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Alkalinity and pH of three lakes in northern New England, U.S.A., over the past 300 years

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Three-hundred-year histories of pH and total alkalinity (alk) have been inferred from diatom and chrysophyte remains in deep-water sediment cores from Mud Pond (pH 4.6, alk -23 μeq l^{-1}) and Little Long Pond (pH 5.7, alk 4 μeq l^{-1}), Maine and Haystack Pond (pH 4.8, alk -18 μeq l^{-1}), Vermont. Three replicate cores were studied from each Mud Pond and Haystack Pond; one core from Little Long Pond; pH and alk inferences from diatoms were based on three different calibration equations: CLUSTER, DECORANA and CCA (CANOCO) (only CCA for chrysophytes). Replication of pH and alk inferences between cores was excellent. Different calibration approaches led to the same conclusions with minor exceptions. There were minor differences between chrysophyte- and diatom-based inferences, but both led to similar conclusions regarding acidification. These were: Mud Pond, *ca.* 1700–1925, pH 5.2–5.3, alk 0 to -15 μeq l^{-1}; 1925–1970, acidification to pH $\simeq$ 4.8 and alk -20 to -30 μeq l^{-1}. Little Long Pond *ca.* 1700–1950, pH $\simeq$ 5.9, alk 20–50 μeq l^{-1}; 1950 ff., possible slight acidification to pH 5.7–5.8. Haystack Pond, *ca.* 1700–1925, pH 5.2–5.3, alk 0 to -10 μeq l^{-1}; 1925–1970, acidification to pH $\simeq$ 4.9 and alk -10 to -30 μeq l^{-1}. Correlation of lake acidification with great increases in sedimentary indicators of air pollution (carbonaceous particles, Pb, polycyclic aromatic hydrocarbons) and absence of correlated catchment disturbance point to anthropogenic acid deposition as the cause of lake acidification. Extreme acid sensitive lakes like these three are atypical for northern New England.

Introduction

The palaeolimnological approach has proven useful for understanding the effects of anthropogenic acid deposition on lakes. The approach has relied heavily on inferences of past pH and total alkalinity (alk) from the remains of diatoms and chrysophytes in sediment cores. This area of palaeolimnology, or major parts of it, have been reviewed by Battarbee *et al.* (1986), Charles & Norton (1986), Davis (1987) and Charles *et al.* (1989). Summary results from northern New England (Vermont, New Hampshire and Maine) were given by Davis *et al.* (1983), Norton *et al.* (1985) and Davis (1987), but much of the information on the study lakes in the region has not been published. This paper aims to partially rectify that situation by giving more complete results for three lakes (figure 1). Additional results are given by Davis *et al.* (1990). This work was part of the Paleolimnological Investigation of Recent Lake Acidification Project (PIRLA) (Charles & Whitehead 1986*a*).

The specific objectives of this project were: (i) methodological (*a*) compare reconstructions of pH and alk from replicate cores from the same lake (can conclusions regarding lake acidification be based on a single core?), (*b*) compare pH and alk reconstructions based on different calibration equations (does the particular calibration equation affect conclusions on

