1.00	22.34	0.29
8.25	2.6034	27.49	0.81	8.16	0.85	19.33	0.27
9.75	3.2303	28.99	0.87	8.14	0.91	20.85	0.27
12.75	4.6380	15.49	0.44	-2.57	0.47	18.06	0.16
15.75	5.7243	33.07	0.71	0.61	0.76	32.46	0.26
18.25	6.5124	28.71	0.78	0.38	0.83	28.33	0.29

Table 8 *Lochan Uaine: ¹³⁷Cs and ¹³⁴Cs data*

Depth cm	¹³⁷ Cs Conc.		¹³⁴ Cs Conc.	
	pCi g ⁻¹	±	pCi g ⁻¹	±
0.25	6.45	0.29	1.89	0.30
0.75	8.31	0.20	1.19	0.18
1.75	9.91	0.25	1.04	0.24
2.75	7.88	0.19	0.00	0.00
3.75	3.25	0.14	0.00	0.00
4.75	1.41	0.10	0.00	0.00
5.75	0.81	0.11	0.00	0.00
6.75	0.35	0.08	0.00	0.00
7.25	0.07	0.09	0.00	0.00
8.25	0.16	0.08	0.00	0.00
9.75	0.11	0.08	0.00	0.00
12.75	0.06	0.03	0.00	0.00
15.75	0.00	0.00	0.00	0.00
18.25	0.00	0.00	0.00	0.00

Table 9 *Lochan Uaine: ^{210}Pb chronology*

Depth cm	Cum. dry mass g cm ⁻²	Chronology			Sedimentation rate		
		Date AD	Age yr	±	g cm ⁻² yr ⁻¹	cm yr ⁻¹	± (%)
0.00	0.0000	1986	0				
0.25	0.0742	1985	1	2	0.0520	0.175	3.5
0.50	0.1487	1983	3	2	0.0449	0.158	3.2
0.75	0.2233	1981	5	2	0.0377	0.141	3.0
1.00	0.2861	1979	7	2	0.0346	0.131	3.0
1.25	0.3488	1977	9	2	0.0314	0.121	3.1
1.50	0.4116	1975	11	2	0.0283	0.111	3.2
1.75	0.4744	1973	13	2	0.0252	0.101	3.3
2.00	0.5369	1970	16	2	0.0245	0.095	3.4
2.25	0.5993	1968	18	2	0.0237	0.089	3.5
2.50	0.6618	1965	21	2	0.0230	0.084	3.6
2.75	0.7243	1963	23	2	0.0222	0.078	3.7
3.00	0.8044	1959	27	2	0.0239	0.081	4.1
3.25	0.8846	1956	30	2	0.0255	0.083	4.4
3.50	0.9647	1953	33	2	0.0271	0.086	4.7
3.75	1.0449	1950	36	2	0.0288	0.089	5.0
4.00	1.1265	1947	39	2	0.0295	0.091	5.4
4.25	1.2082	1944	42	2	0.0302	0.094	5.9
4.50	1.2898	1941	45	2	0.0309	0.096	6.3
4.75	1.3715	1939	47	2	0.0317	0.098	6.7
5.00	1.4506	1936	50	2	0.0317	0.097	7.2
5.25	1.5298	1934	52	2	0.0317	0.096	7.7
5.50	1.6089	1931	55	3	0.0317	0.094	8.2
5.75	1.6881	1929	57	3	0.0318	0.093	8.7
6.00	1.7796	1925	61	3	0.0294	0.084	9.6
6.50	1.9627	1919	67	4			
7.00	2.1571	1912	74	5			
7.50	2.3458	1905	81	6			
8.00	2.5175	1899	87	7	0.0283	0.074	14.5%
8.50	2.7079	1892	94	9			

Diatom analysis and pH reconstruction

A summary diatom diagram is shown in Figure 15. Below about 12 cm the composition of the diatom flora is fairly stable, dominated by circumneutral species such as *Fragilaria virescens*, *Brachysira vitrea*, *Achnanthes minutissima*, together with acidophilous *Achnanthes* species, which contribute about 40% of the assemblage. Above about 10 cm there are a number of floristic changes the most dramatic of which is the sudden increase of *Achnanthes austriaca* v. *alpina*. The acidophilous species *Achnanthes scotica* declines above 10 cm and small increases in species such as *Eunotia exigua*, *Cymbella aequalis*, and *Achnanthes marginulata* f. *major* also occur. Lochan Uaine exhibits many of the taxonomic problems associated with small *Achnanthes* taxa found at upland sites in the Cairngorms and Wales (Flower and Jones 1989).

Weighted averaging and multiple regression of pH preference groups were used to reconstruct pH (Figure 15). Both models show a slight acidification above 9 cm (c. 1885). The weighted average method shows a slight drop in pH from pH 5.4-5.6 to pH 5.1-5.4, with a modern reconstructed pH of 5.1. The present pH of the loch is 5.8. The poor correlation between reconstructed and measured pH at this site can be partly explained by the poor fit above 24 cm between core samples and samples in the SWAP surface sediment data set (Stevenson *et al.* 1991). For example, many of the *Achnanthes* species found at this site are uncommon in the SWAP diatom data base. The results of multiple regression give a better fit with modern measured pH and show evidence for a slight decrease in pH in the top 10 cm.

The high concentration ($>10^7$ valves g^{-1}) of diatoms throughout the core (Figure 16) reflects the high diatomaceous component in the sediment. Concentrations are fairly uniform although there is a drop at 12 cm coincident with lithostratigraphic evidence for an inwash event. Diatom concentrations subsequently recover towards the top of the core.

Pollen analysis

The pollen profile (Figure 17) is dominated by open habitat species; *Calluna*, Gramineae and Cyperaceae. Tree pollen is dominated by *Pinus* and the values of *Picea* in the top 2 cm probably reflect recent coniferous planting at lower elevations in the region. The unusually high values of *Lycopodium annotinum* and *Huperzia selago* spores in the profile reflect the alpine heath vegetation of the catchment. There is no evidence of an increase in *Calluna* at the expense of Gramineae in the recent past, confirming the improbability of major land-use change in the area.

Carbonaceous particle analysis

20 sediment samples from 0-20 cm were analysed; the results are presented in Figure 18. Few carbonaceous particles ($<0.3 \times 10^3$ gDM^{-1}) are present below 3.5 cm; above 3.5 cm (c. 1953) the concentration of carbonaceous particles rises to the surface.

Magnetics analysis

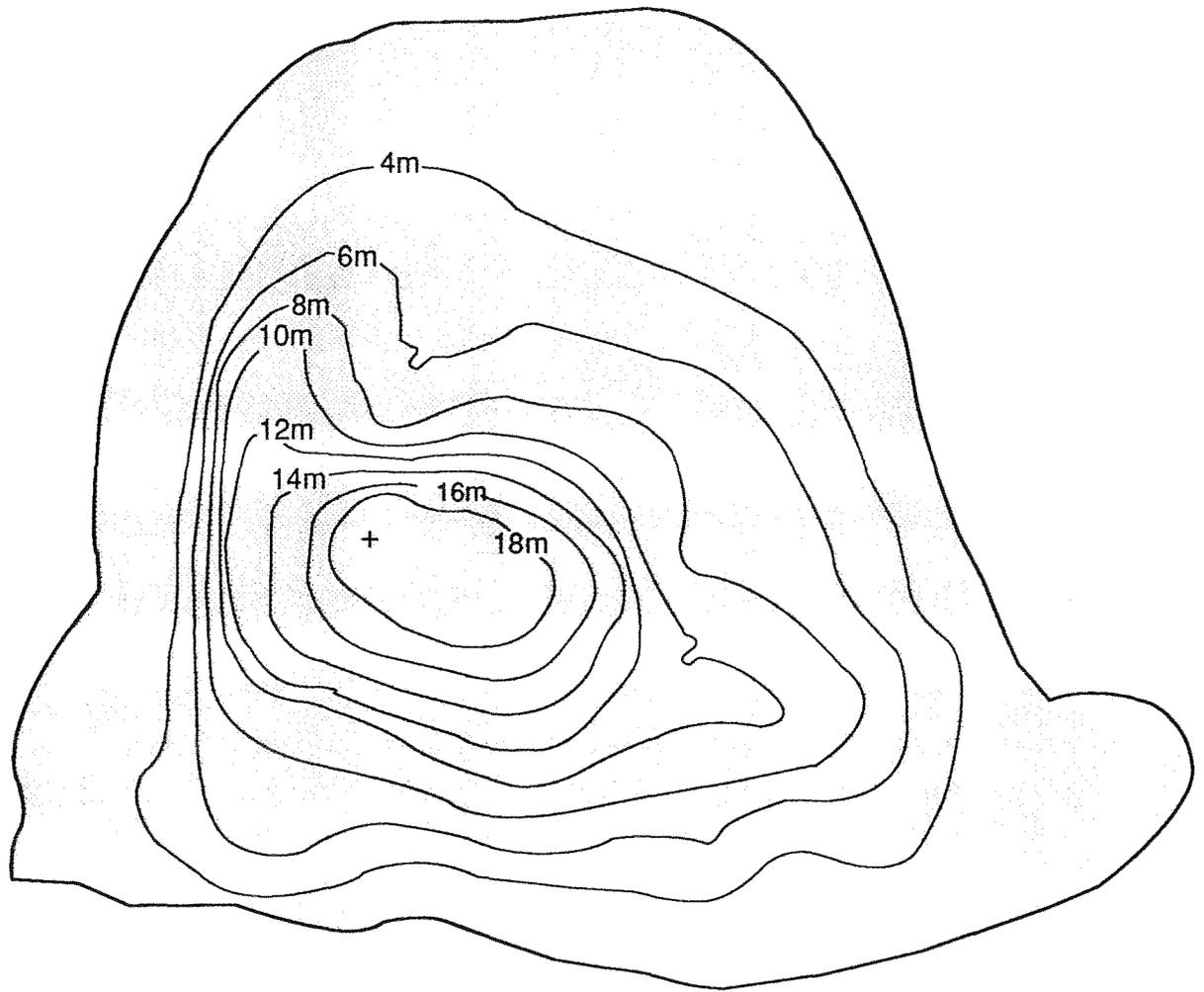
Although there is a significant catchment input of magnetic minerals, the magnetite component of the atmospheric deposition record can be distinguished, and there is an increase in the input of magnetite after about 1950 (Figure 19).

Sediment chemistry

The minerogenic nature of the Lochan Uaine sediments, already commented on, is also reflected in the high sodium and potassium concentrations (Figure 20). The major cation concentrations vary little up the core, only potassium shows a small drop in concentration.

The trace metal concentrations, except nickel, increase in the upper part of the core (Figure 21). Visual inspection suggests that zinc starts to increase above 10 cm and lead above 20 cm. However, Figure 22 shows that if the variation in major cation concentration is taken into account, then the start of lead contamination is also around 10 cm. This depth is just before the dated part of the core, but by extrapolation corresponds to c. 1870. As there is no possibility of trace metal contamination from catchment sources in this remote, upland lake, the origin of the lead and zinc contamination is by material deposited from the atmosphere.

Figure 12 L. Uaine: bathymetry (contour intervals in metres)



