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INTRODUCTION

A decade ago very little was known about acid lochs in Scotland. The implicit assumption was that they were largely free from pollution. In recent years the position has changed and we now have large bodies of data on the chemistry and biology of upland lochs (Harriman *et al.* 1986, Maitland *et al.* 1987, Battarbee *et al.* 1988) and clear evidence for the recent acidification of many sensitive sites (Flower and Battarbee 1983, Battarbee *et al.* 1985, Flower *et al.* 1987, Battarbee *et al.* 1988).

Information on the acidification (pH) history of lochs has been obtained from the analysis of fossil diatom assemblages extracted from dated sediment cores (Battarbee 1984). Diatom-based pH reconstructions have now been completed for 20 Scottish lochs. This brief paper uses these data to describe the distribution of acidified lochs in Scotland in relation to the pattern of total sulphur (S) deposition and shows how such data can be used to define critical S loads for individual sites.

The distribution of sites studied is shown in Figure 1. Almost all sites are sensitive to acidification with values for calcium (Ca^{++}) less than 100 $\mu eq\ l^{-1}$ (Table 1). Only Loch Urr in Galloway has a higher value (170 $\mu eq\ l^{-1}$). This site was selected as a high pH, high calcium comparison to other, more acidic sites in Galloway. Sites throughout the rest of Scotland were selected to represent most of the sensitive regions of the country as well as to form a series along a gradient of decreasing total S deposition from south to the north-west. Some sites for which results are not yet available, mainly in the north-west of Scotland, are also shown (Figure 1).

REGIONAL PATTERNS

The diatom-based pH reconstruction for each site is shown in Table 2 and the following account considers these data region be region.

1. Galloway

In Galloway the most acidified lakes occur on the Loch Doon and Cairnsmore of Fleet granites. Three sites with moorland catchments situated entirely on the Loch Doon granite: Loch Enoch, Loch Valley and the Round Loch of Glenhead, have become gradually more acid since the mid-nineteenth century. Loch Enoch lost its fish population in the 1950s and fish populations in Loch Valley and the Round Loch of Glenhead are poor. Loch Dee situated partly on the granite and at a lower elevation has been less strongly acidified and acidification began somewhat later, towards the end of the nineteenth century and well before conifer planting in the 1970s.

The response of the three main lakes on the Cairnsmore granite (Loch Grannoch, Loch Fleet, Loch Skerrow) to acid deposition has been complicated by the additional effects of afforestation (including deep ploughing), hydrological disturbance and geological variability. Rapid acidification of Loch Grannoch began in the early twentieth century prior to afforestation, but drainage and conifer planting in the 1960s and 1970s has caused soil erosion and may have exacerbated acidification. At Loch Fleet, unusually high groundwater alkalinity caused by secondary calcite veining in the granite (Cook *et al.* 1987) was probably responsible for this site maintaining a relatively high pH until after afforestation. Afforestation was accompanied by substantial soil inwash to the lake and the rapid post-1970 acidification may have been caused more by the combined effects of inwash and afforestation than by acid deposition. The situation at Loch Skerrow is more complex. Only slight acidification has occurred at this site. The very acid drainage from the afforested granitic parts of the catchment to the west (pH 4.0-4.5) is continually countered by much more alkaline inflows from the east (pH 6.5) where the catchment is underlain by Silurian sedimentary strata. The main high-pH inflow is an artificial leat associated with the water supply to the former Loch Skerrow railway halt.

All the sites on the granite are sensitive to acidification (Ca^{++} 20-70 μeq l^{-1}). As a comparison a core from Loch Urr, a nearby site uninfluenced by granitic geology with Ca^{++} of 166 μeq l^{-1} and pH 6.7, was analysed. Despite a clear record of trace metal and carbonaceous particle contamination, no acidification has occurred.

2. Arran

Surface waters on granitic rocks in the north of Arran are acidified. Cores from Loch Tanna show that the lake was naturally very acid prior to 1800 with pH between 5.0 and 5.5. However, diatom changes from 1850 onwards indicate a progressive acidification to the present day with a decline of about 0.5 pH.