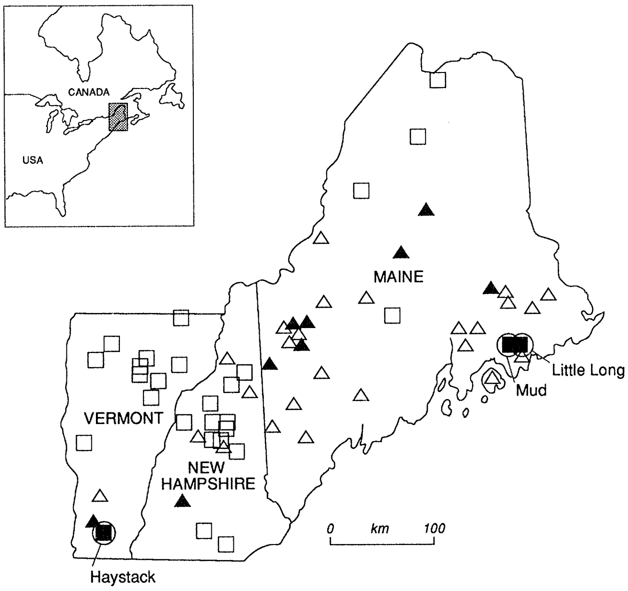

FIGURE 1. Locations of Haystock Pond (1), Mud Pond(2) and Little Long Pond (3), and the additional 60 lakes used for pH and alkalinity calibrations. Squares indicate sites studied as part of the PIRLA Project; triangles as part of earlier projects. Solid symbols indicate lakes where cores were subjected to diatom and chemical stratigraphic analyses, and where pH and alk reconstructions have been made (Davis *et al.* 1990).

lake acidification?) and (*c*) compare pH and alk reconstructions based on diatom against chrysophyte remains from the same core (do the different remains lead to different conclusions regarding lake acidification?). (ii) Interpret pH and alk reconstructions in terms of (*a*) pre-pollution pH and alk conditions of the lakes, (*b*) any changes in conditions during the period of anthropogenic acid deposition, and the timing of such changes and (*c*) correlations between changes and the sedimentary record of air pollutants associated with fossil fuel combustion and acid deposition (carbonaceous particles (CarbP); Pb; polycyclic aromatic hydrocarbons (PAH)).

STUDY LAKES

The study lakes are Haystack Pond in southern Vermont and Little Long Pond and Mud Pond in eastern Maine (figure 1). Water from Mud Pond flows into Little Long Pond via a 200 m long stream. The three water bodies have heavily forested catchments that have been undisturbed since selective logging around 1900. The thin, coarse-grained soils are derived from granite. Haystack Pond and Mud Pond had (1983 and 1984) negative alk and, respectively, mean pH values of 4.8 and 4.6. Little Long Pond had mean alk of about 4 μeq l^{-1} and mean pH 5.7. The lakes are oligotrophic and low in dissolved organic matter (table 1).

SUMMARY OF METHODS

Detailed field and laboratory methods for all aspects of the PIRLA Project were given by Charles & Whitehead (1986*b*). Undisturbed cores of sediment approx. 2 m apart were obtained from the deepest part of each lake by using a 10 cm diameter improved version of a

TABLE 1. CHARACTERISTICS OF THE THREE STUDY LAKES

(Each value is a mean of depth-integrated samples taken on six to eight dates from the mid-lake mixed stratum in 1983 and 1984 ice-free seasons. See Charles & Whitehead (1986*b*) and Davis *et al.* (1990) for methods.)

	Haystack Pond	Mud Pond	Little Long Pond
catchment			
area/ha[a]	52	50	205
total relief/m	132	246	285
mean slope (%)	36	12	10
lake			
surface elevation a.s.l./m	910	110	72
surface area/ha[a]	11	1	22
maximum depth/m	11.6	15.0	24.5
transparency/m	9.5	7.3	10.7
conductance/($\mu S\ cm^{-1}$)	19	31	24
dissolved organic carbon/($mg\ l^{-1}$)	1.5	3.7	1.0
chlorophyll *a*/($\mu g\ l^{-1}$)	0.2	0.5	0.7
total phosphorus/($\mu g\ l^{-1}$)	4.7	4.2	2.4
silicon/($mg\ l^{-1}$)	0.3	1.5	1.4
total aluminium/($\mu g\ l^{-1}$)	129.8	330.3	40.9
labile aluminium/($\mu g\ l^{-1}$)	85.5	219.7	31.0
calcium/($\mu eq\ l^{-1}$)	41.0	23.4	43.9
nitrate/($\mu eq\ l^{-1}$)	5.7	0.7	1.1
non-marine sulphate/($\mu eq\ l^{-1}$)	72.3	89.6	70.6
total alkalinity/($\mu eq\ l^{-1}$)	−18.3	−23.0	3.7
pH	4.83	4.57	5.70

[a] 1 ha = $10^4\ m^2$.

corer described by Davis & Doyle (1969). Three cores were studied from Haystack Pond, three from Mud Pond and one from Little Long Pond.