+ Core location
Contours in metres

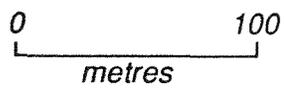


Figure 13 L. Uaine: lithostratigraphy

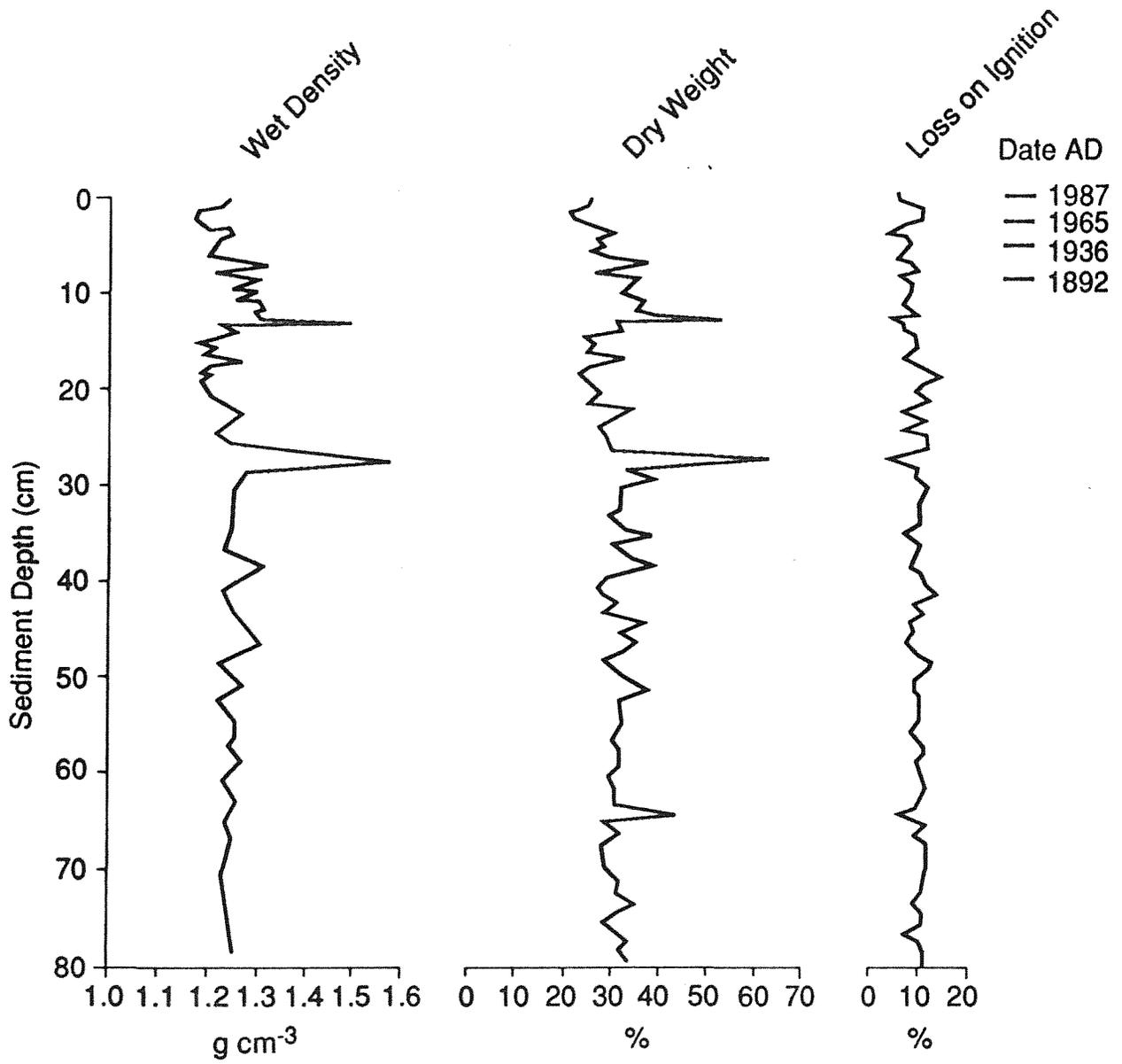


Figure 14 L. Uaine: ^{210}Pb chronology

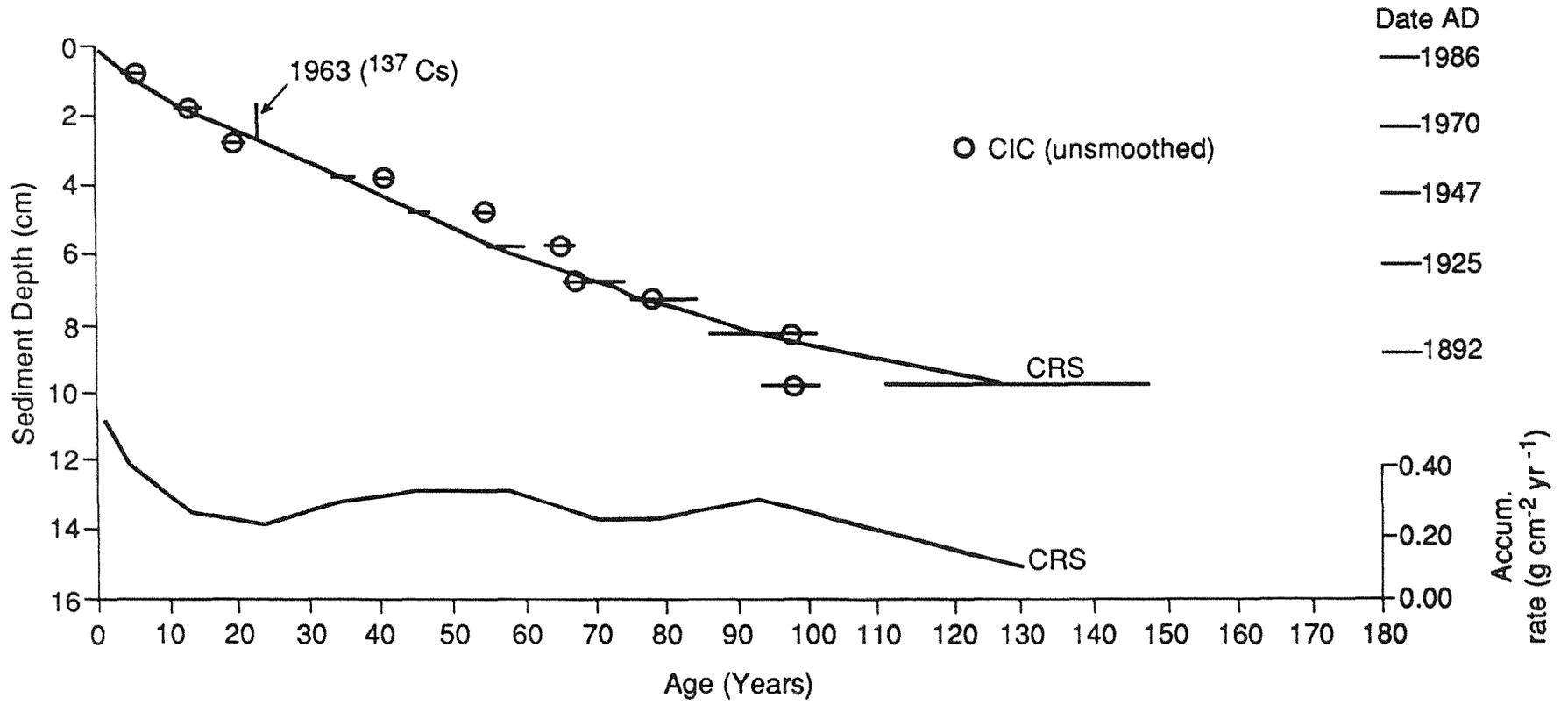


Figure 15 L. Uaine: diatom summary diagram and pH reconstructions

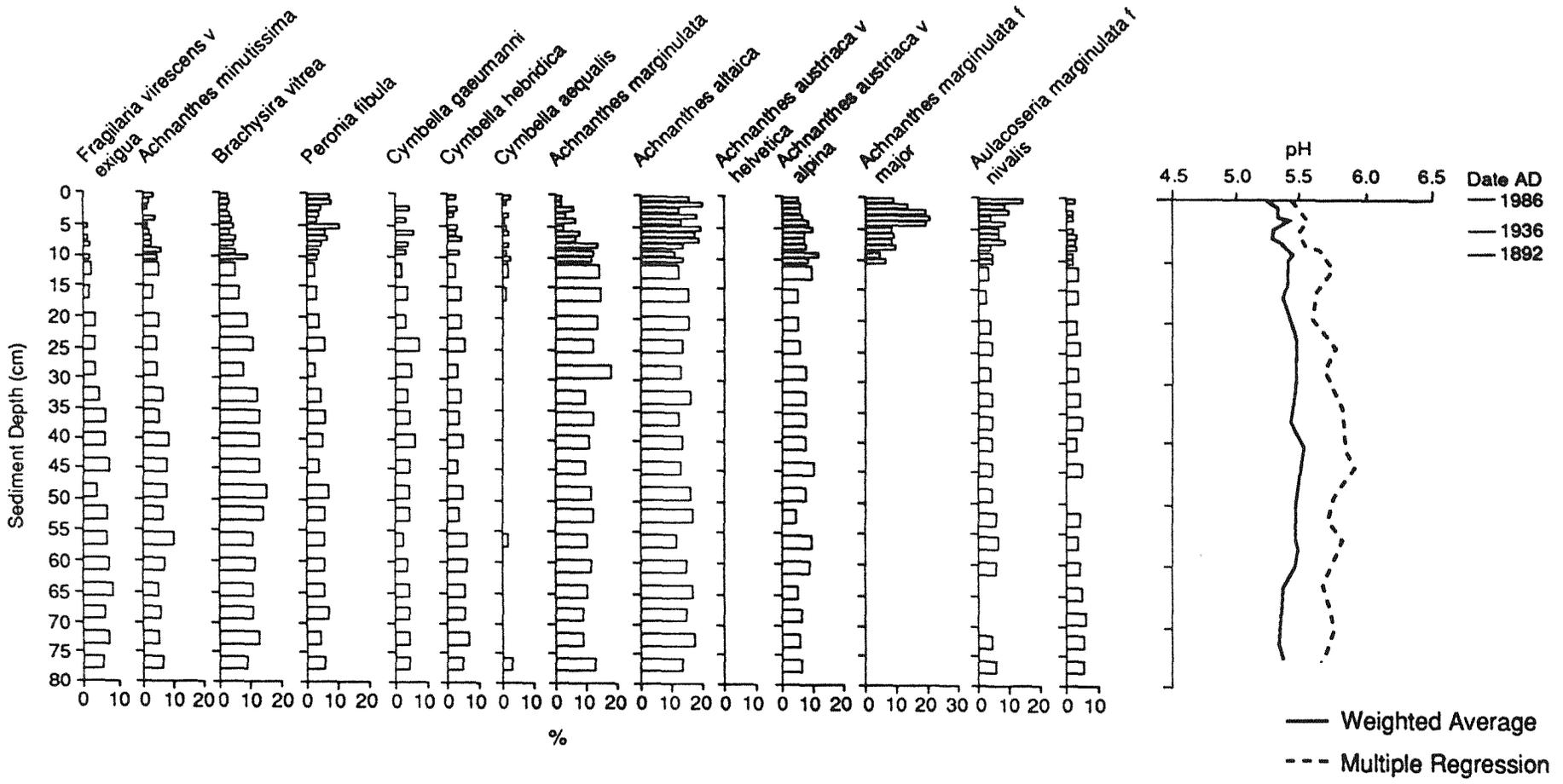


Figure 16 L. Uaine: diatom concentration ($\times 10^8 \text{ g}^{-1}$), showing 95% confidence limits

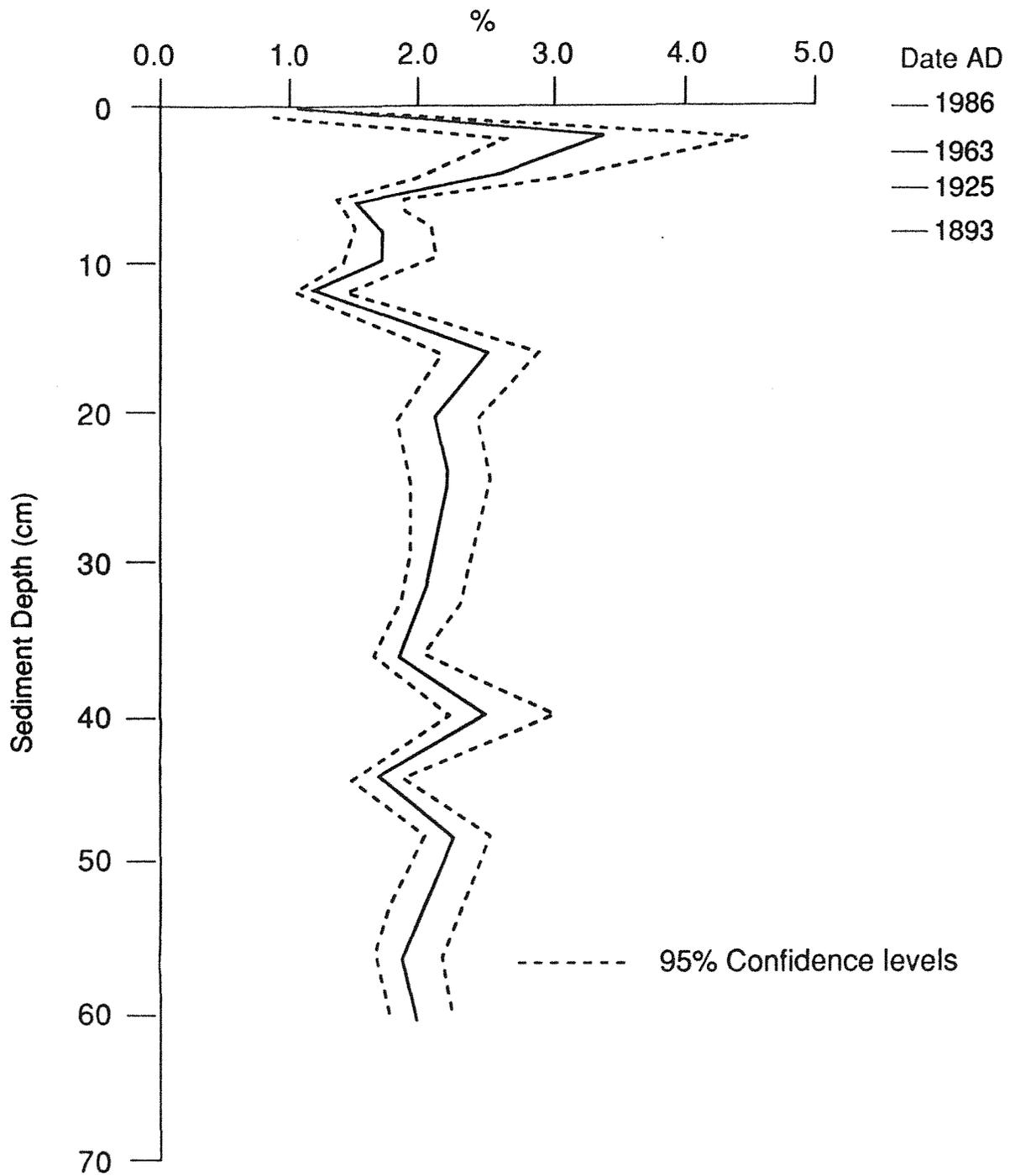


Figure 17 L. Uaine: summary pollen diagram

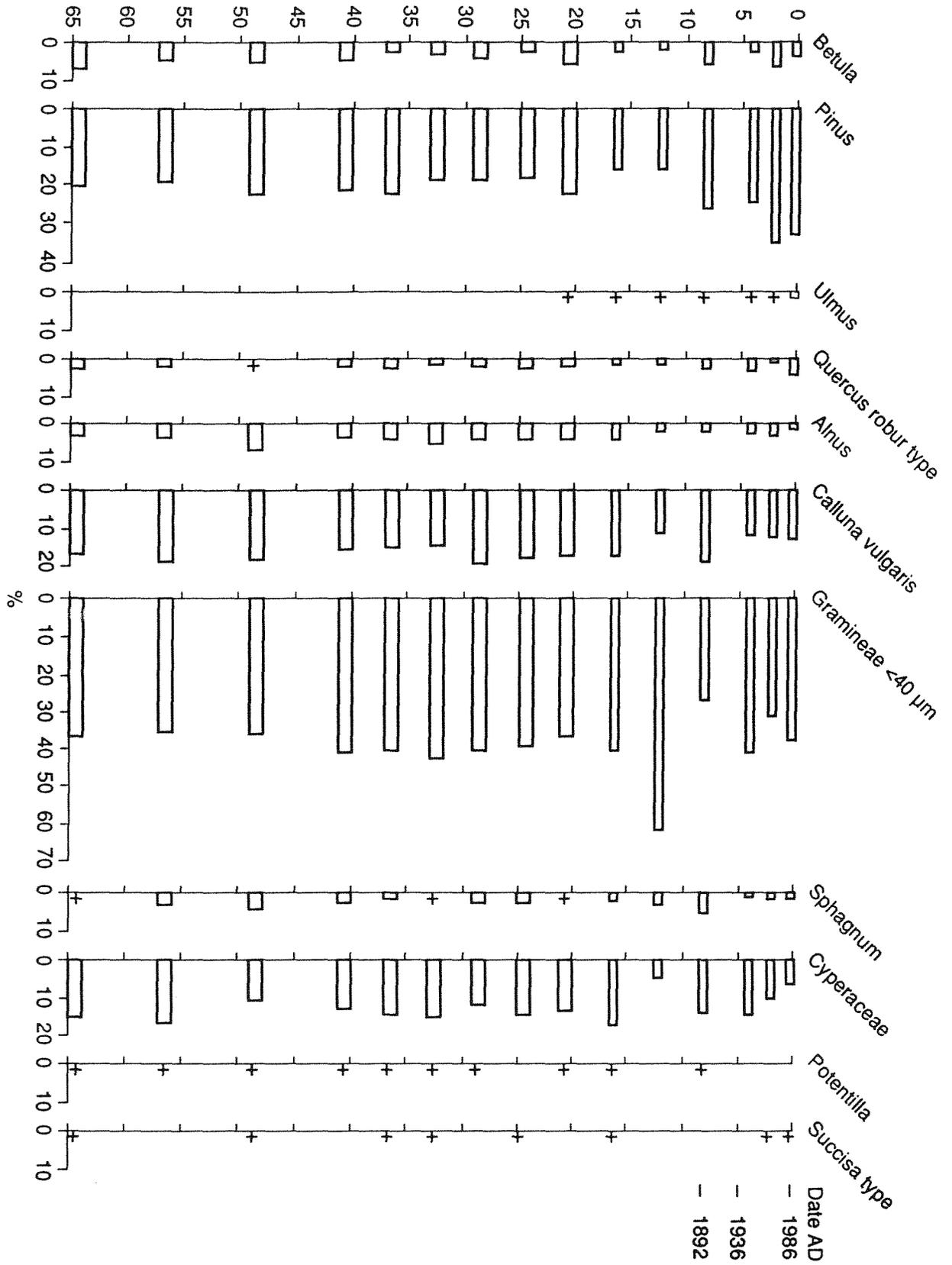


Figure 18 L. Uaine: carbonaceous particle profile ($\times 10^3 \text{ gDM}^{-1}$)