3. Loch Ard forest and the Trossachs

Two sites in this region have been cored to assess the impact of afforestation. Loch Chon and Loch Tinker are situated on the same Upper Dalradian quartzose-schist lithology. The catchment of Loch Chon is extensively afforested whereas Loch Tinker has an open moorland catchment. The analyses show that Loch Tinker has been slightly acidified since 1850 and that Loch Chon has been strongly acidified only after the planting of conifers (post-1970). In this area where catchments have a greater neutralising capacity than the Galloway granites (Loch Tinker and Loch Chon have Ca^{++} of 70-80 μeq l^{-1}), it is apparent that afforestation has helped to promote acidification, probably because of the increased scavenging of dry and occult deposition by the forest canopy.

4. Rannoch Moor

Surface waters on the granite of Rannoch Moor are very sensitive to acidification (Ca^{++} 40 $\mu eq\ l^{-1}$). A core from Loch Laidon, an important fishing loch and part of a National Nature Reserve, shows an acidification of about 0.5 pH from about pH 5.8 to pH 5.3 between 1850 and the present.

5. Cairngorms and Lochnagar

High corrie lochs on the Lochnagar granite (Lochnagar, Dubh Loch and Loch nan Eun) have been acidified by between 0.5 and 1 pH unit from about 1850 to the present day. All sites are extremely remote and at high elevation (>500 m) yet are strongly contaminated by trace metals and carbonaceous particles. Lochan Uaine in the main Cairngorm region is similarly contaminated but is less sensitive to acidification (Ca^{++} 60 $\mu eq\ l^{-1}$) and no pH decrease has occurred.

6. Strontian and Morvern

Data are available for Lochan Dubh and Loch Doilet in this region of relatively low acid deposition. Lochan Dubh has a moorland catchment whilst the catchment of Loch Doilet has been extensively afforested. Both are very sensitive sites and have been acidified by about 0.3 pH unit. The apparent lack of a "forest effect" at Loch Doilet, compared to the effect at Loch Chon (Table 2), suggests that conifer growth by itself does not cause acidification. Loch Uisge, a site somewhat to the south of these sites is less sensitive (Ca^{++} 66 $\mu eq\ l^{-1}$) and no pH change has occurred.

7. The North and North-west

The North and North-west highlands and islands of Scotland are areas of relatively low acid deposition. Work is in progress on sediments from a number of sites. No clear-water (TOC <5 mg l^{-1}) lakes with pH <5.5 are known in this region yet many are sensitive to acidification with Ca^{++} <50 $\mu eq\ l^{-1}$). So far only data from Loch Coire nan Arr, a clear water loch with pH of 6.3 are available. Despite its sensitivity (similar to the Round Loch of Glenhead), no pH change has occurred at this site, and carbonaceous particle concentrations are very low.

COMPARISONS WITH TOTAL S DEPOSITION AND THE DERIVATION OF CRITICAL LOADS

Figure 2 shows the pattern of pH change of lochs in Scotland compared with contours of total S deposition. The sites for which no pH change has occurred are indicated by open circles and include Loch Coire nan Arr, Loch Uisge, Lochan Uaine, and Loch Urr. The location of these sites in relation to the pattern of S deposition illustrates the clear relationship between sensitivity and deposition. Only in the north-west of Scotland, where total S deposition is approximately 0.5 g S m^{-2} yr^{-1} or less, are very sensitive sites (Ca^{++} <50 µeq l^{-1}) not acidified. In Galloway, where the highest total S deposition occurs, sensitive sites are strongly acidified and only sites with Ca^{++} >100 µeq l^{-1} are not acidified. The only significant qualification to this pattern is in the case of afforested catchments where sites such as Loch Chon are more strongly acidified than would be predicted from this relationship.

In Figure 3 total S deposition at each site is plotted against calcium, as an indicator of sensitivity and the diatom-based acidification status of each site is shown. Although there are relatively few data points for non-acidified lochs it is possible to indicate an approximate calcium to deposited sulphur ratio which differentiates acidified from non-acidified waters. This line or ratio then allows the critical S load for any site to be calculated. For example the critical load for Loch Enoch, with a Ca^{++} value of about 20 µeq l^{-1}, is approximately 0.3 g S m^{-2} y^{-1}. To return this site to a non-acidified status, assuming base cation values have and will remain approximately constant as deposition is reduced (cf. Henriksen 1984), requires a reduction in S deposition from about 1.25 to 0.3 g m^{-2} y^{-1}, or approximately 75%. On the other hand for Loch Tinker, a less acidified, less sensitive site although in an area of somewhat higher S deposition (1.4 g m^{-2} y^{-1}) only a very slight reduction should be sufficient for a return to its non-acidified status. If base cation concentrations decrease during de-acidification then critical loads will be proportionally lower and greater percentage reductions will be required.