Cores were analysed for a wide range of chemical variables (Davis *et al.* 1990) but only Pb and PAH are reported here. Diatoms and chrysophyte scales were determined microscopically on the same slides, from a series of depths in each core. Diatom taxonomy in the PIRLA Project was standardized (Camburn *et al.* 1984–1986). ^{210}Pb dating was supplemented by dates based on pollen chronostratigraphic markers (Davis *et al.* 1990; Charles & Whitehead 1986*b*). Carbonaceous particles, determined on pollen slides, consisted of coal and oil combustion spherules as well as other soot particles, but as much as possible did not include carbonized remains of plants (e.g., charcoal). A collection of slides and photomicrographs of emission particles from fossil-fuel combustion facilities and carbonized plant parts including crushed charcoal of several tree species was used for reference.

A data-set for pH and alk calibration of sedimentary remains of diatoms and chrysophytes, comprising 63 lakes in Vermont, New Hampshire and Maine (figure 1) was established (partially reported by Davis & Anderson (1985)). Three different calibration equations were developed for diatom-inferred pH (DIpH) and three for diatom-inferred alk (DIalk). These equations consisted of regressions of pH and alk of: (i) groups of diatoms as determined by a cluster analysis; (ii) diatom scores on the first DECORANA axis; and (iii) diatom scores from a canonical correspondence analysis (CCA) (CANOCO of ter Braak (1987)). Only CCA equations were used for chrysophyte inferred pH (CIpH) and CIalk. Low sedimentary concentrations of chrysophyte remains in five of the lakes led to a reduced number of lakes in the chrysophyte calibration set ($n = 58$). Statistics for the calibration equations are given in table 2. See Battarbee & Charles (1987) for a general explanation of the calibration approach.

Table 2. Statistics for calibration equations

(Chrysophyte statistics from Dixit *et al.* (1990).)

	method	Ho[a]	r^2	*Se*
diatoms ($n = 63$)				
pH	clusters	< 0.001	0.86	0.29
	DECORANA	< 0.001	0.84	0.29
	CCA (CANOCO)	< 0.001	0.93	0.29
alkalinity	clusters	< 0.001	0.79	27
	DECORANA	< 0.001	0.75	27
	CCA (CANOCO)	< 0.001	0.88	29
chrysophytes ($n = 58$)				
pH	CCA (CANOCO)	< 0.001	0.74	0.35
alkalinity	CCA (CANOCO)	< 0.001	0.62	138

[a] Probability that null hypothesis (Ho) is true.

Microfossil stratigraphies

In the following text, a pH value in parentheses following a diatom taxon is its abundance weighted mean pH in the calibration data set. Major shifts in diatom assemblages in the Mud Pond cores occur around and above 8 cm (figure 2). Minor peaks at 7 cm (*ca.* 1900) of *Melosira distans* var. *distans* (5.75) and *Fragilaria virescens* (6.00) may be responses to lake enrichment from