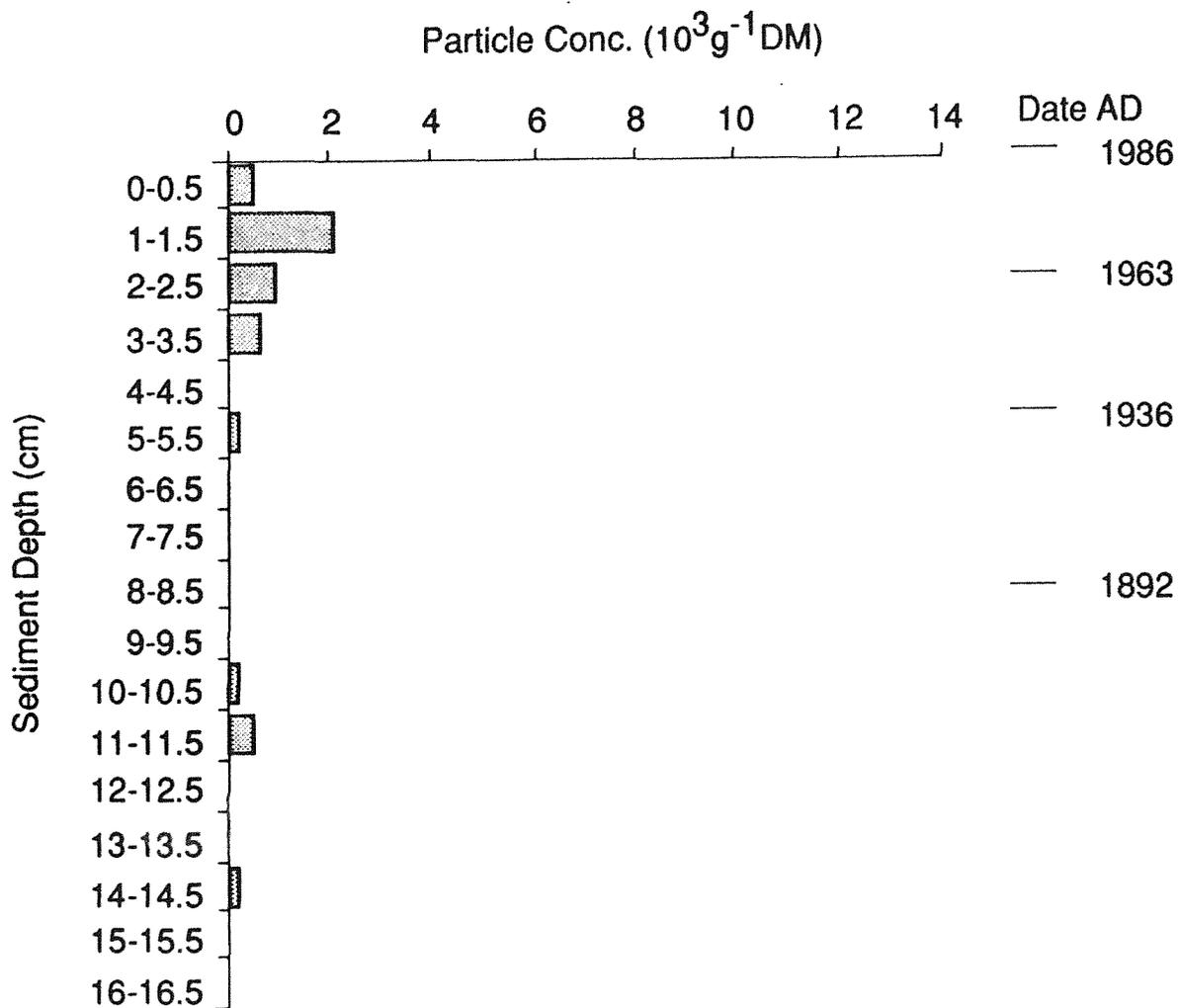


Figure 19 L. Uaine: sediment magnetic record

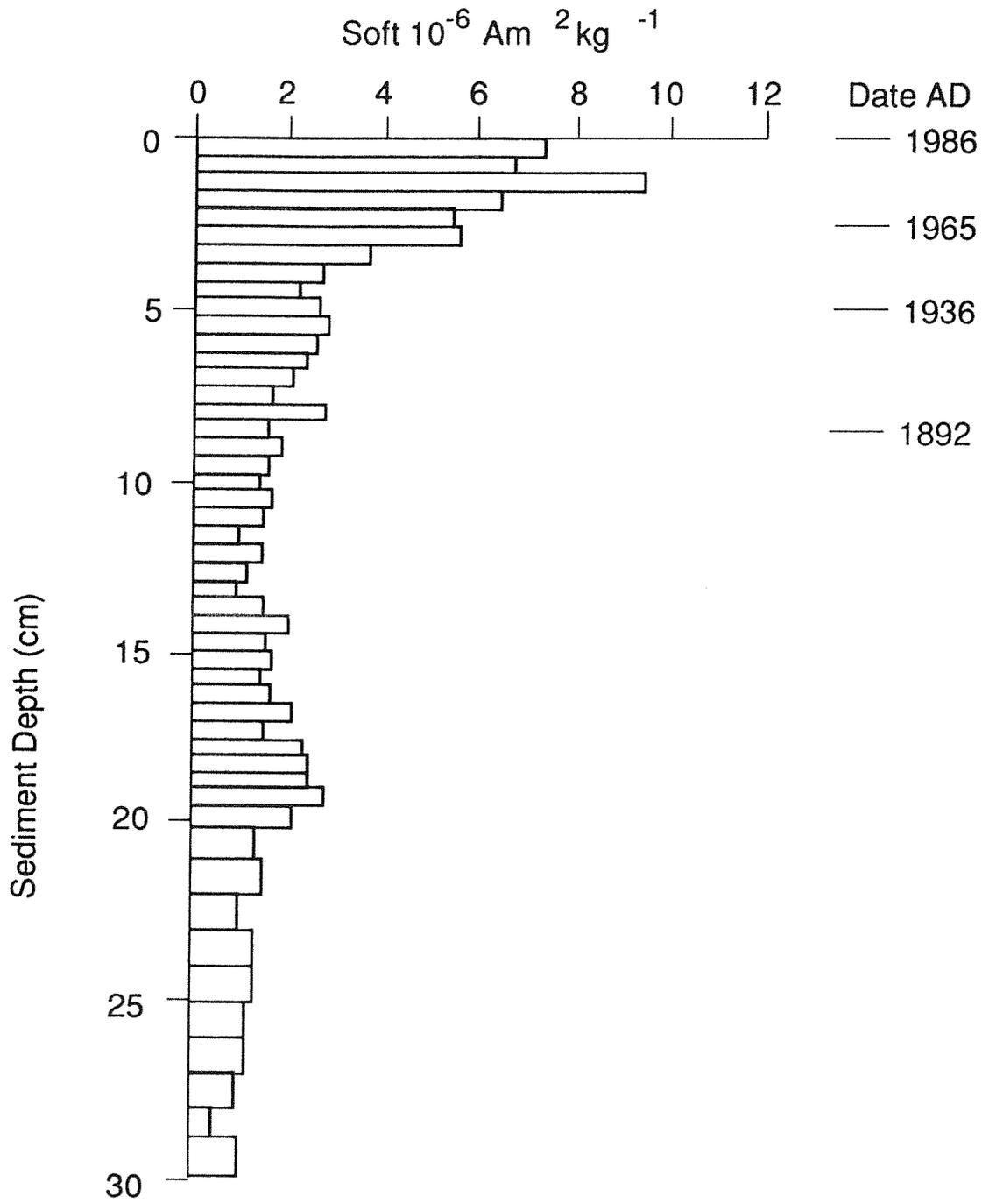


Figure 20

L. Uaine: the variation of major ion concentrations with depth

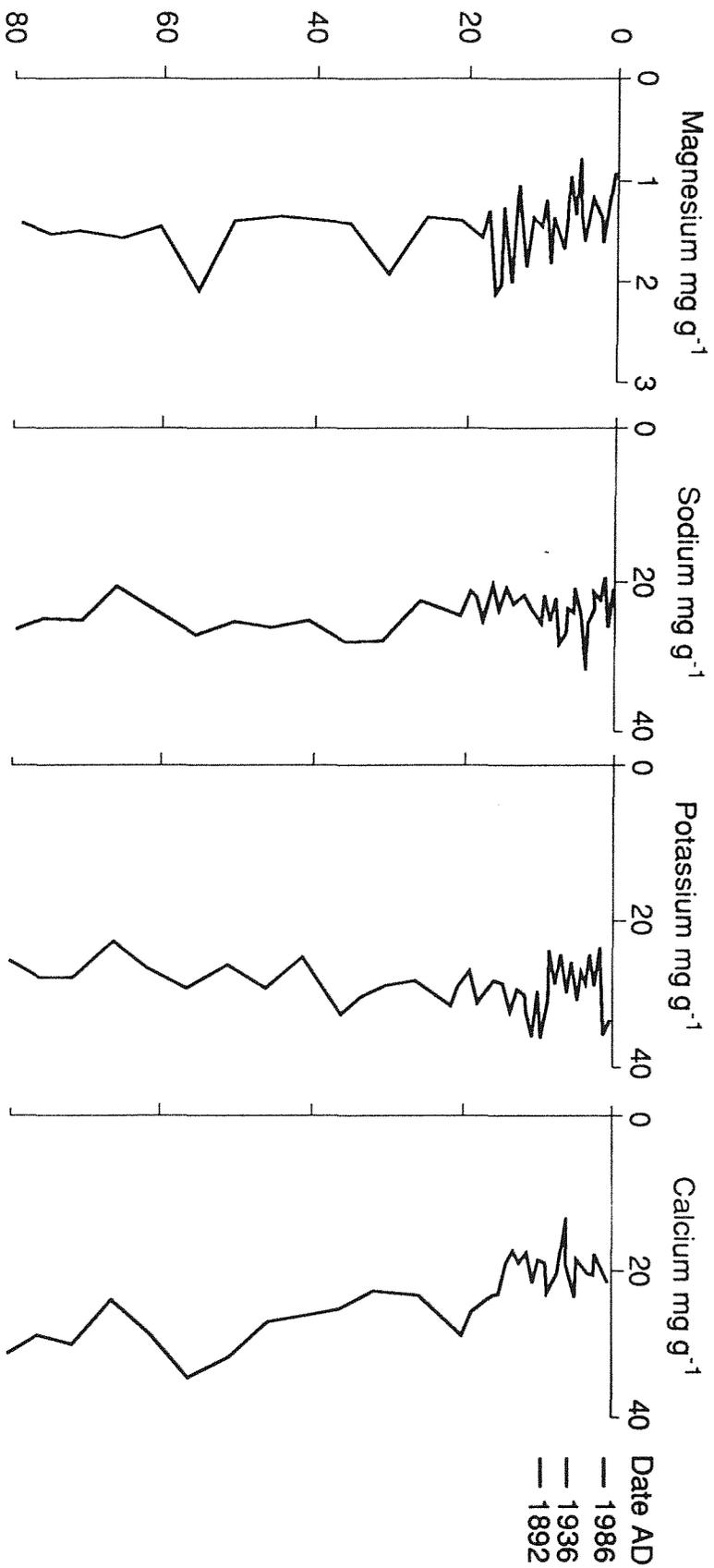


Figure 21 L. Uaine: the variation of trace metal concentrations with depth

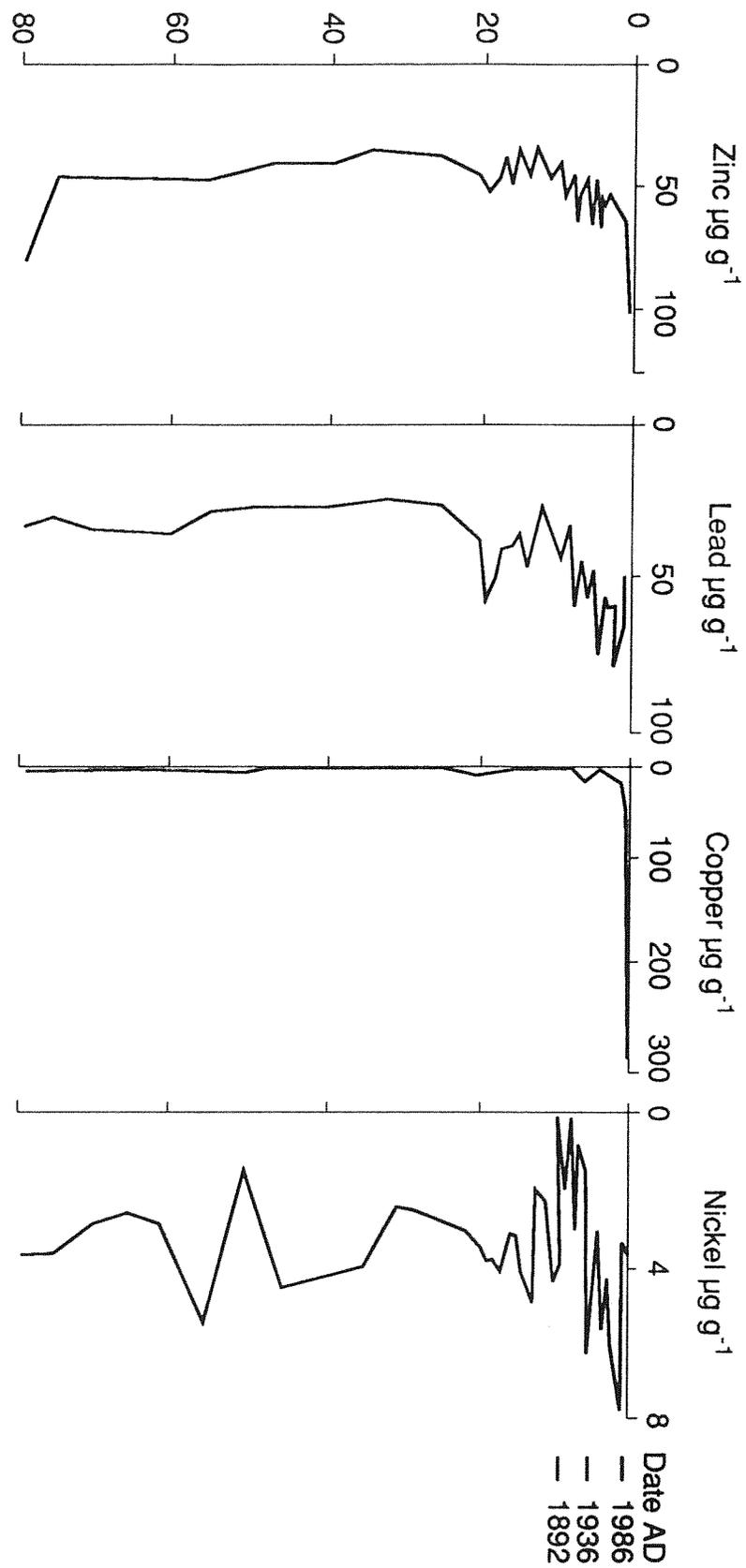
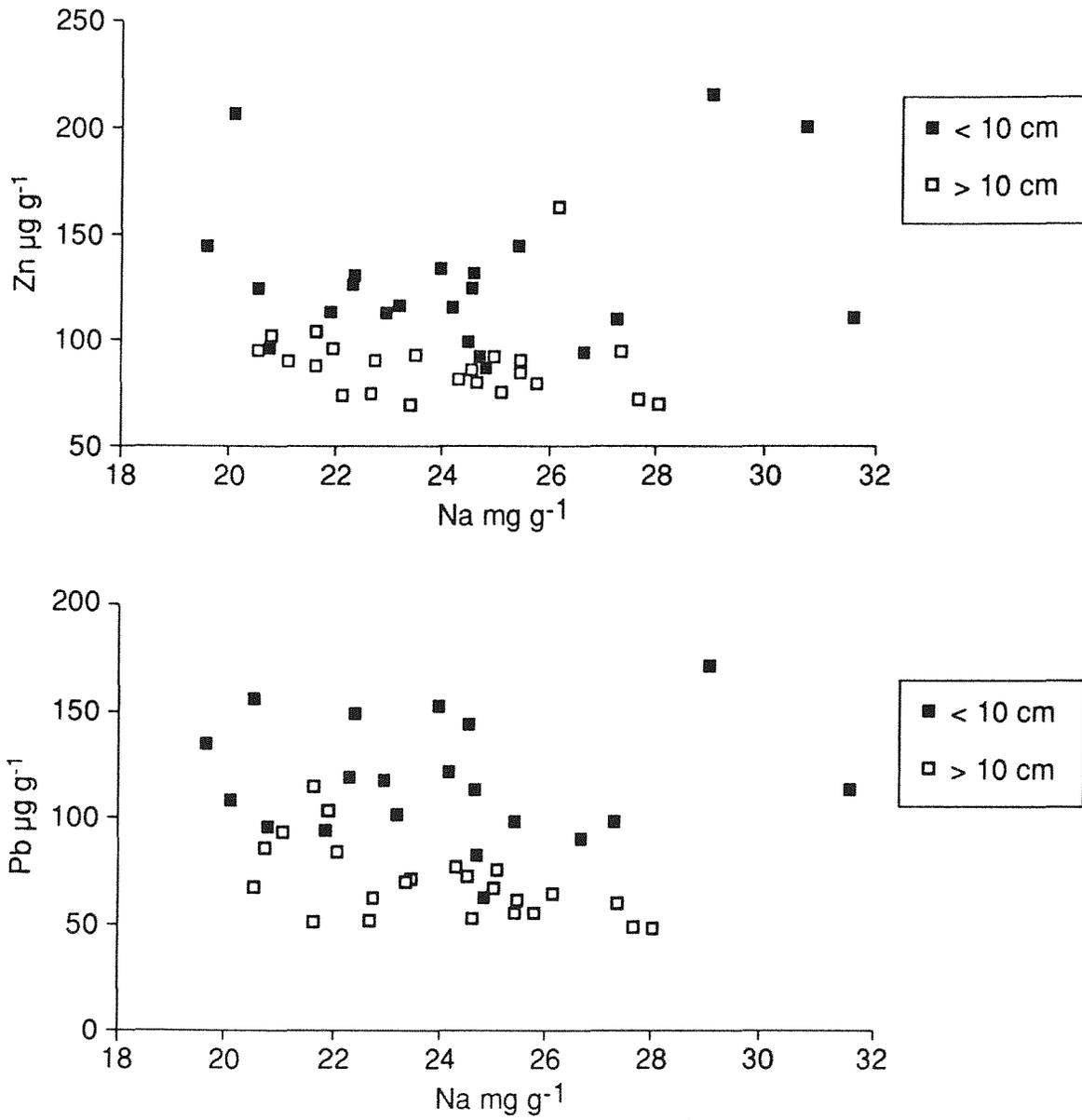


Figure 22

L. Uaine: the relationships between zinc and sodium and lead and sodium concentrations in the sediments



4.3 Loch nan Eun

Bathymetry

The bathymetric survey indicates a deep (maximum depth 23 m) single basin offset to the south west towards the back wall of the corrie in which the lake lies (Figure 23). Sediment cores were taken in 22 m of water.

Lithostratigraphy

The sediment is inorganic diatomaceous mud, with low percentage LOI values throughout the core (Figure 24). Below 25 cm it is dark greyish brown in colour with LOI values of around 20%. Above 25 cm there is a colour change to a black sediment associated with a decrease in the LOI to about 15%, and a sandy lens. LOI values then increase slowly towards the top of the core. There is also an increase in dry weight and wet density values above 25 cm which probably represents a period of increased catchment erosion.

Dating

^{210}Pb and ^{226}Ra results are given in Table 10. ^{137}Cs and ^{241}Am data are presented in Table 11, the short-lived Chernobyl fallout isotope ^{134}Cs was not detected.

The core has high levels of ^{226}Ra activity and a large variation in ^{226}Ra activity with depth. As a consequence of this it is difficult to obtain precise determinations of the unsupported ^{210}Pb activity except near the top of the core and this severely limits the accuracy of the radiometric dates of the older sediments. Nevertheless, the results do indicate that the unsupported ^{210}Pb activity declines more or less exponentially with depth, suggesting a reasonably constant sediment accumulation rate. Above 5 cm, dated 1903, there is little significant divergence between dates given by the CRS and CIC dating models (Appleby and Oldfield 1978). These are shown in Figure 25. Mean accumulation rates have been calculated using both the dating models. The dates given in Table 12 have been calculated using the average value of $0.0092 \text{ g cm}^{-2} \text{ y}^{-1}$ given by these two methods.

The distribution of the ^{137}Cs inventory suggests that 1963 should be placed at a depth of about 1.75 cm, which is in reasonable accord with the ^{210}Pb dates (Figure 25).

Table 10 Loch Nan Eun: ^{210}Pb and ^{226}Ra data

Depth	Dry mass	^{210}Pb Conc.				^{226}Ra Conc.	
		Total		Unsupp.			
cm	g cm^{-2}	pCi g^{-1}	\pm	pCi g^{-1}	\pm	pCi g^{-1}	\pm
0.25	0.0303	26.00	1.60	23.33	1.64	2.67	0.34
0.75	0.0922	24.25	4.95	22.20	5.10	2.05	1.23
1.75	0.2270	21.64	1.29	17.79	1.33	3.85	0.34
2.25	0.3000	18.50	0.79	12.33	0.83	6.17	0.25
3.25	0.4650	10.94	1.15	2.21	1.21	8.73	0.36
4.25	0.6672	9.36	0.94	2.87	0.97	6.49	0.23
5.25	0.8546	5.88	0.67	-0.06	0.69	5.94	0.18
7.25	1.1974	5.75	0.67	1.28	0.71	4.47	0.22
9.75	1.6135	5.32	0.70	0.18	0.73	5.14	0.19

Table 11 Loch Nan Eun: ^{137}Cs and ^{241}Am data

Depth	^{137}Cs Conc.		^{241}Am Conc.	
	pCi g^{-1}	\pm	pCi g^{-1}	\pm
0.25	6.73	0.34	0.10	0.06
0.75	10.03	1.08	0.48	0.20
1.25	6.38	0.82	0.08	0.04
1.75	5.27	0.29	0.00	0.00
2.25	4.14	0.23	0.04	0.02
3.25	2.72	0.23	0.13	0.06
4.25	1.14	0.16	0.00	0.00
5.25	0.86	0.12	0.00	0.00
7.25	0.18	0.12	0.00	0.00
9.75	0.00	0.00	0.00	0.00

Table 12 *Loch Nan Eun: ²¹⁰Pb chronology*

Depth cm	Cum. dry mass g cm ⁻²	Chronology			Sedimentation rate	
		Date AD	Age yr	±	g cm ⁻² yr ⁻¹	cm yr ⁻¹
0.00	0.0000	1986	0			
0.25	0.0303	1983	3	2		0.074
0.50	0.0621	1979	7	2		0.074
0.75	0.0939	1976	10	3		0.074
1.00	0.1258	1972	14	4		0.074
1.25	0.1576	1969	17	5		0.074
1.50	0.1960	1965	21	6		0.061
1.75	0.2344	1961	25	7		0.061
2.00	0.2729	1956	30	8		0.061
2.25	0.3113	1952	34	9		0.061
2.50	0.3497	1948	38	10		0.061
2.75	0.3881	1944	42	10	0.0092	0.061
3.00	0.4266	1940	46	11		0.061
3.25	0.4650	1936	50	12		0.061
3.50	0.5155	1930	56	10		0.048
3.75	0.5661	1925	61	12		0.048
4.00	0.6166	1919	67	13		0.048
4.25	0.6672	1914	72	14		0.048
4.50	0.7140	1909	77	16		0.048
4.75	0.7609	1903	83	17		0.048
5.00	0.8077	1903	88	18		0.048
5.25	0.8546	1900	93	20		0.048
5.50	0.8974	1898	97	20		0.048

Diatom analysis and pH reconstruction

A summary diatom diagram is shown in Figure 26. A series of clear floristic changes occur beginning at about 20 cm, where values of the circumneutral species *Cymbella lunata* fall. At 10 cm the circumneutral planktonic species *Cyclotella kuetzingiana* shows a dramatic decline, from levels of 30-40% below 10 cm to levels of <5% at 5 cm. At 6 cm the circumneutral species *Achnanthes minutissima* and *Brachysira vitrea* decline together with *Achnanthes scotica*. Above 10 cm there are increases in acidophilous taxa particularly *Achnanthes marginulata* and *A. marginulata* f. *major* and there is also an increase in the acidobiontic *Aulacoseira distans* v. *nivalis*.

Reconstructed pH values were calculated by the weighted average method (Figure 26). This suggests a slight drop in pH above 16 cm, with a more substantial fall in pH above 10 cm. This represents a pH decline from pH 6.1-6.4 below 16 cm to pH 4.9 at the top of the core, which agrees well with the current measured pH of the lake (pH 5.0). The results of multiple regression also show a steep pH decline from 5.7 to 4.7 above 10 cm. However, this major pH change can not be ^{210}Pb dated (see Table 12).

There is a decline in diatom concentration throughout the core (Figure 27) from values of around 10^9 valves g^{-1} at the bottom to values of around $2-3 \times 10^8$ valves g^{-1} at the core top. These changes do not appear to be closely associated with any lithostratigraphic features and could possibly reflect changing sediment accumulation rates. Above about 25 cm the low diatom concentrations could be due to their dilution by inwashed catchment material (see above).

Pollen analysis

Figure 28 presents a pollen diagram from this site. The core can be divided into two major zones at 20 cm. Below 20 cm the pollen spectra suggest an early to mid-postglacial vegetation type dominated by high percentages of tree pollen. The base of the core can be tentatively dated to about 7000 years BP., as values of *Alnus* rise and *Pinus* values fall with the establishment of the mid-Atlantic forests of central Scotland. At about 50 cm values of *Ulmus* pollen decline and levels of ruderals increase (eg. *Plantago lanceolata*, *Rumex*), which can be roughly dated to about 5000 years ago. Above this level there is an expansion of peatland indicator taxa (*Calluna*, Gramineae and *Sphagnum*) and intensification of forest clearance is recorded from 40 cm onwards, where values of *Plantago lanceolata* and *Rumex* reach 3-5%.

If this interpretation of the pollen diagram is correct and the bottom of the core can be dated to approximately 7000 years ago, then either this core represents an extremely low sediment accumulation rate, or there is at least one hiatus in the sediment record. The former possibility is unlikely since the core would represent a rate of accumulation ten times slower than that found at other upland Scottish lochs. The possibility that an inwash event with an accompanying increase in sediment accumulation rate occurred around 25 cm has already been discussed. This would also explain the decline in *Isoetes* at this point (Figure 28), an aquatic species known to be sensitive to changes in water transparency caused by

inwash events (Birks 1972). A hiatus could also occur at this point, since the pollen assemblages above and below 20 cm are quite different.

Carbonaceous particle analysis

Twenty sediment samples from 0-20 cm were analysed for carbonaceous particles, the results are presented in Figure 29. Carbonaceous particles are found throughout the profile, low amounts (generally $<0.4 \times 10^3 \text{ gDM}^{-1}$) are found below 7 cm. Above this level the concentration begins to increase, with a further more substantial increase above 2 cm (1956), giving values of $9.1 \times 10^3 \text{ gDM}^{-1}$ at the sediment surface.

Sediment chemistry

The high sedimentary sodium and potassium concentrations in Loch nan Eun (Figure 30) are a result of the minerogenic nature of the sediments. Other features of the lithostratigraphy also indicate the inorganic nature of the sediments (Figure 24). The major cation concentrations increase steadily from the base of the core to 10 cm and then decrease.