Afforested sites cannot be treated in this way since the actual S deposition at these sites is probably 50-100% greater (Fowler, pers. comm.) than calculated from the regional Warren Springs data (Barret *et al.* 1987) used in this Figure.

CONCLUSIONS

On the basis of these observations and from other published and unpublished results a number of conclusions can be made about the acidification of Scottish lochs. At sites with non-afforested or moorland catchments it is now clear that:

a. with the exception of some very brown waters in the north almost all lakes with present pH <5.5 have been recently acidified, some slightly, some severely;

b. acid deposition is the main cause of acidification;

c. non-acidified, but sensitive clear-water sites still occur in the far north of Scotland;

d. the boundary between acidified and non-acidified systems for the most sensitive sites is approximately along a line somewhat to the north of the Caledonian fault;

e. acidification throughout the rest of Scotland depends on sensitivity, or acid neutralising capacity, and lochs with $Ca^{++} > 100$ $\mu eq\ l^{-1}$ have not been acidified;

f. The worst affected areas are parts of Galloway, north Arran, Rannoch Moor and the Cairngorms, especially Lochnagar;

g. critical sulphur loads depend on lake sensitivity and can be defined from diatom-inferred acidification histories of a range of sites.

For afforested sites less data are available. So far, however, it is possible to conclude that:

a. at very sensitive sites acidification usually predates afforestation;

b. at less sensitive sites (50-100 $\mu eq\ Ca^{++}\ l^{-1}$) in areas of high S deposition afforestation exacerbates acidification.

c. at sensitive sites in areas of low S deposition afforestation does not appear to cause acidification or to exacerbate it.

d. the afforestation effect must therefore be related to S deposition (through an enhanced interception mechanism) rather than to a direct effect of forest growth.

FURTHER QUESTIONS

Although the main cause of acidified waters in Scotland is now well understood and work in progress will establish the broad extent of acidification, a number of issues remain. Relatively few lochs with afforested catchments have been studied and there is a need to investigate the forest effect at sites with intermediate sensitivity where afforestation might cause local S deposition to exceed the critical load.

In addition, S deposition in Scotland is now decreasing and is likely to decrease further as Flue Gas Desulphurisation systems are installed in the 1990s. The extent to which lochs will respond to such reductions, both chemically and biologically is not yet known. Some lochs in Galloway already show minor signs of improvement (Battarbee *et al.* 1988). Close examination of recent sediments (1970-1990) at other lochs throughout Scotland is needed to determine present trends and to evaluate the impact of S reduction strategies in the future.

Finally, the extent of improvement in the future can only be measured against the yardstick of non-acidified control sites. There is an urgent need to identify and study a range of such "pristine" sites which can still be found in north-west Scotland (eg. Loch Coire nan Arr) and in other countries (Muniz 1987). The suitability of sites can be assessed by palaeoecological means and chemical and biological survey techniques can be used to define targets for ecosystem restoration.

TABLE AND FIGURE CAPTIONS

Table 1: Mean water chemistry of sites for which pH reconstruction is available (see Table 2).

Table 2: Diatom-inferred pH change for 20 Scottish lochs. Inference model based on the multiple regression of pH preference groups with measured pH for a Galloway calibration set.

Figure 1: Distribution of core sites in relation to the map of susceptibility of surface and groundwaters to acid deposition (from Edmunds and Kinniburgh 1986).

Figure 2: pH change at core sites in relation to the modelled deposition of S (g m^{-2} yr^{-1}) for Scotland (from Derwent *et al.* 1988).

Figure 3: The relationship between calcium concentration in lake water, as an index of sensitivity to acidification and total S deposition for core sites in the United Kingdom. The acidification status of each lake is shown and the data can be used to derive the critical S load for each site. Sites are indicated in Table 2 and Battarbee *et al.* 1988.