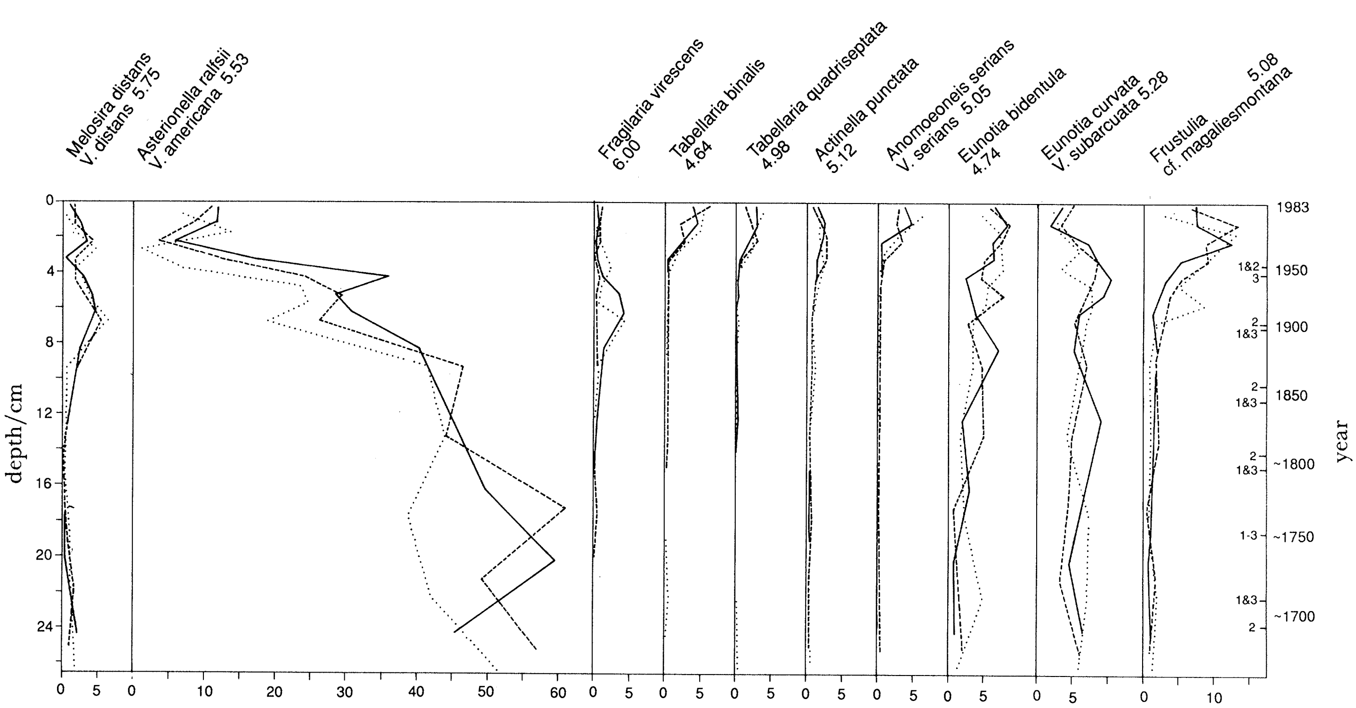

Figure 2. Percent abundance of common and informative diatom taxa in Mud Pond core 2 (master core) (——) and in cores 1 (-----) and 3 (· · · ·) (three replicate cores). The abundance weighted mean pH in the calibration data set is given for each taxon. Stratigraphy is presented on a scale of equal increments of depth (cm). Dates (1983, 1950, 1900, etc.) along that depth scale for each numbered core are given by tick marks on the right. Dates are based on ^{210}Pb (to *ca.* 1850) and pollen chronostratigraphic markers (to *ca.* 1800), and for older–deeper sediment on down-core linear extrapolation of the slope of the 1850–1800 part of the date–depth curve (Davis *et al.* 1990).

selective logging of the catchment at that time. Above 6 cm (*ca.* 1920) the changes indicate an acidification. Taxa associated with water pH of *ca.* 4.5 to 5.0 increase, and taxa associated with water pH of *ca.* 5.3 to 6.0 decrease. For example, the benthic *Tabellaria binalis* (4.64) and *T. quadriseptata* (4.98) increase and the planktonic *Asterionella ralfsii* var. *americana* (5.53) decrease. Chrysophyte shifts (Smol & Dixit 1990) support the diatom indications of a recent acidification. Most notable are the replacements of *Mallomonas crassisquama* and *M. allorgei* by *M. acaroides* var. *muskokana*, *M. hamata* and *M. punctifera*.

Microfossil stratigraphies for Haystack Pond also suggest a recent acidification (Davis *et al.* 1990). At that lake, *Tabellaria flocculosa* strain 3 (5.73) (Koppen 1975) is largely replaced by *T. quadriseptata* (4.98) and *Eunotia pectinalis* var. *minor* (5.00). Chrysophyte counts are overwhelmingly dominated (more than 90%) by one taxon (*M. crassisquama*) and are not useful for pH and alk inference. Microfossil assemblages at Little Long Pond exhibit little change above 40 cm (since *ca.* 1700) except that in the top few centimetres, *Cyclotella stelligera* (6.6) and *Tabellaria flocculosa* strain 3P (6.30) are partially replaced by more acidophilic forms including *Melosira perglabra* (5.6).