As a result of these changes in the basic constitution of the sediment, the trace metal concentrations would be expected to vary even the absence of contamination. This is further compounded by the high dry weight and low LOI values in the 3 to 8 cm band (Figure 24). The trace metal concentrations do increase above 10 cm depth (Figure 31), but the evidence suggests that the point where contamination begins is around 8 cm (Figure 32). This depth is before the dated portion of the core but may be extrapolated to c. 1840, and is close to the depth where the diatom results indicate that acidification began. The only source of contamination in this remote, high altitude lake is deposition from the atmosphere.

The zinc, but not lead, concentration declines rapidly above 2 cm (Figure 31) and this is probably due to a drop in the zinc sedimentation efficiency as the pH drops. This effect starts at pHs below 5.0-5.5 and this is in keeping with a current pH of 5.0 as shown by the diatom result and the measured pH.

Figure 23 L. nan Eun: bathymetry (contour intervals in metres)

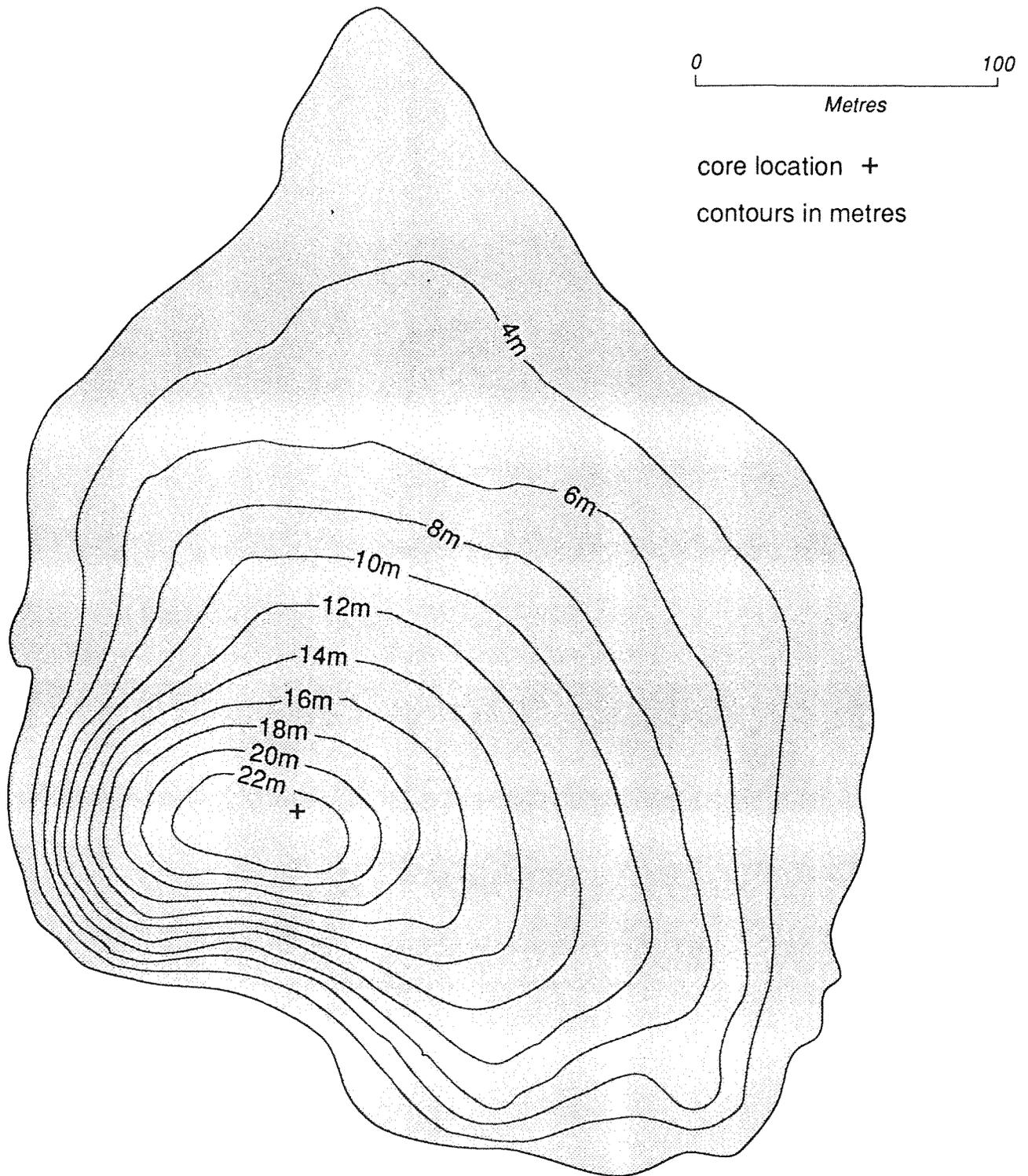


Figure 24 L. nan Eun: lithostratigraphy

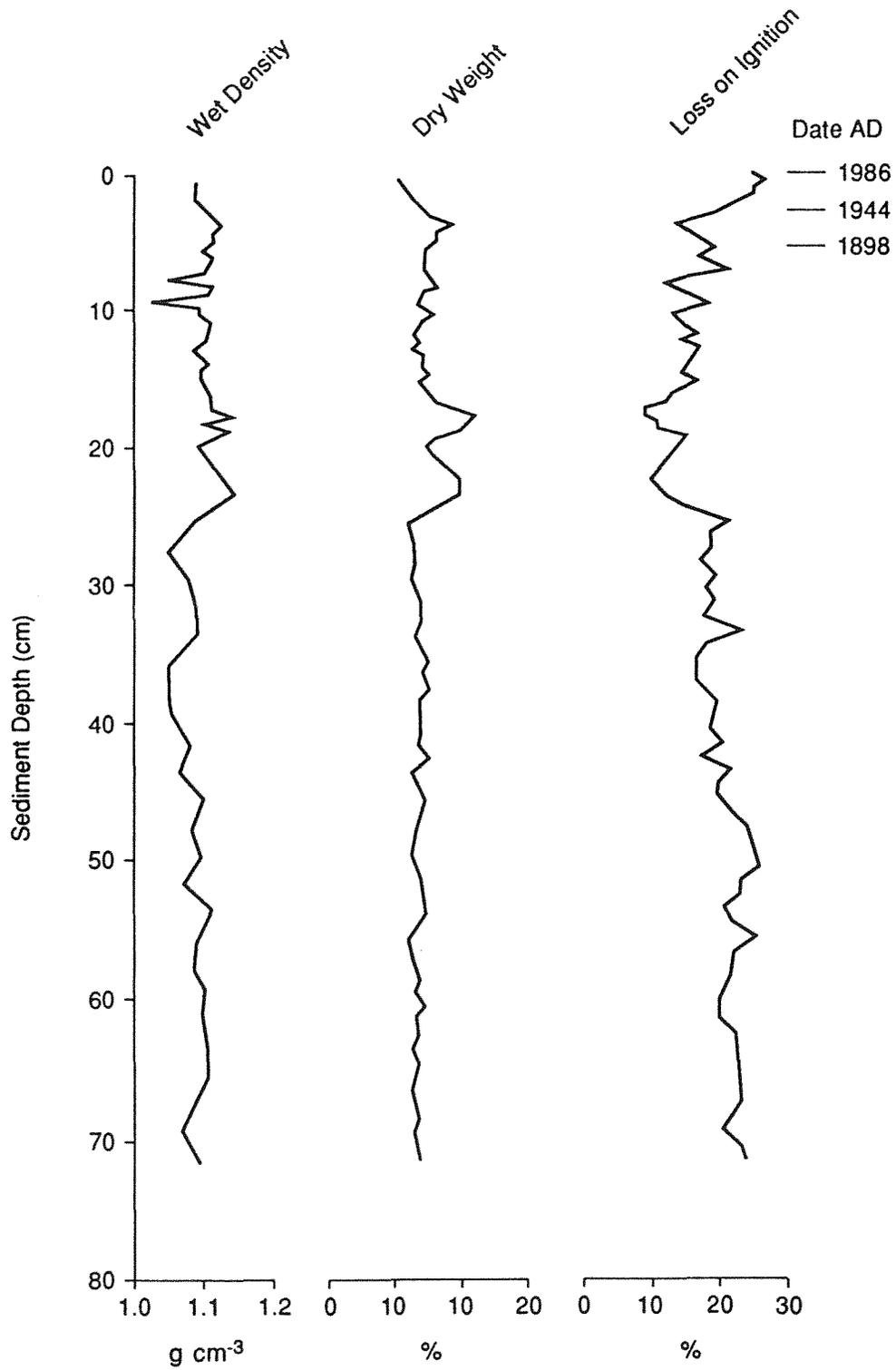


Figure 25 L. nan Eun: ^{210}Pb chronology

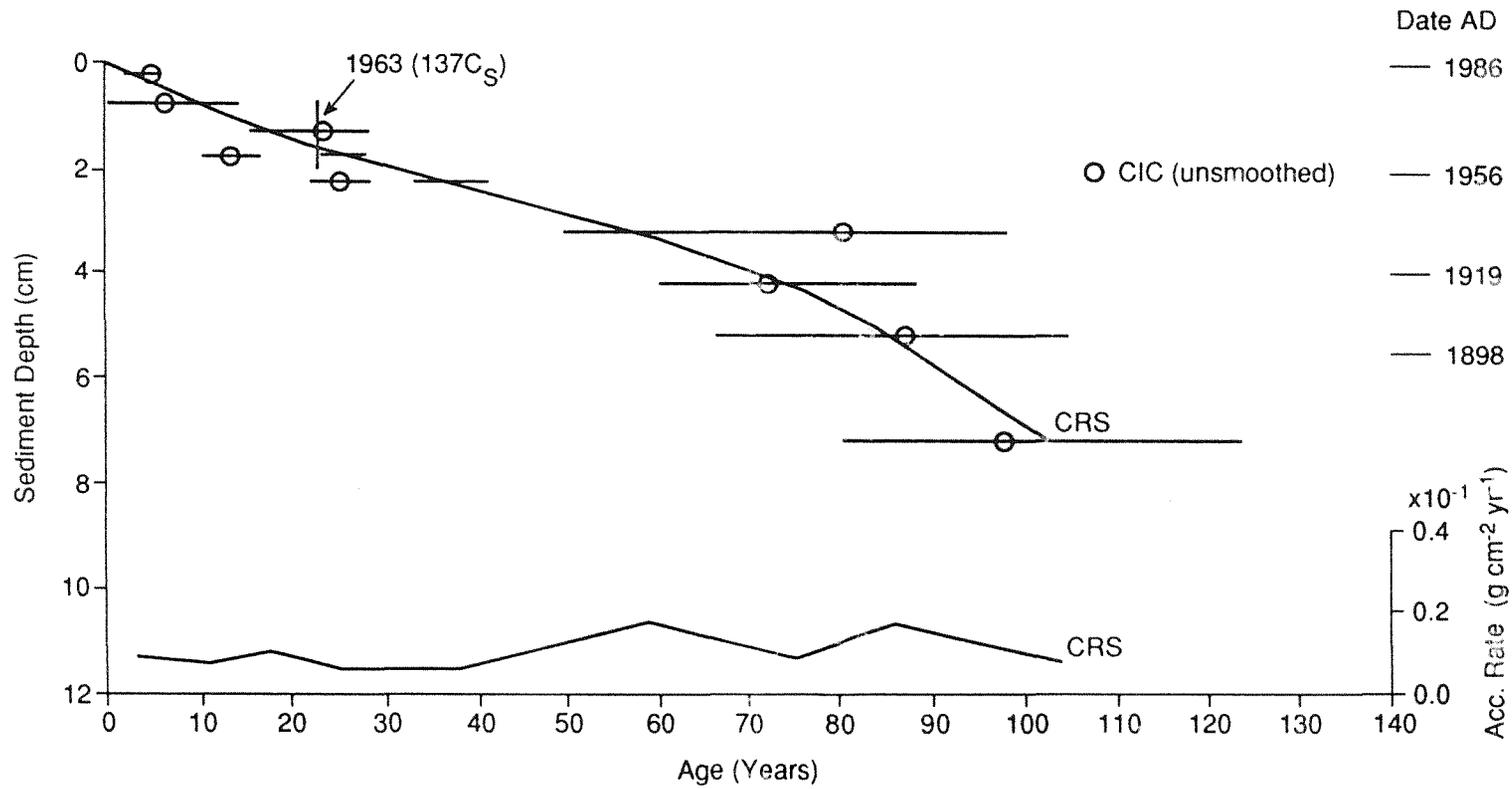


Figure 26

L. nan Eun: diatom summary diagram and pH reconstructions

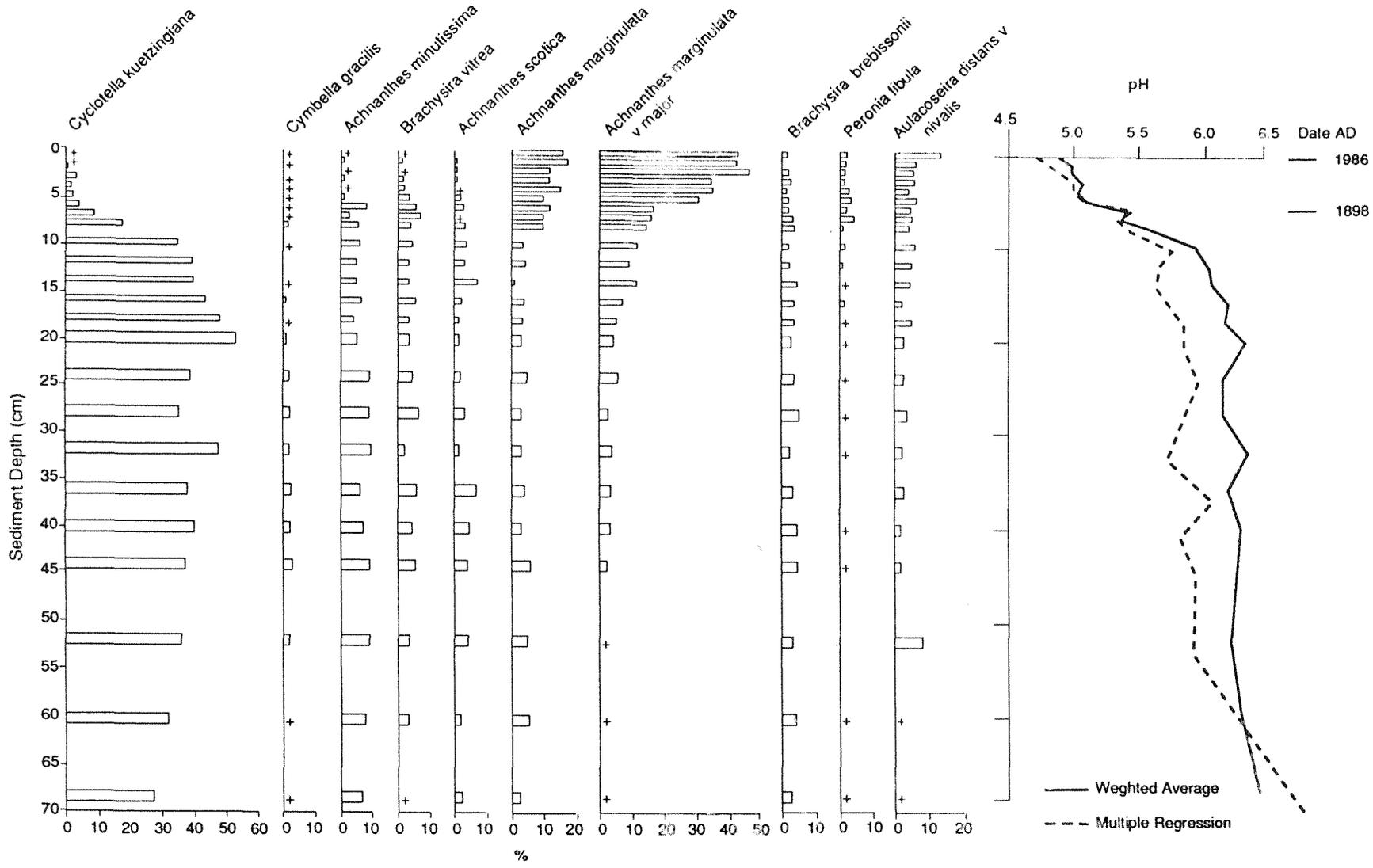


Figure 27 *L. nan Eun*: diatom concentration ($\times 10^8 \text{ g}^{-1}$), showing 95% confidence limits