Table 1 Scottish sites — mean water chemistry

Site	Mean pH	Conductivity$_{25°C}$ µS cm^{-1}	Ca^{2+} µeq l^{-1}	Mg^{2+} µeq l^{-1}	K^{+} µeq l^{-1}	Na^{+} µeq l^{-1}	Cl^{-} µeq l^{-1}	SO$_4^{2-}$ µeq l^{-1}	Alkalinity µeq l^{-1}	TOC mg l^{-1}	Labile Al µg l^{-1}
1. L. Enoch	4.5	37.3	18.5	37.4	6.5	167.2	197.5	37.5	0.00	–	–
2. L. Valley	4.7	37.3	25.0	44.4	7.2	171.4	268.0	85.4	0.00	–	–
3. Round Loch of Glenhead	4.7	41.0	29.8	45.2	6.1	177.9	265.2	80.2	0.00	–	–
4. L. Dee	5.3	41.0	66.5	58.0	16.1	191.0	228.5	82.2	34.90	–	–
5. L. Grannoch	4.7	50.0	49.0	53.1	5.2	221.8	287.7	115.6	0.00	–	–
6. L. Fleet	4.5	54.0	80.5	71.6	15.6	294.5	282.1	110.3	43.45	–	–
7. L. Skerrow	5.3	58.0	107.5	80.6	6.9	263.6	355.4	133.3	21.72	–	–
8. L. Urr	6.8	63.3	171.5	137.8	11.0	205.3	287.7	153.0	115.68	–	–
9. L. Tanna	5.0	47.0	36.7	67.6	12.1	238.6	218.3	91.0	0.00	1.8	87
10. L. Tinker	5.7	29.2	79.2	38.8	8.0	121.0	141.8	64.2	18.12	3.6	5
11. L. Chon	5.2	39.0	79.4	49.4	7.2	175.2	188.4	85.4	2.32	2.2	30
12. L. Laidon	5.4	24.8	41.3	31.0	6.3	132.0	130.0	45.5	12.81	3.1	3
13. Lochnagar	5.0	21.4	30.2	33.7	7.5	89.0	76.2	66.0	0.00	0.8	57
14. Dubh Loch	5.3	17.3	27.3	21.3	5.3	72.3	67.7	54.0	1.37	1.6	55
15. L. nan Eun	5.0	22.3	29.7	26.3	5.7	82.3	55.0	62.3	0.00	0.5	128
16. Lochan Uaine	5.8	40.5	69.0	59.5	10.0	191.0	215.0	82.0	14.79	0.4	56
17. Lochan Dubh	5.6	29.0	33.2	40.2	8.0	163.0	185.4	109.6	9.75	2.8	11
18. L. Doilet	5.6	43.8	54.0	60.5	12.5	238.7	277.0	70.6	15.43	2.4	7
19. L. Uisge	6.2	38.0	66.5	56.8	15.7	213.3	220.0	50.0	33.10	4.7	–
20. L. Coire nan Arr	6.2	39.2	43.3	59.2	13.3	216.2	263.0	45.8	27.90	2.2	2

Table 2 Scottish sites — pH change

	Grid reference	Modern pH (measured)	Date of core	pH 1800 (inferred)	Modern pH (inferred)	First point of decrease	pH change
1. L. Enoch	NX 446851	4.5	1982	5.3	4.4	1840	0.9
2. L. Valley	NX 444817	4.7	1981	5.6	4.6	1860	1.0
3. Round Loch of Glenhead	NX 450804	4.7	1981	5.9	4.9	1860	1.0
4. L. Dee	NX 466788	5.3	1980	6.2	5.5	1890	0.7
5. L. Grannoch	NX 542700	4.7	1980	5.7	4.7	1930	1.0
6. L. Fleet	NX 560698	4.5	1985	5.8	4.6	1975	1.2
7. L. Skerrow	NX 605682	5.3	1980	5.9	5.8	–[1]	–[2]
8. L. Urr	NX 760845	6.8	1984	6.5	6.8	–[1]	–[1]
9. Loch Tanna	NR 921428	5.0	1986	5.0	4.6	1850	0.4
10. L. Tinker	NN 445068	5.7	1985	6.4	6.0	1850	0.4
11. L. Chon	NN 421051	5.2	1986	6.4	5.5	1900[3]	0.9
12. Loch Laidon	NN 380542	5.4	1985	5.8	5.3	1860	0.5
13. Lochnagar	NO 252859	5.0	1986	5.7	4.8	1850	0.9
14. Dubh Loch	NO 238828	5.3	1986	5.7[4]	5.2	–[5]	0.5
15. Loch nan Eun	NO 230854	5.0	1986	6.0[4]	4.8	–[5]	1.2
16. Lochan Uaine	NO 001981	5.8	1986	5.8	5.7	–[1]	–[2]
17. Lochan Dubh	NM 895710	5.6	1986	5.5	5.2	–[5]	0.3
18. L. Doilet	NM 808678	5.6	1986	6.0[4]	5.7	–[5]	0.3
19. L. Uisge	NM 808550	6.2	1986	6.2[4]	6.1	–[1]	–[2]
20. L. Coire nan Arr	NG 808422	6.2	1986	6.2[4]	6.4	–[1]	–[1]