Different cores from the same lake

The diatom stratigraphies of Mud Pond cores 1, 2 and 3 are very similar to each other (figure 2). As for diatoms, stratigraphic replication of Mud Pond chrysophytes is very good (Smol & Dixit 1990). At Haystack Pond, stratigraphic replication for diatoms and chrysophytes is not as precise as at Mud Pond but is still quite good (Davis *et al.* 1990; Smol & Dixit 1990).

Replication of DIpH based on the same equation applied to multiple cores from the same lake is excellent for both absolute pH and chronology of change (figure 3). For Haystack Pond and Mud Pond, pre-1900 inferences for the same date generally agree to within 0.1–0.2 IpH units. For post-1900 at Haystack Pond, IpH ranges more widely (to about 0.3 unit), but acidification is indicated in all cases. Replication of CIpH at Mud Pond is about as precise as for DIpH.

Core replication for DIalk appears to be less precise than for DIpH (figure 3). Nevertheless, replicate Ialk values for the same date generally have small differences (less than or about 15 μeq l^{-1}); post-1900 trends are in good agreement, with certain exceptions, for example, slopes for CCA at Haystack Pond and variance for cluster at Mud Pond. Core replication of CIalk at Mud Pond is also very good.

Different equations applied at the same lake

For pre-1900, DIpH based on the three equations at the same lake ranges only about 0.1 units, except for Little Long Pond where the CCA equation gives values about 0.2 units higher than the other two equations (figure 3). Post-1900 inferences range slightly more widely. Trends are similar to each other but are dampened for DECORANA IpH (especially at Haystack Pond). The three equations for DIalk give results for pre-1900 at Haystack Pond that vary little (less than or about 10 μeq l^{-1}). At Mud Pond, DECORANA DIalk is about 20 μeq l^{-1} higher than cluster DIalk. Pre-1900 DIalk at Little Long Pond ranges a little more widely: about 20–30 μeq l^{-1}. Post-1900 trends in DIalk at each lake are similar for the three equations, except