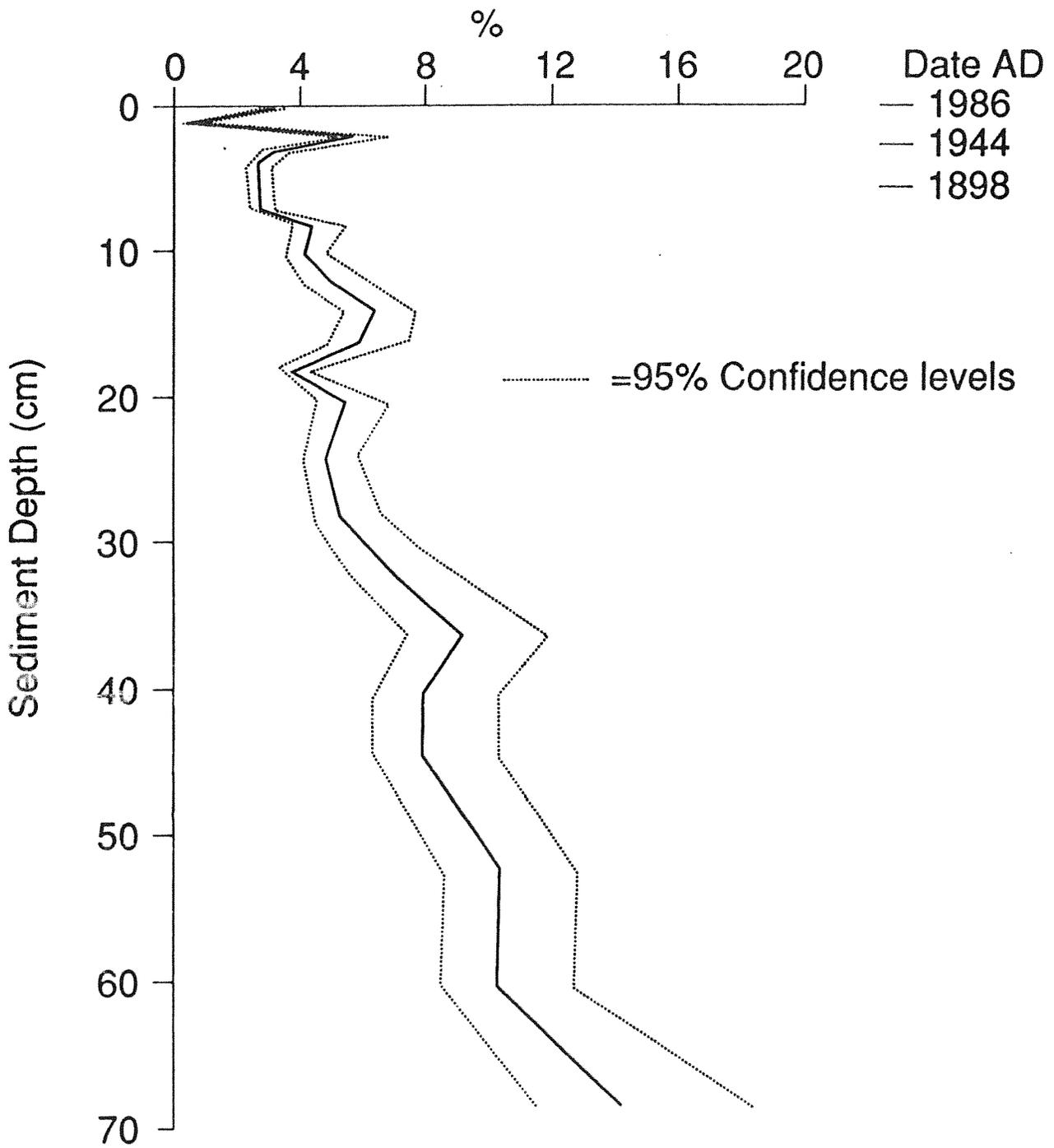


Figure 28 L. nan Eui: summary pollen diagram

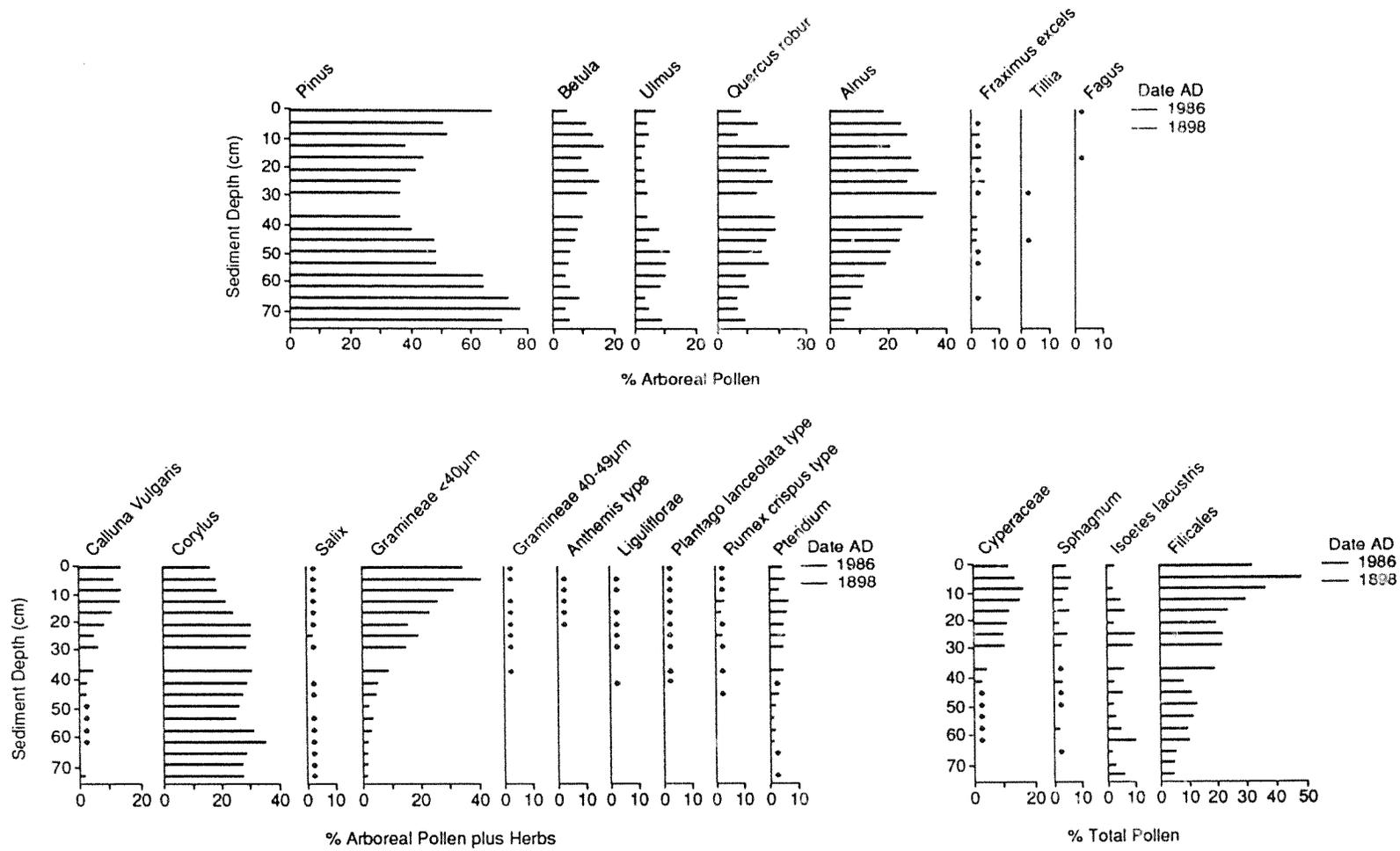


Figure 29 L. nan Eun: carbonaceous particle profile ($\times 10^3 \text{ gDM}^{-1}$)

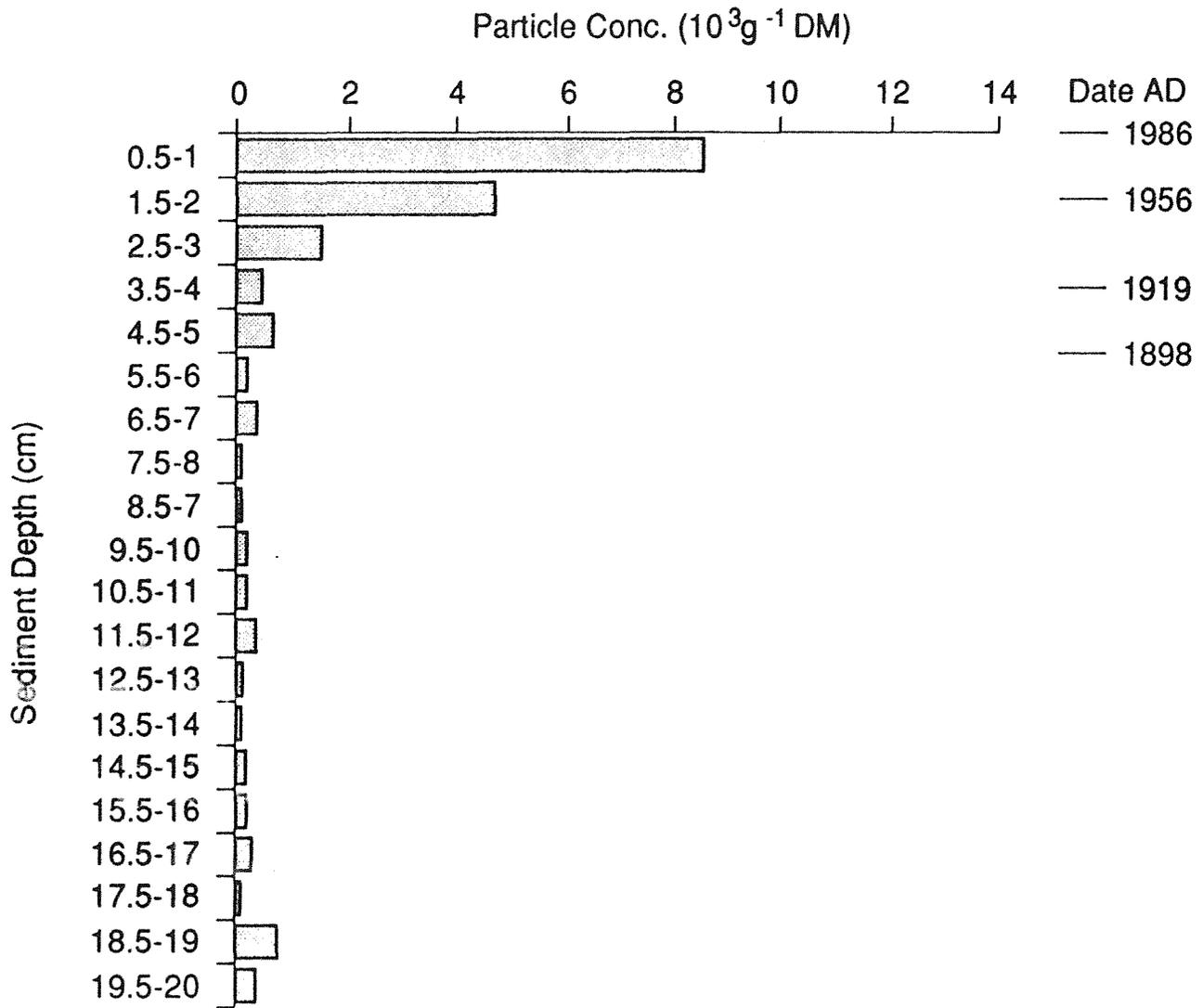


Figure 30

L. nan Eun: the variation of major ion concentrations with depth

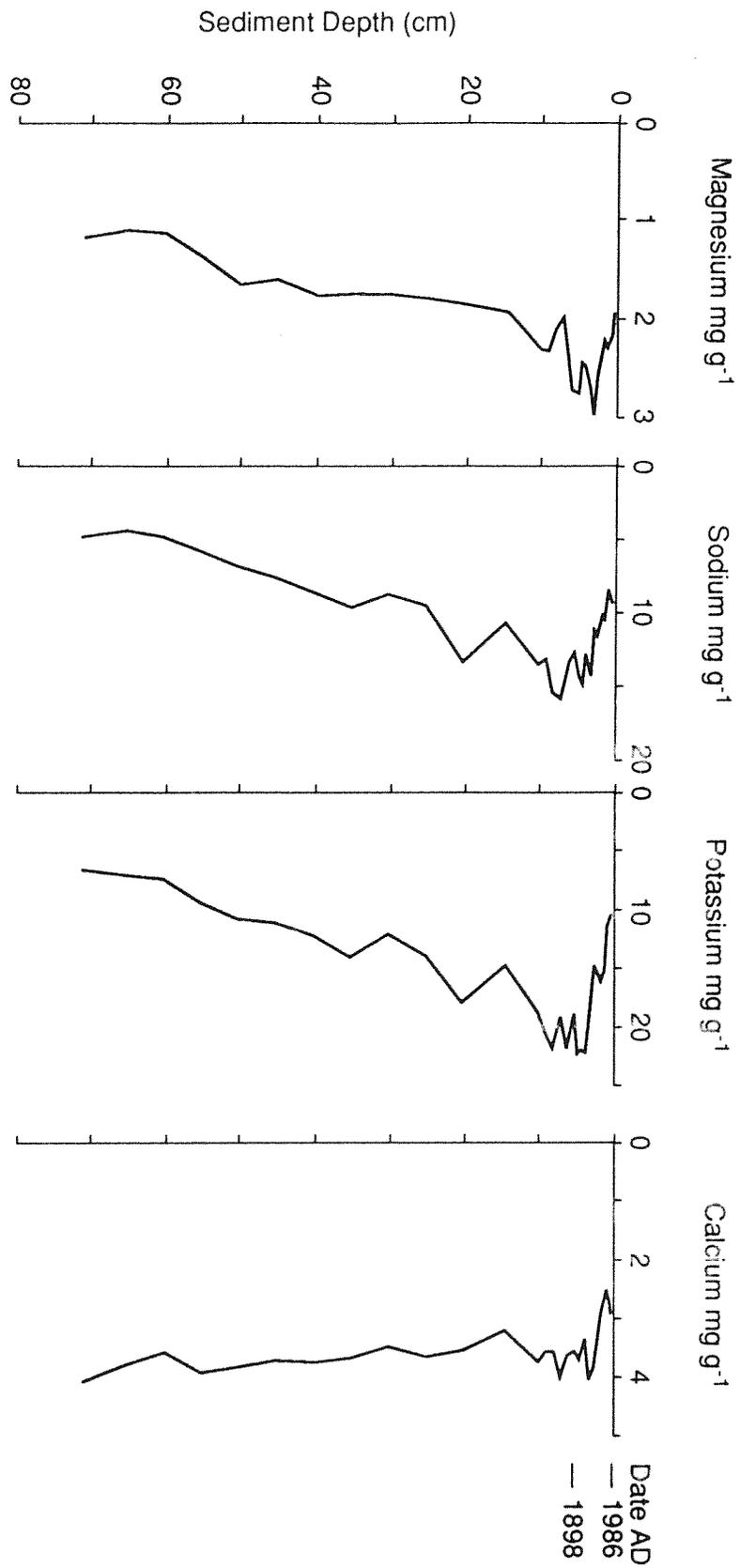


Figure 31 L. nan Eun: the variation of trace metal concentrations with depth

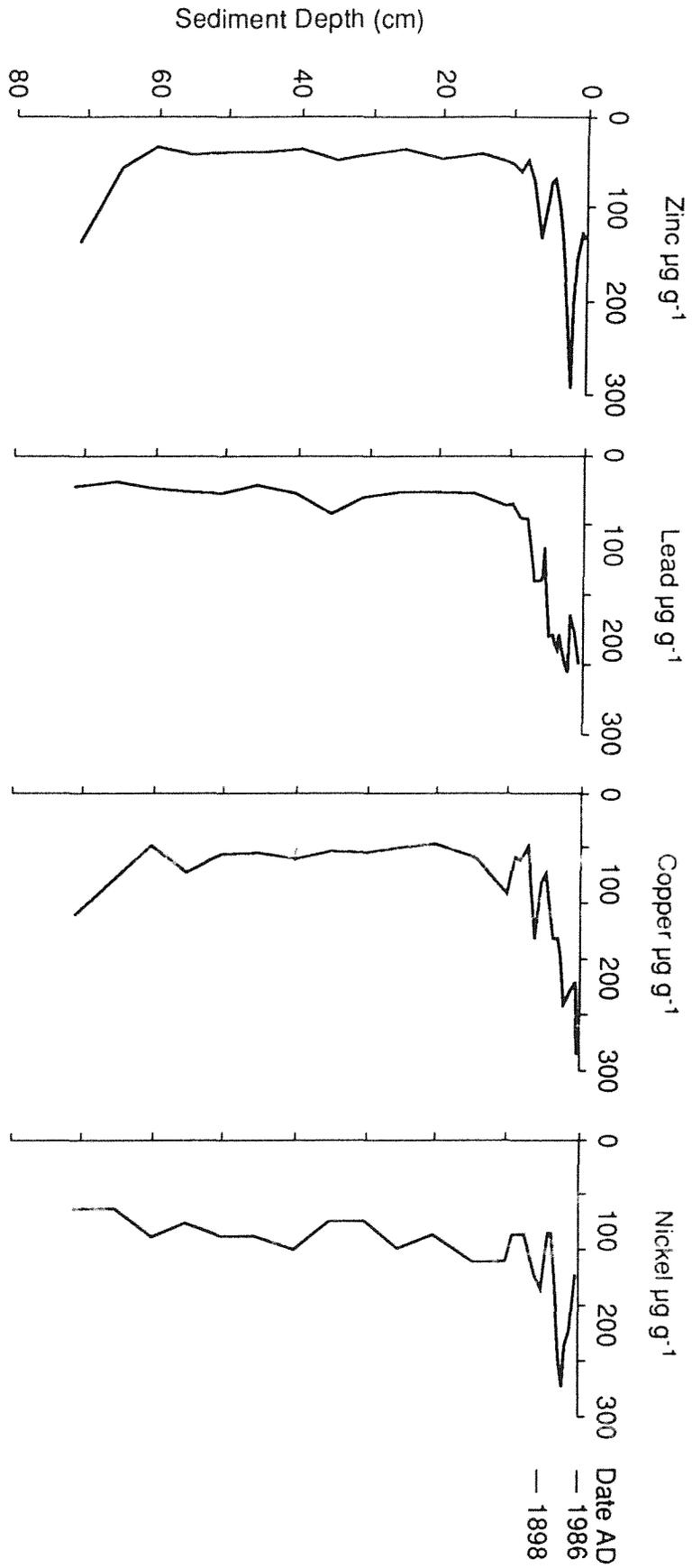
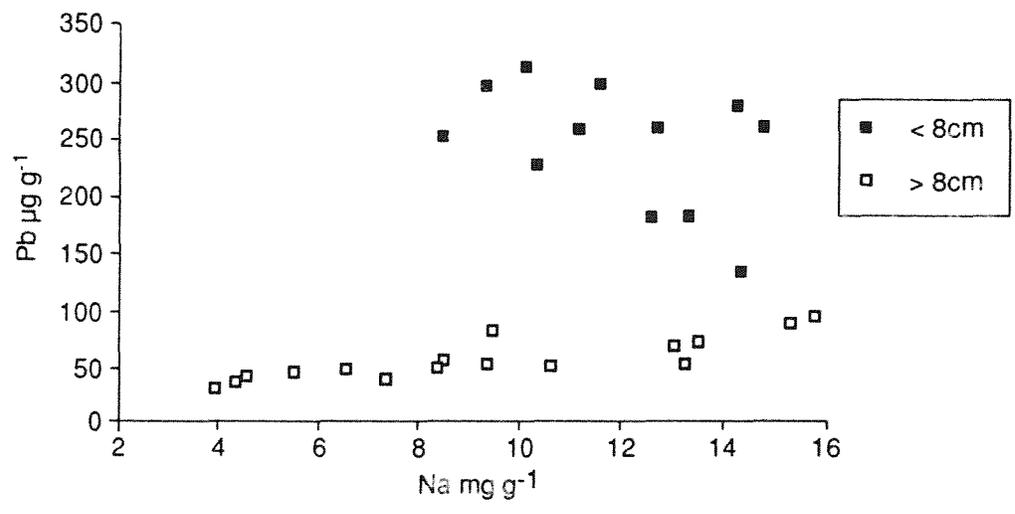
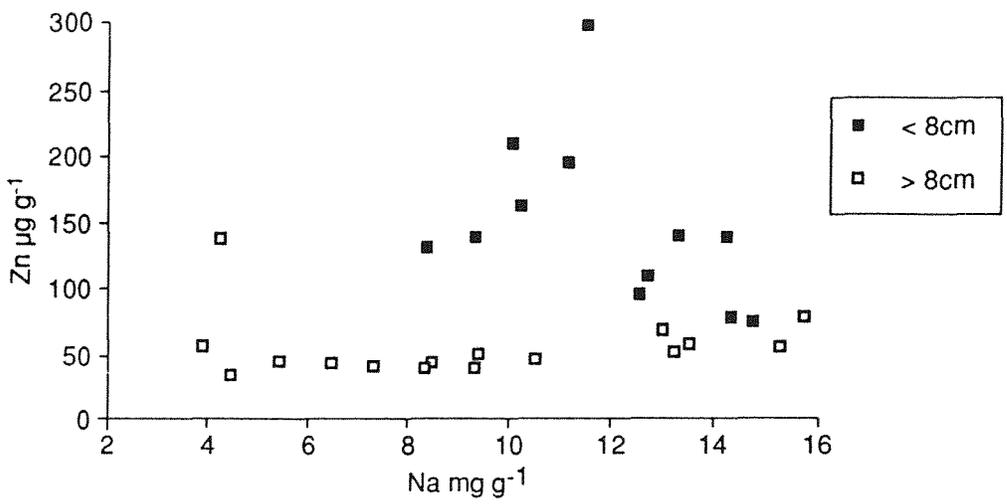


Figure 32 L. nan Eun: the relationships between zinc and sodium and lead and sodium concentrations in the sediments



4.4 Dubh Loch

Bathymetry

The bathymetry survey of Dubh Loch (Figure 33) reveals a deep (maximum depth 21 m) basin confined in the centre of the loch within the steep valley sides. To the north and south the contours rise more gently to the broader, shallower areas adjacent to the inflow and outflow. A sediment core was taken from 19 m water depth.