NOTES: [1] Not applicable. [2] Not significant. [3] Main change after afforestation (post-1970).
[4] Refers to the base of the core. [5] Core not yet dated.

Figure 1

The susceptibility of surface and groundwaters to acid deposition

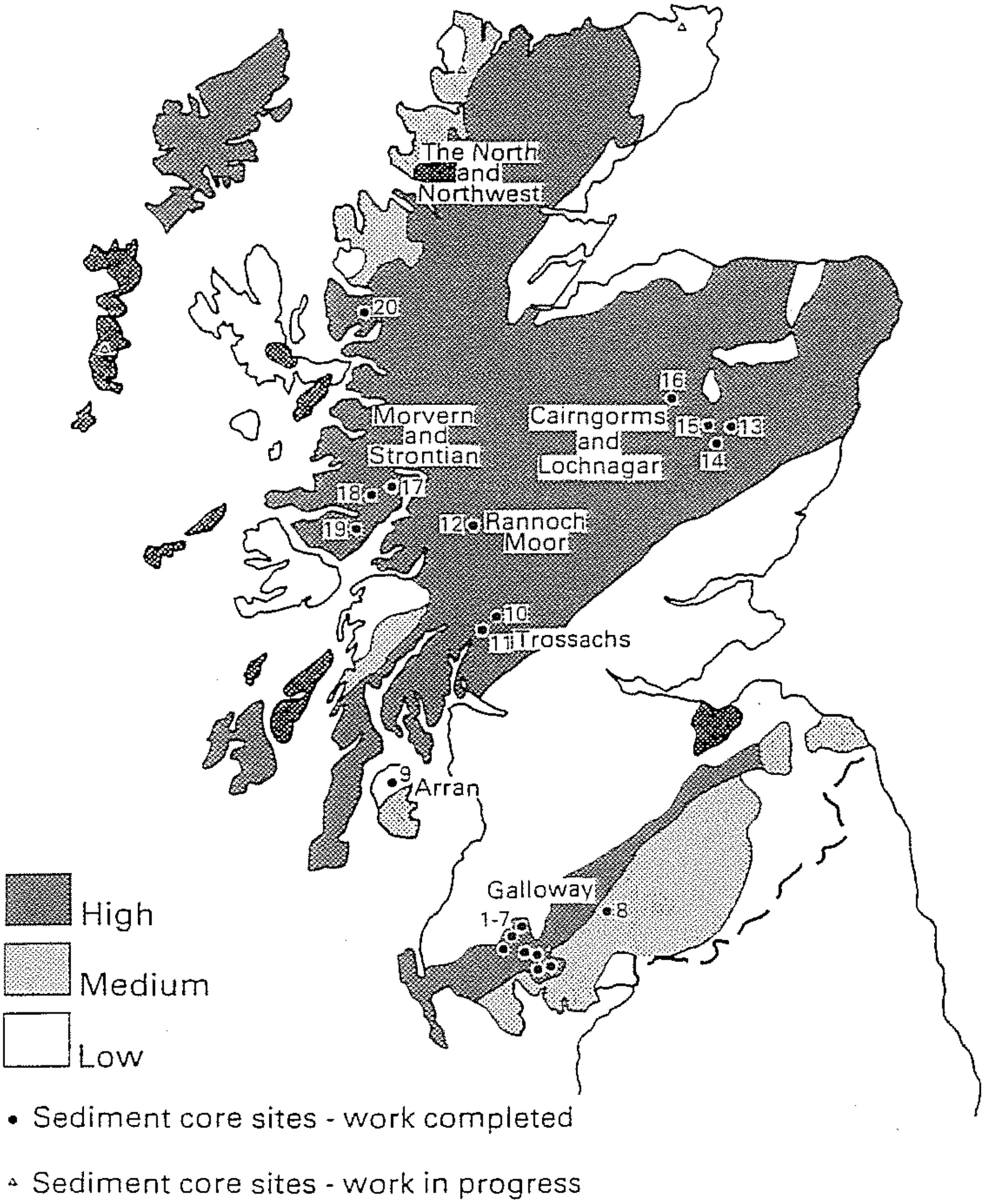
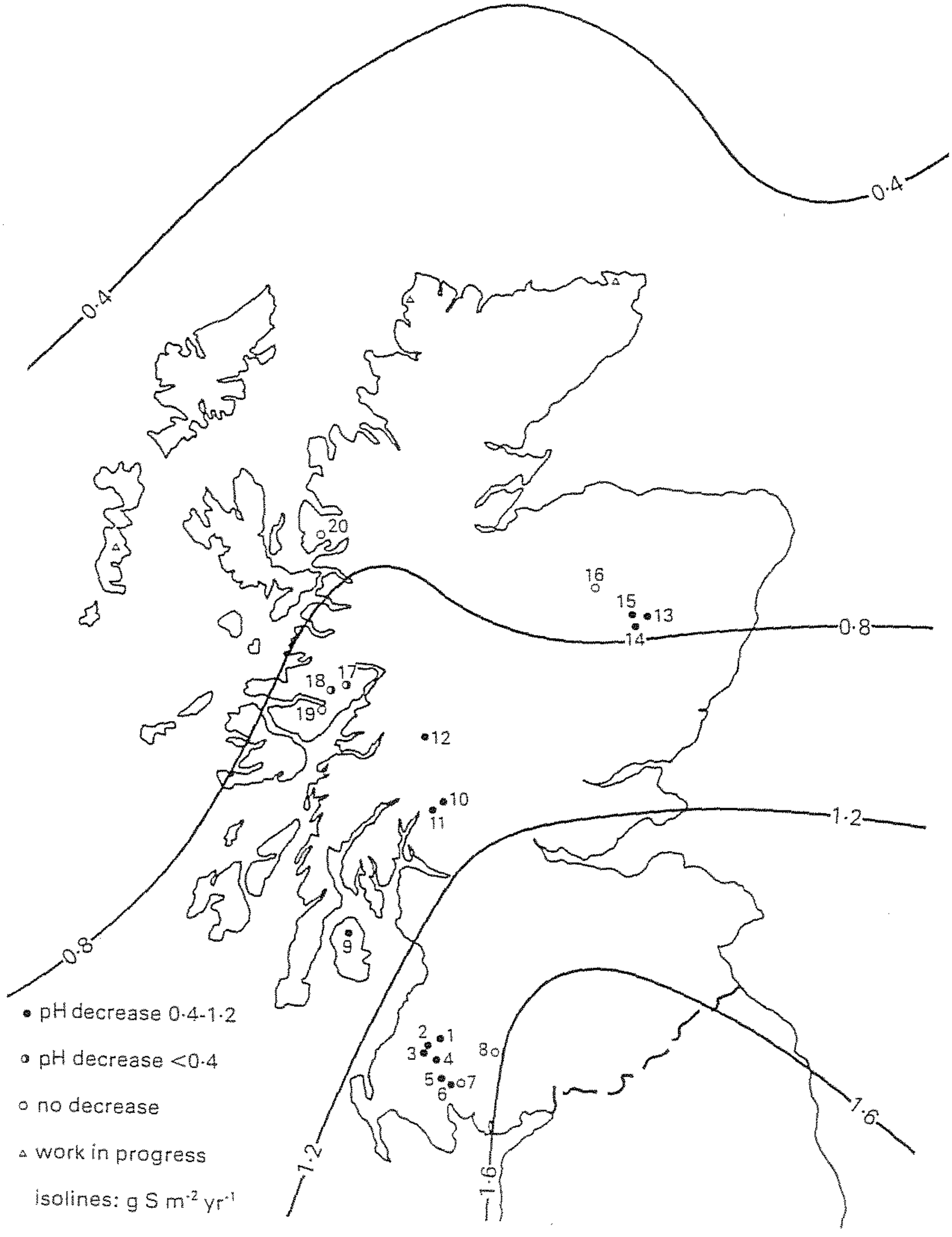