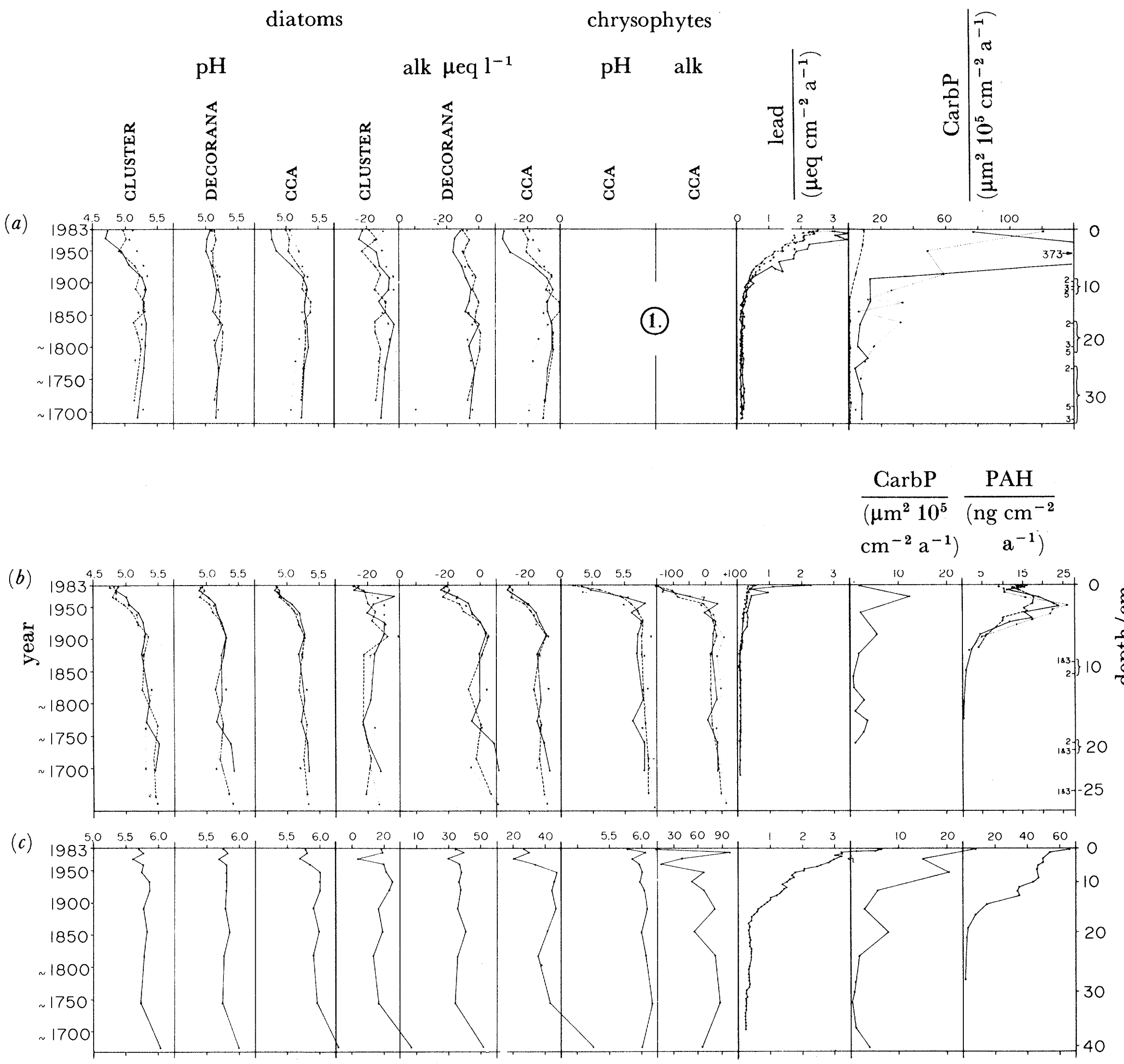

FIGURE 3. Replicate core pH and alkalinity reconstructions for (*a*) Haystack Pond and (*b*) Mud Pond, and single core reconstructions for (*c*) Little Long Pond based on diatoms and scaled chrysophytes and on three calibration approaches (CLUSTER, DECORANA and CCA). Sedimentary indicators of fossil fuel combustion: lead, carbonaceous particles (CarbP) and polycyclic aromatic hydrocarbons (PAH) are given for comparison. Stratigraphy is presented on a scale of equal increments of time (year date) (see legend of figure 2 for dating techniques). Depths (10 cm, 20 cm, etc.) along that time scale for each numbered core are indicated by tick marks on the right. ((*a*), (——) core 5, (----) core 3, (····) core 2; (*b*), (——) core 2, (----) core 1, (····) core 3; (*c*) core 2 only; (1.), overwhelming dominance by *Mallomonas crassisquama* (unsuitable for pH and alk inferences).)

are most invariable for DECORANA at Haystack Pond, most variable for CLUSTER at Mud Pond, and most pronounced for CCA at Little Long Pond.

DIATOMS VERSUS CHRYSOPHYTES

At Mud Pond, diatom analysis leads to slightly different conclusions than chrysophyte analysis (figure 3). Pre-1900 DIpH and DIalk values are lower than CI values; and post-1960 DIalk is much higher than CIalk. The CIalk of about -150 μeq l^{-1} derived from the surface sediment sample is inconsistent with the mean measured alk of -23 μeq l^{-1}, whereas diatom ialk based on surface sediment is in good agreement with the measured value (table 1). Statistics for cialk CCA equations are poorer than for the diatom alk equations (table 2). Although the direction and timing of change of cipH and cialk are the same as for the diatom inferences, the amount of change is greater for chrysophytes. At Little Long Pond, cipH is in fair agreement with dipH, but ialk values are somewhat higher when based on chrysophytes. At Little Long Pond, cialk displays more extreme responses (than dialk) to shifts in individual taxa.

pH and alkalinity of the lakes during the past 300 years

These three lakes had low pH and alk in the 18th and 19th centuries, before any suggestion of anthropogenic acidification (figure 3). The pre-1900 dipH is about 5.2–5.3 and dialk about 0 to -20 μeq l^{-1} for Haystack Pond and Mud Pond and, respectively, about 5.7–6.0 and about +20 to 70 for Little Long Pond. A rapid acidification started between 1920 and 1930 in Haystack Pond and in Mud Pond. Little Long Pond changed little in the three centuries covered by the core, although there is some suggestion of slight acidification starting about 1950.