Lithostratigraphy

The sediment is a dark brown organic detritus mud, and LOI values fluctuate between 50-70% over most of the core (Figure 34). There is a distinct drop in the LOI at 14-15 cm, which is associated with a peak in the percentage dry weight. Wet density values are fairly uniform throughout the core.

Dating

^{210}Pb and ^{226}Ra results are given in Table 13. Results for the artificial fallout radionuclides ^{137}Cs , ^{134}Cs results (corrected for decay since the Chernobyl incident) and ^{241}Am are given in Table 14.

The unsupported ^{210}Pb inventory of the core was calculated to be 16.4 pCi cm^{-2} , representing a constant ^{210}Pb flux of $0.51 \text{ pCi cm}^{-2} \text{ yr}^{-1}$. The total ^{137}Cs inventory of the core was calculated to be 3.7 pCi cm^{-2} . Assuming a $^{137}\text{Cs}/^{134}\text{Cs}$ ratio in Chernobyl fallout of about 0.6, the component of the ^{137}Cs inventory deriving from Chernobyl fallout is estimated to be only 0.2 pCi cm^{-2} , leaving a balance of 3.5 pCi cm^{-2} attributable to nuclear weapons testing fallout.

The unsupported ^{210}Pb profile is fairly linear and there is consequently little divergence between the chronologies determined by the CRS and CIC models (Appleby and Oldfield 1978) until near the base of the core, where the relatively high ^{226}Ra activity gives rise to large uncertainties in the dating parameters. The chronologies given by these models are shown in Figure 35 and Table 15. Both models date 17 cm to c. 1920 and give a mean sediment accumulation rate since then of $0.0263 \pm 0.0042 \text{ g cm}^{-2} \text{ yr}^{-1}$. The chronology given in Table 15 has been calculated using this mean accumulation rate. The ^{210}Pb dating horizon is estimated to be c. 1890, at about 24 cm. Extrapolated dates are given down to 30 cm.

The ^{210}Pb dates suggest sediments dating from 1963 should occur at about 6-7 cm depth in the core and that 1954 should occur at about 9 cm. The ^{137}Cs data give some general support to these dates, although there is some ^{137}Cs down to 25 cm, the principle rise (representing 1954-1958) occurs at about 8 cm. In addition, when Chernobyl ^{137}Cs is excluded, the residual weapons fallout component would appear to peak at between 4-7 cm. Further support to the ^{210}Pb dates is given by the ^{241}Am data, which show a small peak at 6-7 cm, dated by ^{210}Pb to 1962-1964.

Table 13 Dubh Loch: ^{210}Pb and ^{226}Ra data

Depth cm	Dry mass g cm ⁻²	^{210}Pb Conc.				^{226}Ra Conc.	
		Total		Unsupp.		pCi g ⁻¹	±
		pCi g ⁻¹	±	pCi g ⁻¹	±		
0.25	0.0219	21.81	1.31	18.37	1.41	3.44	0.51
0.75	0.0656	23.11	1.41	20.01	1.49	3.10	0.48
2.75	0.2382	21.33	0.83	15.41	0.89	5.92	0.31
4.75	0.4358	16.44	0.95	12.07	1.01	4.37	0.34
6.75	0.6337	11.65	0.77	7.28	0.82	4.37	0.28
8.25	0.7958	12.53	0.96	8.36	1.03	4.17	0.37
9.75	0.9579	8.86	0.99	5.42	1.06	3.44	0.37
12.75	1.2626	7.49	0.79	4.67	0.84	2.82	0.28
16.75	1.7364	5.93	0.69	2.77	0.74	3.16	0.28
19.75	2.0567	5.15	0.41	1.09	0.44	4.06	0.17
24.50	2.5756	4.77	0.47	1.92	0.50	2.85	0.17
29.50	3.1232	3.25	0.26	0.20	0.31	3.05	0.17
34.50	3.6586	3.24	0.30	-0.60	0.33	3.84	0.14
38.50	4.0245	3.53	0.22	0.51	0.26	3.02	0.14

Table 14 Dubh Loch: ^{137}Cs , ^{134}Cs and ^{241}Am data

Depth cm	^{137}Cs Conc.		^{134}Cs Conc.		^{241}Am Conc.	
	pCi g ⁻¹	±	pCi g ⁻¹	±	pCi g ⁻¹	±
0.25	6.60	0.29	1.46	0.34	0.00	0.00
0.75	5.73	0.30	0.78	0.41	0.00	0.00
2.75	4.15	0.17	0.00	0.00	0.00	0.00
4.75	3.60	0.19	0.00	0.00	0.00	0.00
6.75	3.34	0.17	0.00	0.00	0.06	0.03
8.25	1.82	0.18	0.00	0.00	0.00	0.00
9.75	0.54	0.17	0.00	0.00	0.00	0.00
12.75	0.17	0.14	0.00	0.00	0.00	0.00
16.75	0.22	0.12	0.00	0.00	0.00	0.00
19.75	0.13	0.07	0.00	0.00	0.00	0.00
24.50	0.14	0.07	0.00	0.00	0.00	0.00
29.50	0.00	0.00	0.00	0.00	0.00	0.00
34.50	0.00	0.00	0.00	0.00	0.00	0.00
38.50	0.00	0.00	0.00	0.00	0.00	0.00

Table 15 *Dubh Loch: ²¹⁰Pb chronology*

Depth cm	Cum. dry mass g cm ⁻²	Chronology			Sedimentation rate		
		Date AD	Age yr	±	g cm ⁻² yr ⁻¹	cm yr ⁻¹	± (%)
0.00	0.0000	1987	0		0.0263	0.250	16
1.00	0.0872	1984	3	1	0.0263	0.250	16
2.00	0.1735	1980	7	1	0.0263	0.250	16
3.00	0.2629	1977	10	2	0.0263	0.250	16
4.00	0.3617	1973	14	2	0.0263	0.250	16
5.00	0.4605	1969	18	3	0.0263	0.250	16
6.00	0.5595	1966	21	3	0.0263	0.250	16
7.00	0.6607	1962	25	4	0.0263	0.250	16
8.00	0.7688	1958	29	5	0.0263	0.250	16
9.00	0.8768	1954	33	5	0.0263	0.250	16
10.00	0.9833	1950	37	6	0.0263	0.250	16
11.00	1.0849	1946	41	7	0.0263	0.250	16
12.00	1.1864	1942	45	7	0.0263	0.250	16%
13.00	1.2922	1938	49	8	0.0263	0.250	16
14.00	1.4107	1933	54	9	0.0263	0.250	16
15.00	1.5291	1929	58	9	0.0263	0.250	16
16.00	1.6476	1924	63	10	0.0263	0.250	16
17.00	1.7631	1920	67	11	0.0263	0.250	16
18.00	1.8699	1916	71	11	0.0263	0.250	16
19.00	1.9766	1912	75	12	0.0263	0.250	16
20.00	2.0840	1908	79	13	0.0263	0.250	16
21.00	2.1933	1904	83	13	0.0263	0.250	16
22.00	2.3025	1899	88	14	0.0263	0.250	16
23.00	2.4117	1895	92	15	0.0263	0.250	16
24.00	2.5210	1891	96	15	0.0263	0.250	16
26.00	2.7399	1883	104				
28.00	2.9589	1874	113				
30.00	3.1767	1866	121				

Diatom analysis and pH reconstruction

A summary diatom diagram is presented in Figure 36. Only very slight floristic changes occur, values of *Achnanthes minutissima* and *Fragilaria vaucheriae* decline at about 30 cm, and above this level *Cymbella aequalis*, *Cymbella hebridica*, and *Eunotia exigua* are more abundant. The circumneutral species *Brachysira vitrea* and *Tabellaria flocculosa* decline slightly above 18 cm and this is accompanied by an increase in *Achnanthes marginulata*. Above 3 cm the acidobiontic species *Tabellaria quadrisepitata* increases.

The results of weighted average pH reconstruction (Figure 36) suggest a slight acidification from 30 cm (c 1866). There is a reconstructed pH of 5.3 below 30 cm with a drop in pH of about 0.4 pH unit to the present day reconstructed pH of 4.9. The modern measured pH is 5.3. Multiple regression reconstructs surface pH at 5.4 and produces higher values than weighted averaging throughout the core.

Diatom concentrations increase from the bottom of the core to 26 cm (Figure 37), they then decrease to 14 cm before increasing again towards the sediment surface. There does not appear to be any obvious correlation with the lithostratigraphy and sediment accumulation rates are fairly constant over this period.

Pollen analysis

On the whole the pollen profile is very stable (Figure 38), an open peatland community must have existed throughout the period represented by the entire core. The tree pollen is dominated by *Pinus* and there is a slight increase in Gramineae over *Calluna* in the top 20 cm.

Carbonaceous particle analysis

22 sediment samples from 0-24 cm were analysed for carbonaceous particles. The results are presented in Figure 39. Below 16 cm (c. 1924) there are very low levels of carbonaceous particles ($<0.5 \times 10^3 \text{ gDM}^{-1}$). From 16-8 cm the concentrations increase slightly and above 8 cm (c. 1958) they increase sharply to give a peak of $15.2 \times 10^3 \text{ gDM}^{-1}$ at 2.5-3 cm (c. 1978). The concentrations then fall slightly to the surface. The carbonaceous particle record therefore indicates the start of atmospheric contamination in the 1920s, with an increase after 1958 and a peak in the late 1970s, which is in general agreement with the ^{210}Pb and ^{241}Am dating.

Magnetics analysis

The magnetically 'soft' magnetite component indicated by SIRM-IRM^{-20mT} increases in two steps, initially at c. 13 cm (1938) and then more steeply from c. 4 cm (1973) (Figure 40).

Sediment chemistry

The concentrations of major cations are relatively constant over the length of the core (Figure 41), suggesting that there has been no major changes in the delivery of material from the catchment to the loch. Zinc, lead and to a lesser extent copper concentrations, increase from c. 30 cm (1866) (Figures 42 and 43) indicating contamination from the atmosphere. Although the amount of contamination is low, it is larger than that established at Loch Coire and Lochan, Lochan Uaine and Loch nan Eun.

Figure 33 Dubh Loch: bathymetry (contour intervals in metres)