Figure 2


0·4
0·4
0·8
0·8
1·2
1·2
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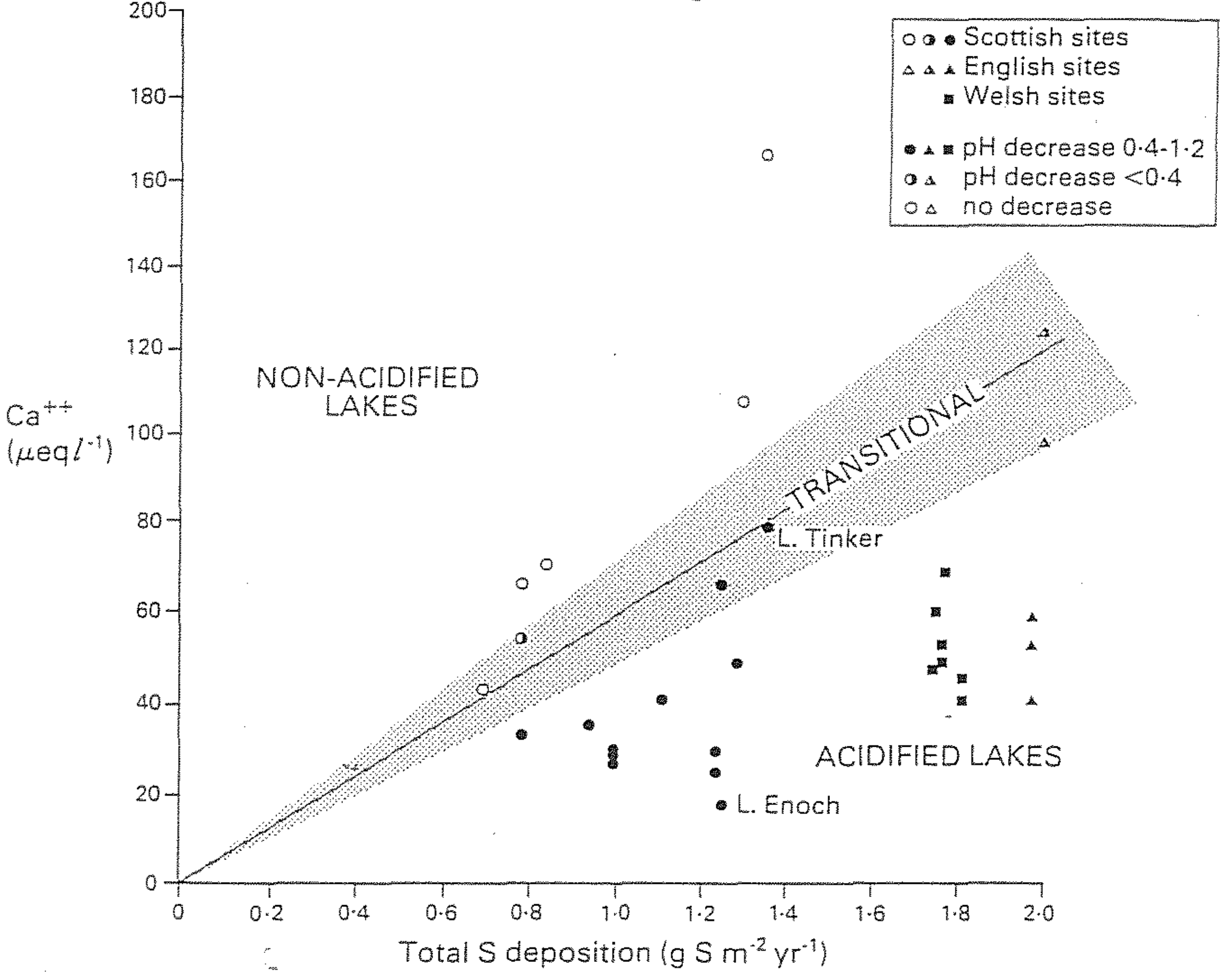
18
19
20
● pH decrease 0·4-1·2
◦ pH decrease <0·4
○ no decrease
△ work in progress
isolines: g S m⁻² yr⁻¹

Figure 3

Critical Sulphur loads for U.K. based on Palaeolimnological Data



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