The aforementioned recent acidifications of Haystack Pond and Mud Pond are correlated with steep increases in sedimentary indicators of fossil fuel combustion (Pb, CarbP, PAH; see figure 3).

Discussion

These results are encouraging in regard to the use of single cores from the deep part of lakes. In Mud Pond and in Haystack Pond, microfossil stratigraphies and environmental inferences from different cores are very similar. These results say nothing about differences that would arise from coring over wide ranges of water depths in the same lake. That consideration is somewhat peripheral here because for both the calibrations and the palaeolimnological reconstructions only deepest-water sediments were used. It has not been our intention to reconstruct whole lake communities of siliceous algae. If one wished to do so, the study of cores from a wide range of water depths would be necessary. See Frey (1988) regarding differential representation of littoral versus pelagial remains in sediments.

These results demonstrate the comparability of different well-established calibration approaches for pH and alk inference. Although only three different approaches (equations) for each parameter were used, the results do suggest that conclusions would only occasionally be misleading because of the use of different calibration approaches. Comparisons of results from different lakes where different calibration approaches were used, both within and between regions, have been extensively and effectively made in the review papers cited in the Introduction. However, in the present study a few differences between inferences from different equations did arise. Those differences do point to the desirability in comparative studies of using the same calibration approach as much as possible.

Some differences arose at Mud Pond between the diatom and chrysophyte reconstructions. Although the timing and direction of inferred change (acidification) in the 20th century were the same regardless of the algal group used, the absolute ipH and ialk differed. Given the greater amounts of variance explained (r^2) and smaller standard errors of the diatom equations (table 2), we place more confidence in them at this time. Apart from any statistical or methodological considerations, however, we would expect to encounter real differences between inferences based on the two groups. In lakes, diatom taxa characteristic of both planktonic–pelagial and benthic–littoral communities occur. However, in most strongly acidic (pH less than or about 5.0) lakes, planktonic diatom taxa are virtually absent. On the other hand, scaled chrysophytes occur only in the planktonic community and may be abundant in strongly acidic lakes. The seasonality of these groups also differs. Although the use of combined diatom–chrysophyte calibrations can be more appropriate for some inferences (Charles & Smol 1988), it may also be desirable to carry out separate diatom and chrysophyte reconstructions for the different habitat information they may reveal.

Haystack Pond and Mud Pond started to acidify rapidly around 1920–1930 during the period of maximum sulphur emission in northeastern United States (Husar 1986). Close correlation of the acidifications with sedimentary indicators of fallout of pollutants from fossil fuel combustion, and the usual association of these pollutants with acid deposition, suggest that the acidifications are due to acid deposition. Absence of correlated changes in land-use and any substantial changes in vegetation and soils in the catchments largely eliminate such possible causes of acidification.

The catchment of Little Long Pond includes that of Mud Pond. The land-use history of the combined catchment has been approximately uniform. However, Little Long Pond has not undergone a major anthropogenic acidification. The Mud Pond catchment (24 % of Little Long's) has steeper slopes and thinner soils. Water residence time in Mud Pond is about a tenth of that in Little Long Pond. For these reasons, Mud Pond responds more directly to rain-water chemistry and acid deposition than Little Long Pond.

Haystack Pond and Mud Pond had inferred pH values of less than 5.5 and lacked alk before anthropogenic acidification. Little Long Pond has not had quite as low a pH, and has had positive, but very low, alk values during the period of the reconstruction. These water chemistries, and the characteristics of the catchments, have made these lakes very sensitive to acidification by acid deposition (Little Long Pond least so). More than 50 % of the lakes that have been shown by palaeolimnological study to have undergone a recent rapid acidification were in this extremely acid-sensitive condition before acidification (Charles *et al.* 1989).

The three lakes studied here are far more sensitive to acid deposition than is typical for lakes in northern New England. Although many hundreds of lakes (and catchments) in the region have low acid neutralizing capacity (Brakke *et al.* 1988), only a very small percentage (probably less than 1 %) of these have the extreme characteristics of these three lakes and their catchments. This is not to say that fallout of air pollutants is not having detrimental effects on the larger population of lakes.

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