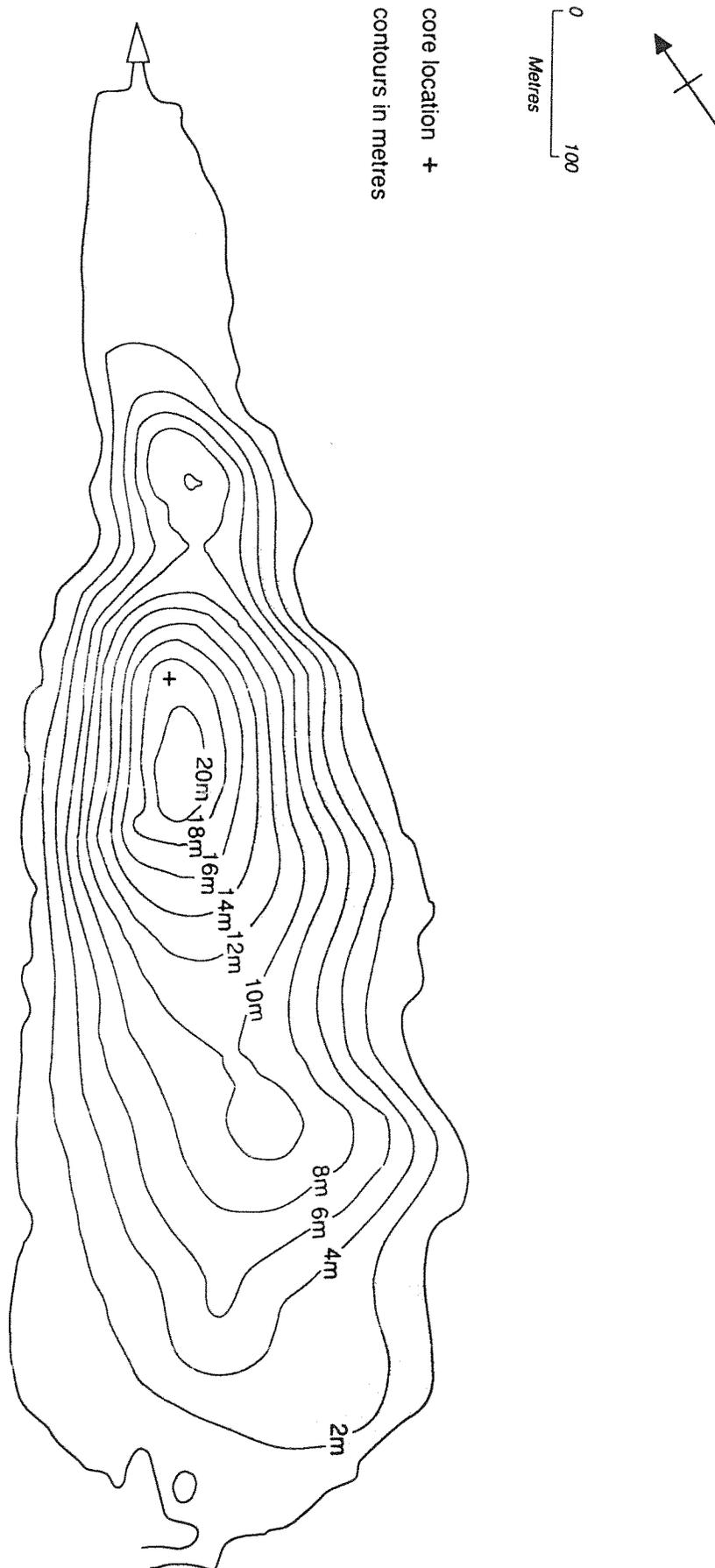


Figure 34 Dubh Loch: lithostratigraphy

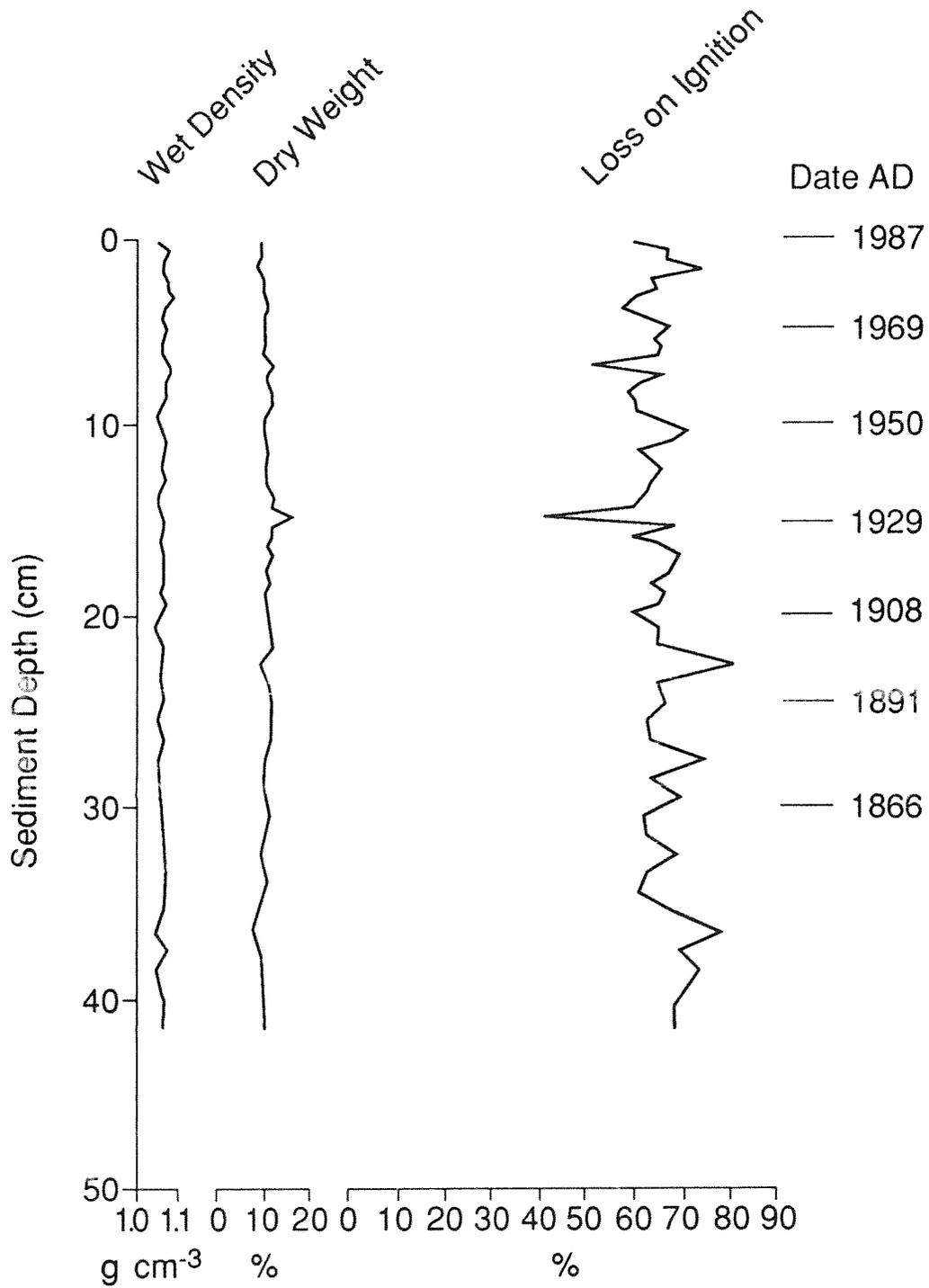


Figure 35 Dubh Loch: ^{210}Pb chronology

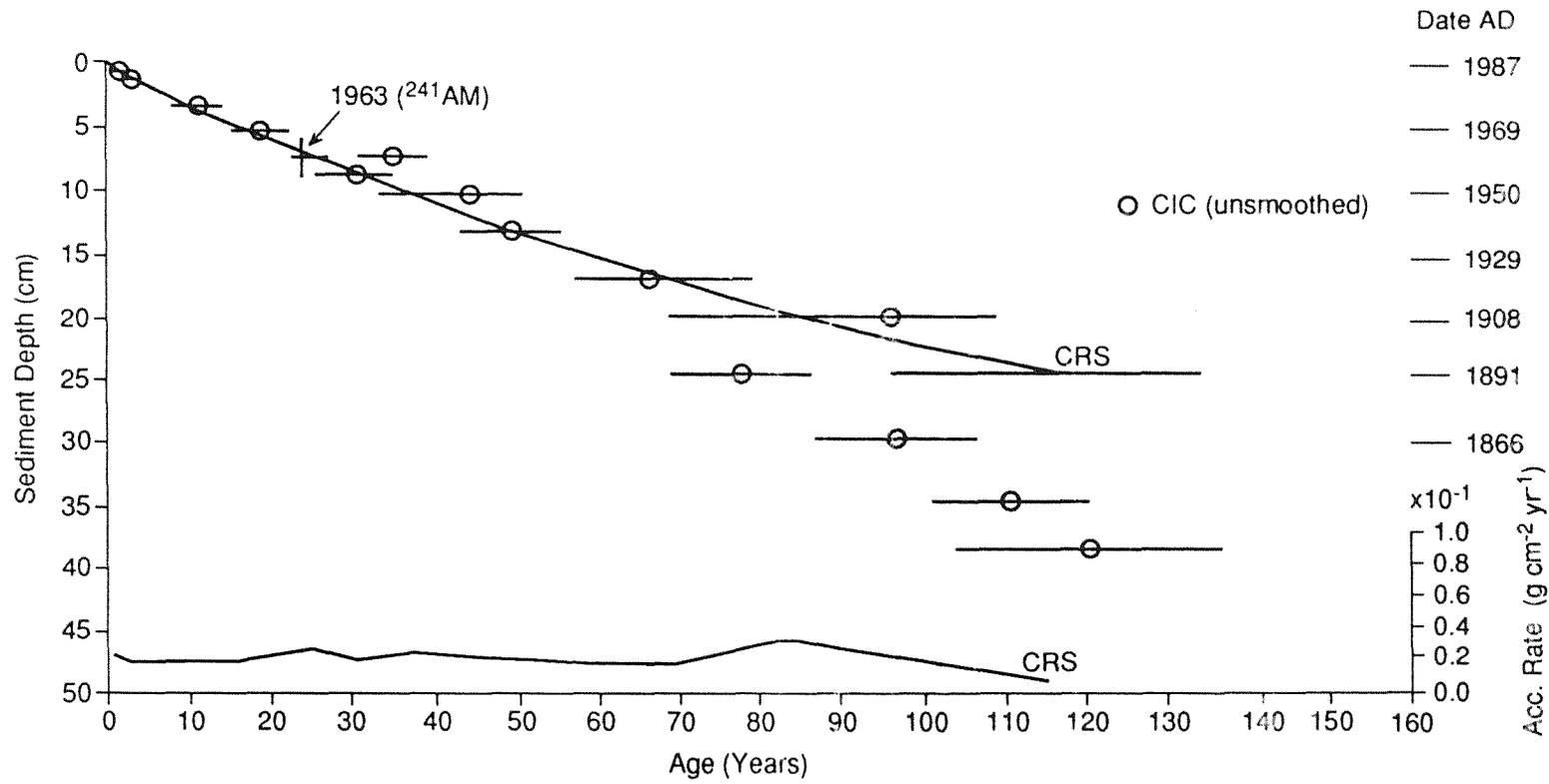


Figure 36 Dubh Loch: diatom summary diagram and pH reconstruction

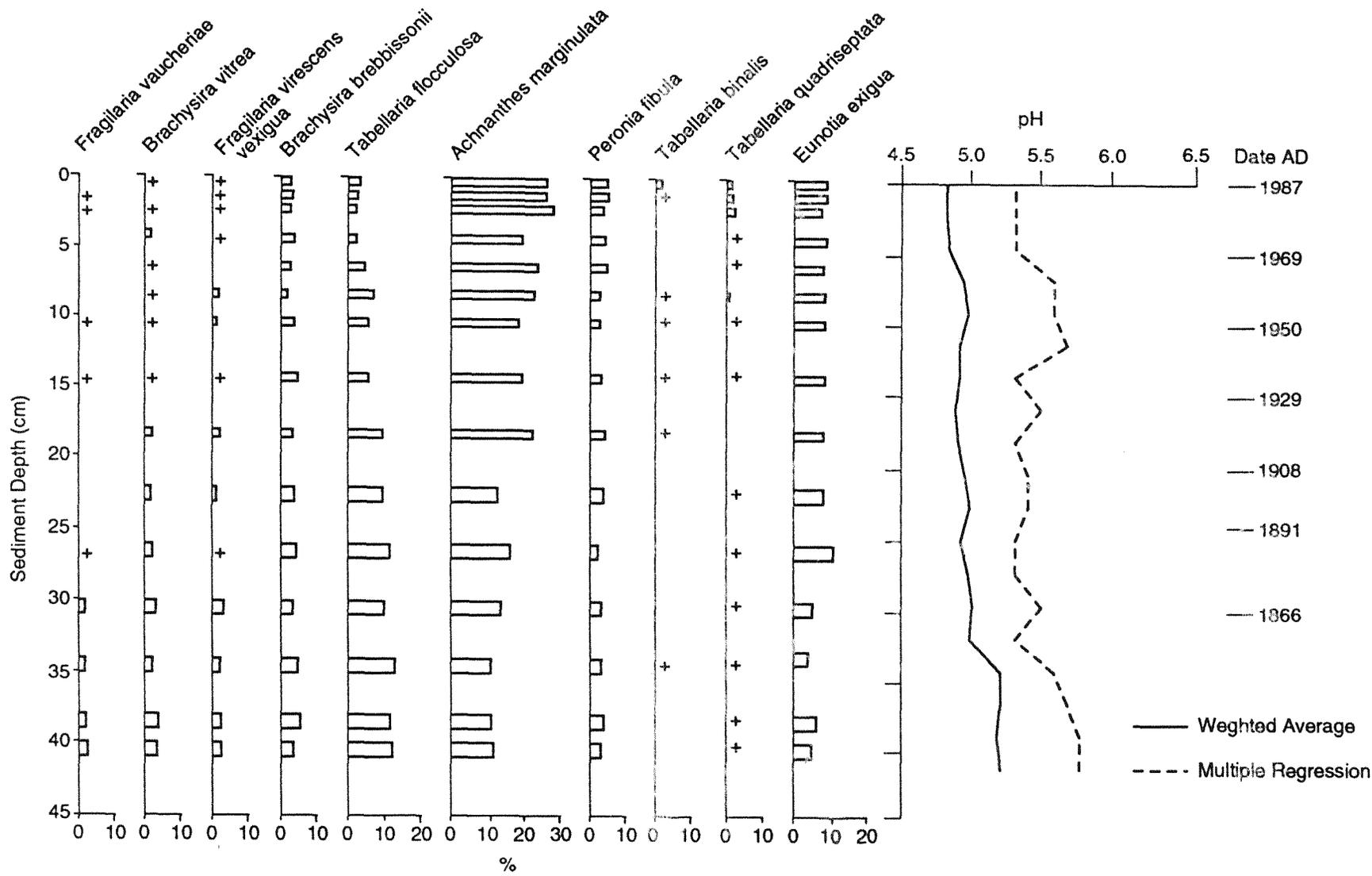


Figure 37 Dubh Loch: diatom concentration ($\times 10^8 \text{ g}^{-1}$), showing 95% confidence limits

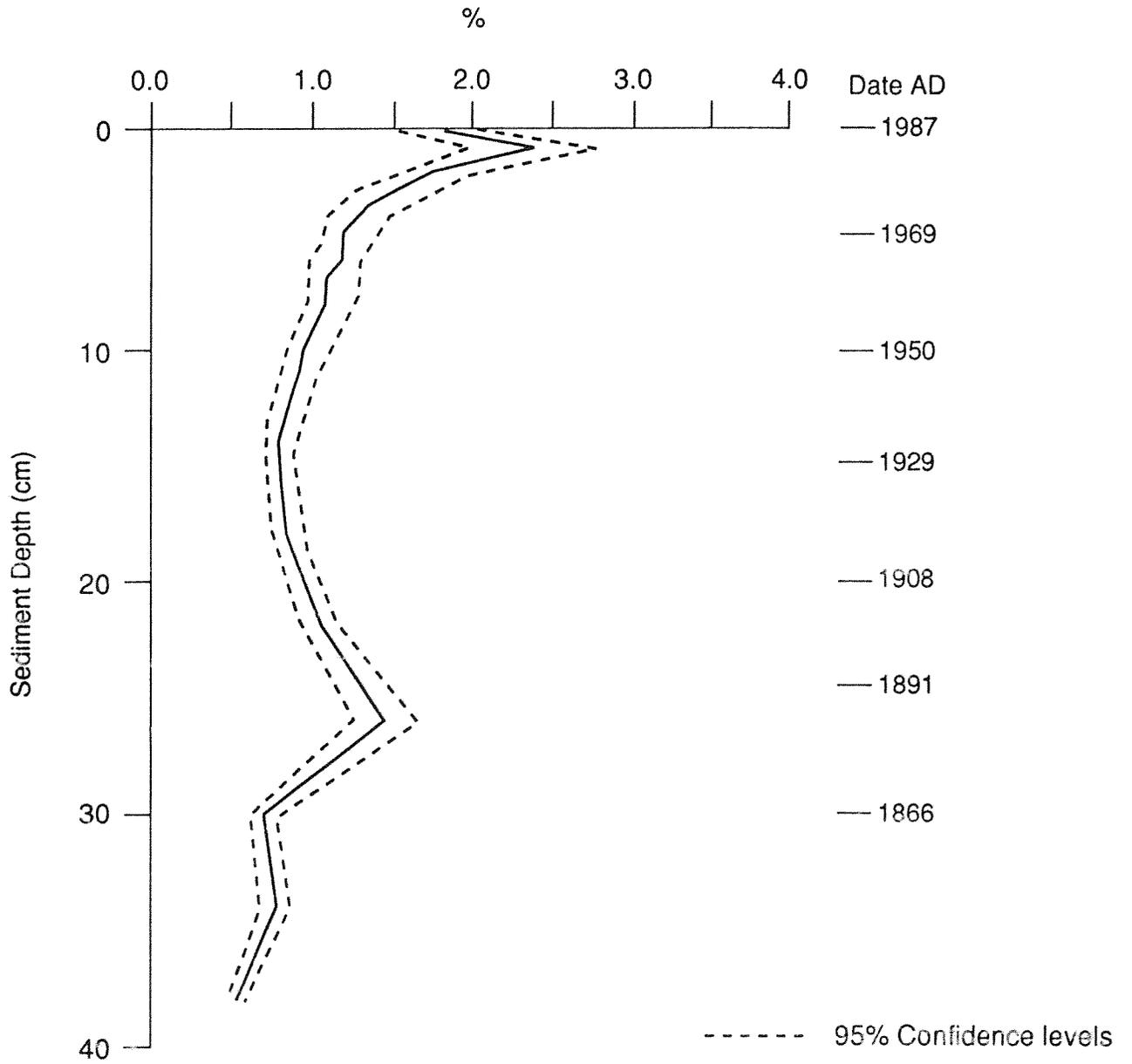


Figure 39 Dubh Loch: carbonaceous particle profile ($\times 10^3 \text{ gDM}^{-1}$)

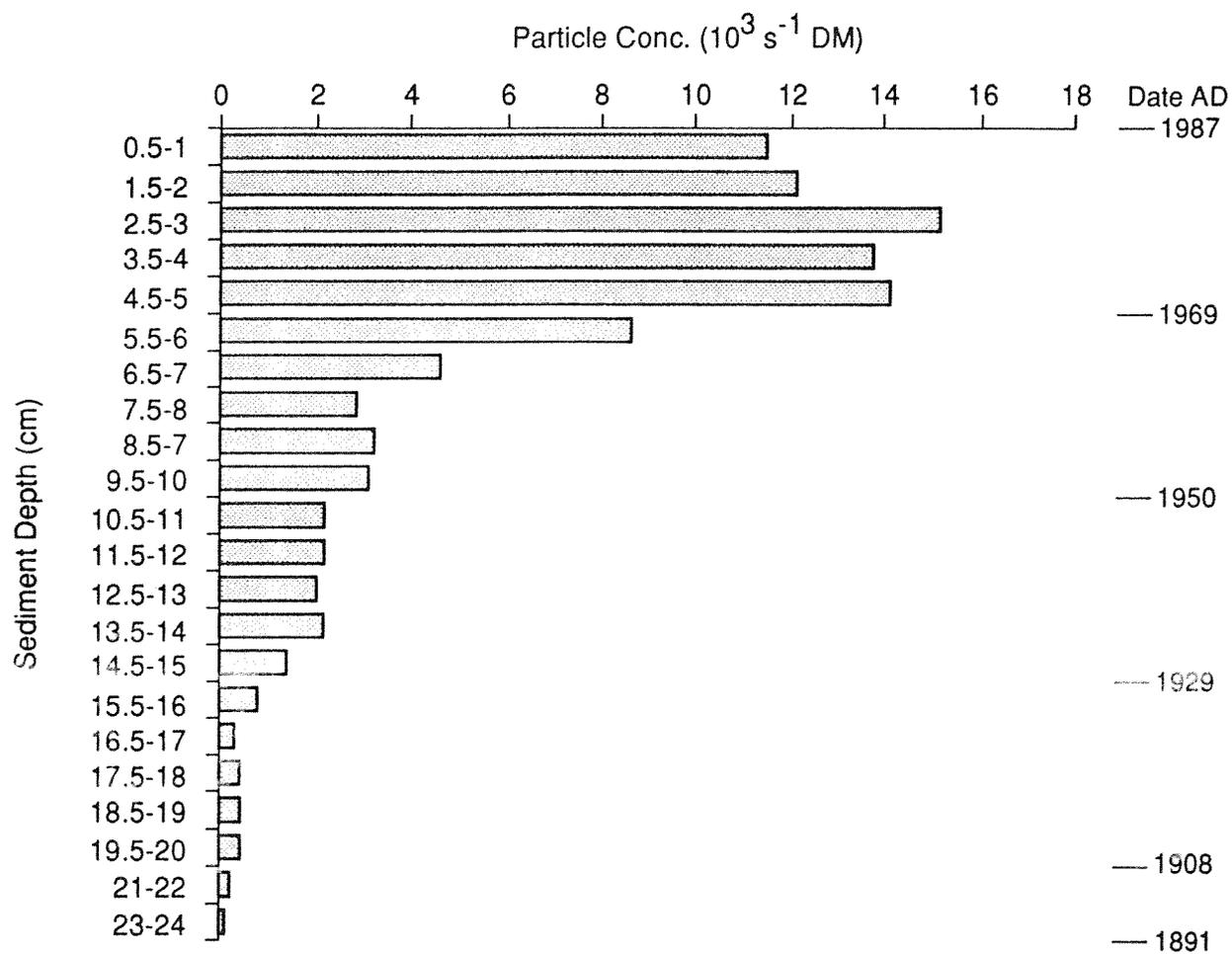


Figure 40 Dubh Loch: sediment magnetic record

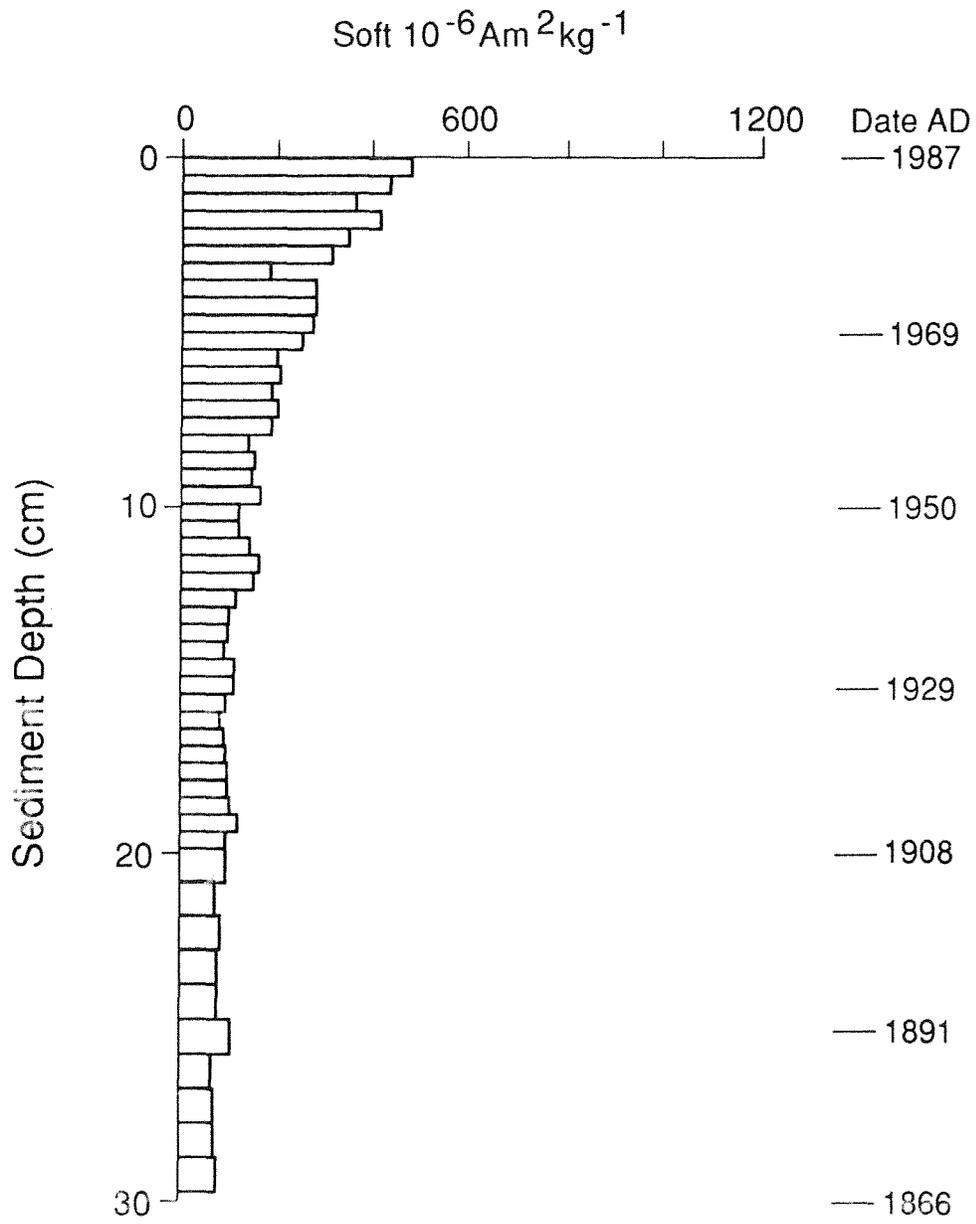


Figure 41 Dubh Loch: the variation of major ion concentrations with depth

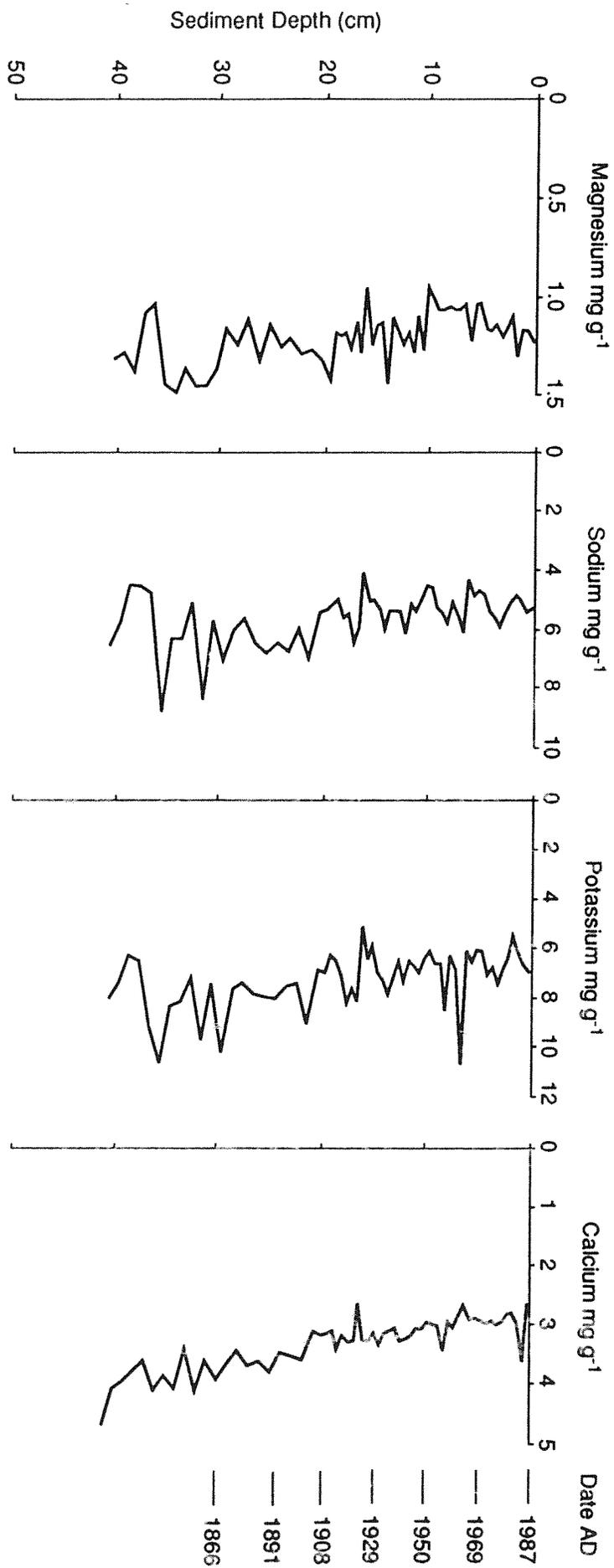


Figure 42 Dubh Loch: the variation of trace metal concentrations with depth

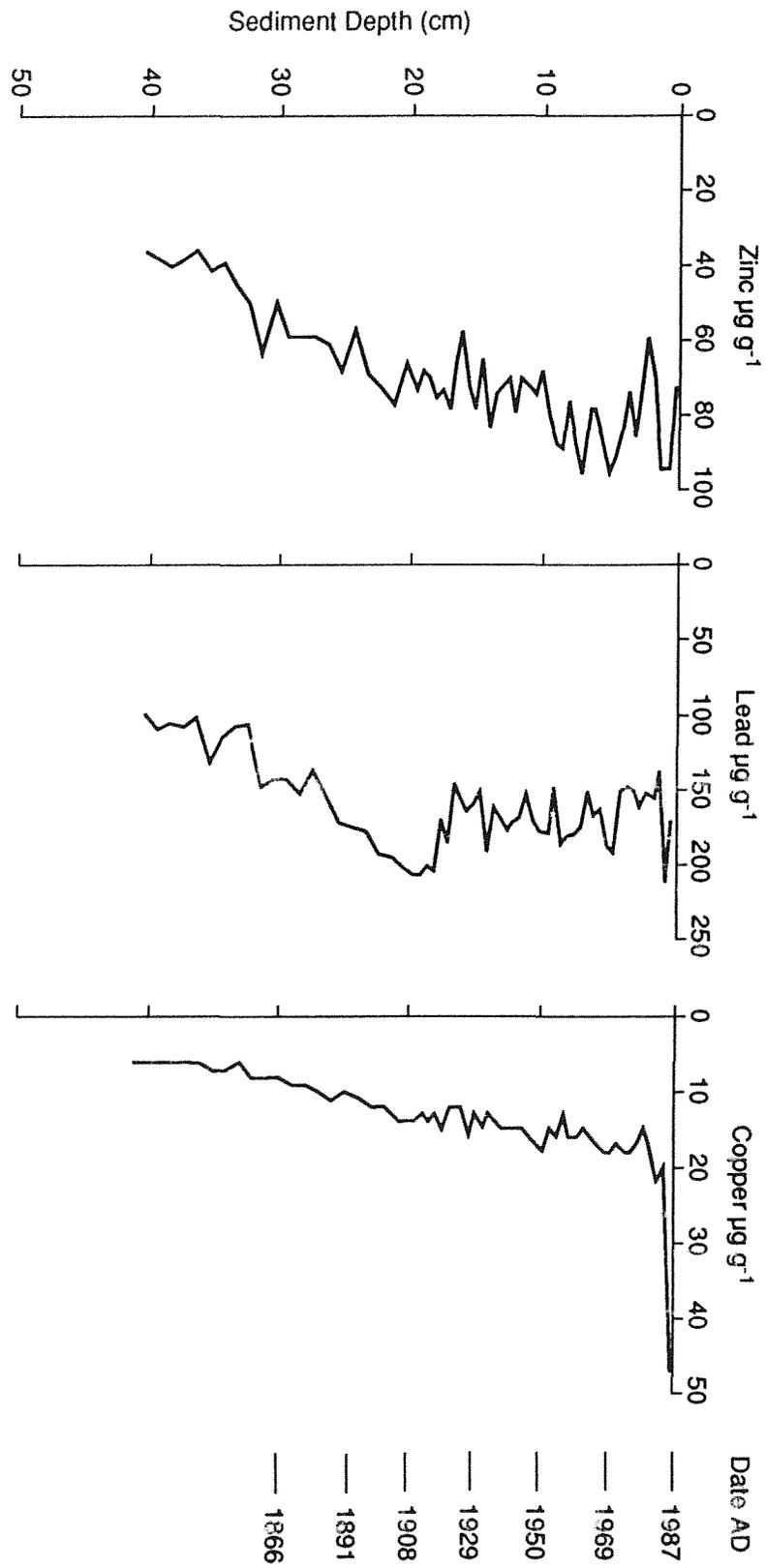
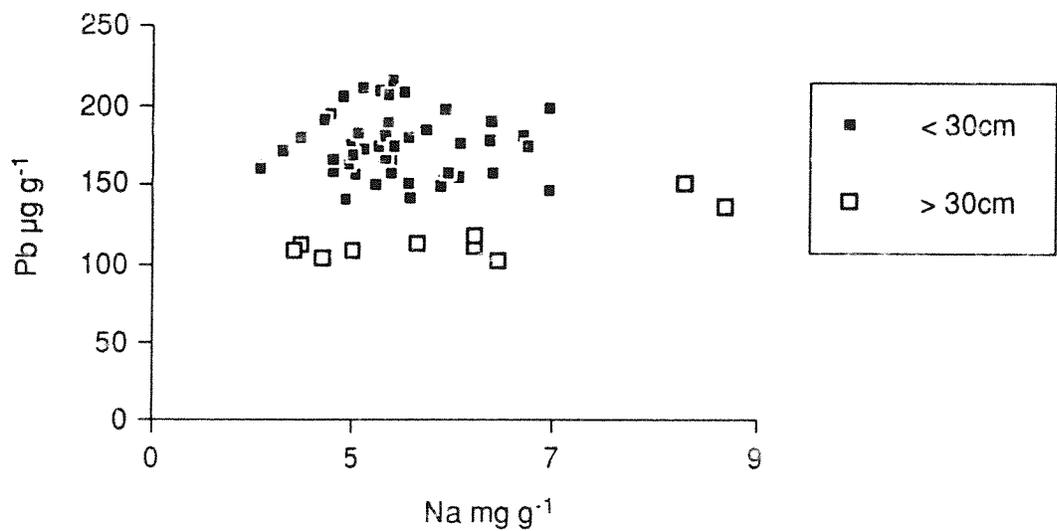
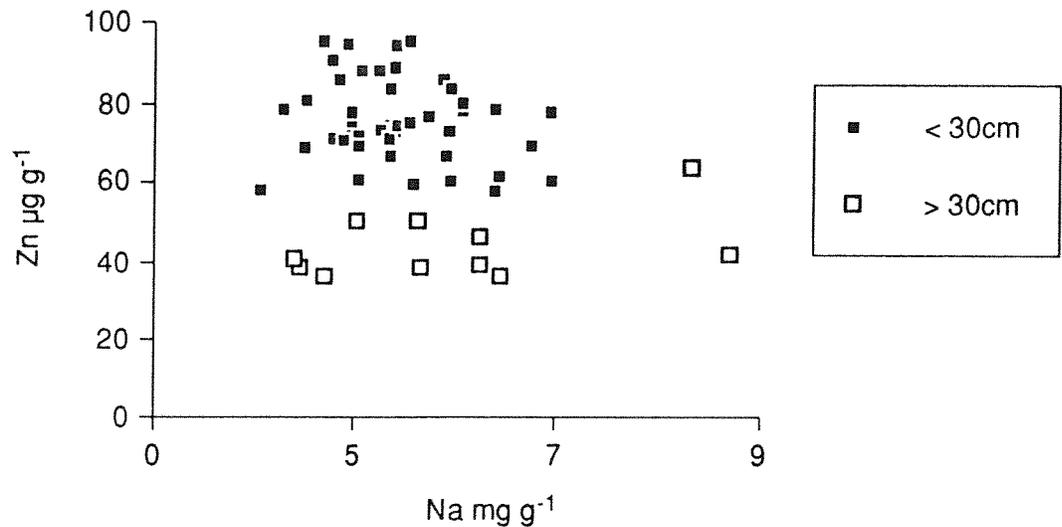


Figure 43 Dubh Loch: the relationships between zinc and sodium and lead and sodium concentrations in the sediments



5 DISCUSSION

The sedimentary record

In any palaeolimnological study the quality of the sedimentary record is important. This becomes crucial in lake acidification studies where the ideal lake sediment core should have a high temporal resolution to enable hypotheses about the causes of lake acidification to be evaluated. The lakes in this study have produced sedimentary records of variable quality, ranging from Dubh Loch where about 30 cm of sediment has accumulated in a linear way over the last 150 years to Loch nan Eun where ^{210}Pb dating is unreliable. At Loch nan Eun unsupported ^{210}Pb is only found in the very top of the core, but it is unlikely that this represents a low sediment accumulation rate since ^{137}CS is also present and the carbonaceous particle evidence suggests that the top 3-4 cm is mid-twentieth century in age. Loch Coire an Lochan has a very low sediment accumulation rate and therefore poor temporal resolution, however there is no evidence of an hiatus in the sedimentary record. At Lochan Uaine a reliable ^{210}Pb chronology was obtained for about the last 120 years, which is represented by the top 9.75cm of sediment. At Lochnagar a conformable, ^{210}Pb dated, sedimentary sequence was found for about the last 90 years, however from 8-11 cm an inwash occurred and an accurate extrapolation of ^{210}Pb dates beyond c. 1880 AD was not possible (Battarbee *et al.* 1988, Patrick *et al.* 1989, Jones *et al.* 1992)

Evidence for lake acidification

The evidence for lake acidification at the four sites plus Lochnagar is summarised in Table 16 and Figure 44. All the sites show signs of acidification, and at three of the sites (Lochnagar, Lochan Uaine and Dubh Loch) this begins in the mid-nineteenth century. At Loch Coire an Lochan a pre-nineteenth century acidification seems to have occurred and there is no evidence of any further acidification in the nineteenth and twentieth centuries. However, the extremely low sediment accumulation rate at this site makes interpretation very difficult. At Loch nan Eun there is a decline of over 0.5 pH units from 1900 to the present day and although the onset of acidification can not be dated accurately, a rough chronology constructed on the basis of the carbonaceous particle profile, by correlating this to other ^{210}Pb dated profiles, gives a late-nineteenth century date for the acidification at this site.

It would appear that lakes in this area of north east Scotland have acidified to a varying extent since about 1850. Differences in the degree of sensitivity to acidification between individual sites can probably be explained in relation to variations in geology, hydrology and soil type between lake catchments.

Causes:

Evidence for atmospheric contamination

The changing concentration of carbonaceous particles through time is shown in Figure 44 which illustrates that all the sites have a very similar pattern of increasing concentrations towards the top of the cores. At Lochnagar, Dubh Loch and Loch Coire an

Lochan small amounts of carbonaceous particles appear from the early to mid-nineteenth century onwards. This period is then followed by a sharp increase in concentration of carbonaceous particles beginning at around 1920 in Lochnagar and Dubh Loch, and rather later in Loch Coire an Lochan (after about 1970). At Lochan Uaine significant quantities of carbonaceous particles are not present until the early twentieth century. At Lochnagar, Dubh Loch and Lochan Uaine peak concentrations are found in the 1970s followed by declining concentrations to the present day. A similar pattern is not found in Loch Coire an Lochan due to the extremely low sediment accumulation rate. At Loch nan Eun evidence for atmospheric contamination is found with increasing concentrations of carbonaceous particles above 7 cm (early/mid-nineteenth century), with a further increase above 2 cm (1956).

Additional evidence for atmospheric contamination emerges from the geochemical analyses of sediment cores from these sites. At Lochan Uaine, Loch nan Eun, Dubh Loch and Lochnagar the geochemical evidence indicates atmospheric contamination from at least the middle of the last century (Jones *et al* 1992). At Loch Coire an Lochan any atmospheric contamination has been at very low levels. The magnetic record from Lochan Uaine and Dubh Loch provides further evidence of atmospheric contamination at these sites.

Evidence for land-use change

It has been argued that changes in land-use or land-management could cause an increase in the production of acid humus producing surface water acidity of a similar magnitude to that from acid deposition (the "land-use" hypothesis Rosenqvist 1977, 1978, 1981). In upland areas this would most commonly be manifested as a change from a grassland community to one dominated by *Calluna*. In the catchments of the highest sites in the Cairngorms (Lochan Uaine and Loch Coire and Lochan) there is currently no *Calluna*, and it is unlikely that *Calluna* ever grew at such altitudes. At these sites the "land-use" hypothesis is irrelevant. At other remote sites such as Lochnagar and Loch nan Eun there is little or no expectation of land-use or land-management changes and this is supported by available documentary evidence (cf. Patrick *et al.* 1989, 1990). The only site in which *Calluna* is currently important is the lowest site, Dubh Loch.

The ratio of *Calluna*:Grass pollen in lake sediments has been used to determine if major changes in land-use or land-management related to grazing and/or burning have occurred (Battarbee *et al.* 1985, Patrick and Stevenson 1990). There is no evidence of any increase in *Calluna* at these sites in the time period where lake acidification has taken place, indeed the evidence at Dubh Loch, the only site with a substantial *Calluna* community, indicates an increase in the amount of grass. The "land-use" hypothesis can be rejected at these sites where land-use and land-management changes likely to cause lake acidification have not occurred.

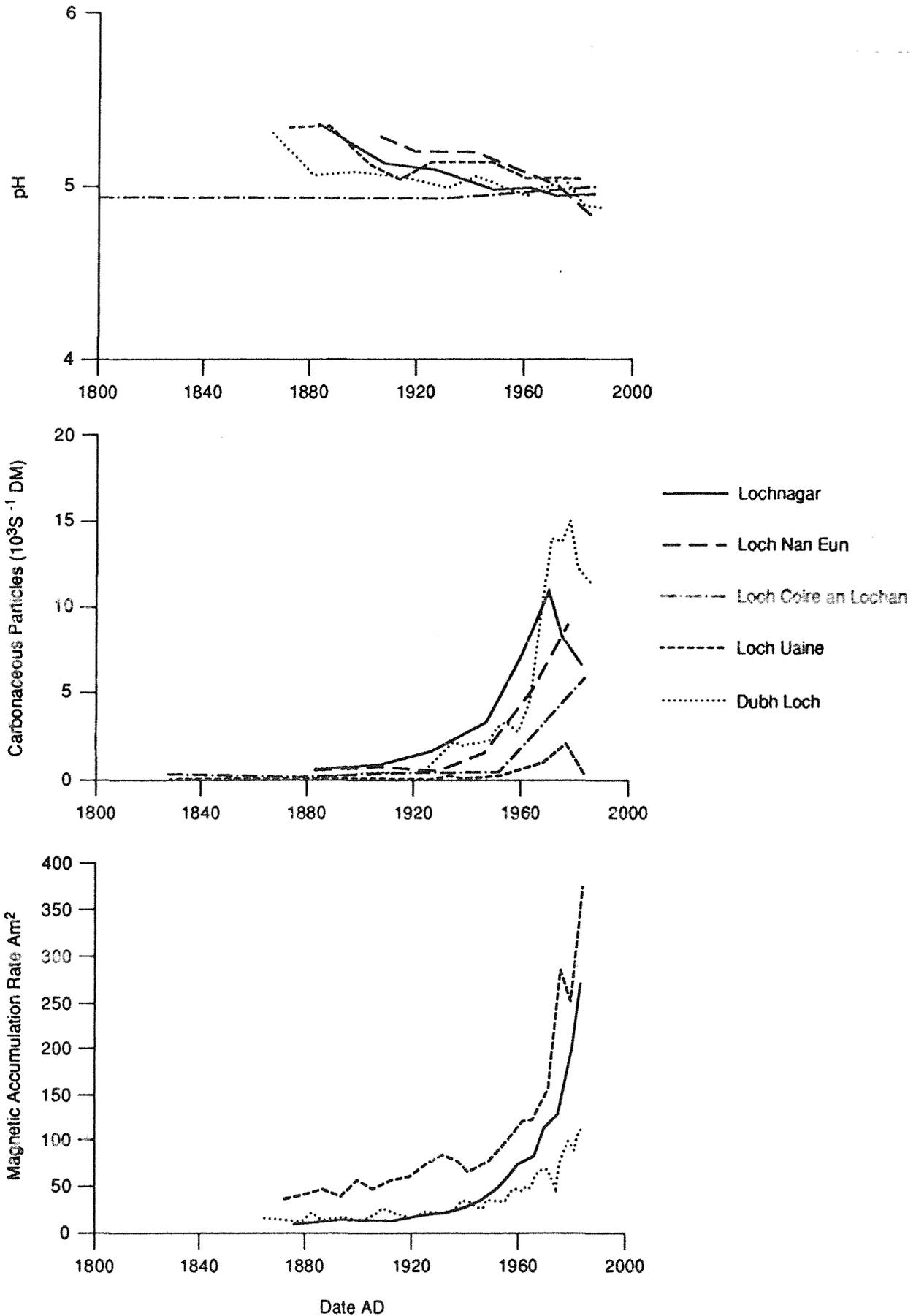
Table 16 Lake acidification in the Cairngorms and Lochnagar: summary derived from the weighted average method of pH reconstruction

	pH before acidification	Date of acidification	pH after acidification	Modern measured pH	Change
Loch Coire an Lochan	5.3	pre-19th C.	4.9	5.6	0.4
Loch nan Eun	6.2	unknown*	4.9	4.9	1.3
Dubh Loch	5.3	c. 1842	4.9	5.3	0.4
Loch Uaine	5.5	c. 1885	5.1	5.8	0.4
Loch-nagar	5.5	mid-19th C.**	5.0	5.0	0.5

* pre-dates reliable ²¹⁰Pb dates

** estimated date from extrapolation of ²¹⁰Pb chronology

Figure 44 Summary diagram



6 CONCLUSION

The most likely cause of the recent acidification of these remote upland lakes is acid deposition. The timing and nature of the changes at these sites are consistent with the acid deposition hypothesis. Although these sites are sensitive to acid deposition they are generally not as strongly acidified as lakes in areas with higher sulphur deposition levels. For example sensitive lakes in the Galloway region of south west Scotland have commonly acidified by about 1 pH unit (Flower *et al.* 1987).

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There is a nominal charge of £4.00 per copy, (titles marked ** £15 per copy owing to reproduction of colour plates).

Acknowledgements

This work was supported by grants from the Department of the Environment (UK) and the Royal Society (Surface Waters Acidification Programme). The authors wish to thank members of the Environmental Change Research Centre, University College London and the Nature Conservancy Council for assistance with fieldwork and Ron Harriman of the SOAFD Laboratory, Pitlochry for water chemistry analyses. We are grateful to the Scottish Wildlife Trust for their assistance and permission to access sites. Diagrams were prepared by Louise Saunders and Tim Aspden of the Cartographic Unit, Department of Geography, University College London.