Paleolimnological Investigation of Freshwater Lake Sediments in Insular Newfoundland

Part 2: Downcore Diatom Stratigraphies and Historical pH Profiles for Seven Lakes

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PALEOLIMNOLOGICAL INVESTIGATION OF FRESHWATER LAKE SEDIMENTS IN INSULAR NEWFOUNDLAND

PART 2: DOWNCORE DIATOM STRATIGRAPHIES AND HISTORICAL pH PROFILES FOR SEVEN LAKES

by

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ABSTRACT

Scruton, D. A., J. K. Elner, and G. D. Howell. 1987. Paleolimnological investigation of freshwater lake sediments in insular Newfoundland. Part 2: Downcore diatom stratigraphies and historical pH profiles for seven lakes. Can. Tech. Rep. Fish. Aquat. Sci. 1521 (Part 2): v + 67 p.

The pH profiles of seven insular Newfoundland lakes were reconstructed from fossilized diatom remains in one-cm strata from sediment cores ranging from 25 cm to 60 cm in length. Inferred pH for each strata was calculated from a region specific Index B transfer function developed from surface sediments for 34 Newfoundland lakes of wide ranging acidity (pH 4.86 to 7.72) (see Part 1 of this report series). Attempts to put a time scale on the pH profiles, using ^{210}Pb dating, were successful for four of the seven lakes.

Three lakes (#'s 8, 204, and Stephenson's Pond) demonstrated a statistically significant trend ($P < 0.01$) to increasing acidity over the core length. A trend to increasing pH was statistically significant in two lakes (lake 660 and Aides Pond) while no significant trend was evident in lakes 12 and 219. In the recent strata, a pH decline of 0.2 to 0.3 of a pH unit was evident in four of the lakes. This decline was postulated to be attributable to acid deposition and possibly watershed disturbances (forest fires, soil humification). In one lake the approximate onset of acidification was dated at circa 1946. The magnitude of pH change compares favourably with similar studies in New Brunswick, Nova Scotia, and New England but is considerably less than lakes studied from severely acidified regions of the world.

RÉSUMÉ

Scruton, D. A., J. K. Elner, and G. D. Howell. 1987. Paleolimnological investigation of freshwater lake sediments in insular Newfoundland. Part 2: Downcore diatom stratigraphies and historical pH profiles for seven lakes. Can. Tech. Rep. Fish. Aquat. Sci. 1521 (Part 2): v + 67 p.

On a reconstitué les profils de pH de sept lacs terre-neuviens à partir de strates de 1 cm de diatomées fossiles révélées dans les carottes sédimentaires dont la longueur varie de 25 à 60 cm. Le pH supposé de chaque strate a été calculé à l'aide d'une fonction de transfert pour l'indice B particulière à chaque région, élaborée en fonction des valeurs obtenues des sédiments superficiels de 34 lacs terre-neuviens à grande gamme d'acidité (pH 4,86 à 7,72) (voir Partie 1 de la présente série de rapports). La détermination d'une échelle temporelle des profils de pH par datation au ^{210}Pb a réussi dans le cas de quatre des sept lacs.

Trois lacs (nos 8, 204 et Stephenson's Pond) ont montré une tendance statistiquement significative (P < 0,01) vers une acidité croissante, comme l'a révélée la carotte. Une tendance à la hausse du pH était statistiquement significative dans deux lacs (lac 660 et Aides Pond) tandis qu'aucune tendance n'était évidente dans les lacs 12 et 219. Dans les strates récentes, une baisse du pH de 0,2 à 0,3 unité était évidente dans quatre lacs. On formule l'hypothèse que ce déclin est le résultat du dépôt d'acides ou peut-être de perturbations du bassin versant (feux de forêt, humidification du sol). Dans un lac particulier, le début de l'acidification a été daté aux environs de 1946. L'importance de la variation du pH soutient la comparaison avec les résultats d'études semblables menées au Nouveau-Brunswick, en Nouvelle-Écosse et en Nouvelle-Angleterre mais elle est beaucoup moins marquée que celle relevée dans des lacs de régions gravement acidifiées autour du globe.

INTRODUCTION

The acidification of freshwater lakes in response to the deposition of strong acids in precipitation is now a well-documented global phenomenon. Acidification damage has been reported from Scandinavia, mostly southern Norway and Sweden (Almer et al. 1974; Overrein et al. 1980) and in other regions of Northern Europe, including the United Kingdom (Wells et al. 1985), Finland (Anonymous 1982) and West Germany (Kraus et al. 1985). In North America, lake acidification was first reported from LaCloche Mountains in Ontario (Beamish and Harvey 1972) and the Adirondack Mountains in the northeastern U.S.A. (Schofield 1976), and is now recognized as a more widespread problem in northeastern North America (Harvey et al. 1981; Bangay and Riordan 1983; Environmental Protection Agency 1983).

In Atlantic Canada, pH declines in 16 lakes in the Halifax region over a 21-year period were first reported by Watt et al. (1979). The acidification of seven Atlantic salmon rivers in southeastern Nova Scotia was subsequently reported (Watt et al. 1983). Underwood and Josselyn (1980), however, have suggested that natural organic acidity and proximity to local emission sources may be the primary causes of lake acidification in Nova Scotia.

In insular Newfoundland, recent lake surveys have confirmed the extreme susceptibility of the island's freshwaters to anthropogenic acidification but at present there is no evidence of widespread acidification damage (Scruton 1983, 1986; Clair 1981). Sensitive freshwaters are demonstrating considerable alkalinity depletion but many of the most acidic systems (pH < 5.0) are weak organic acid dominated. Due to the preponderance of organic acid dominated systems, it has been difficult to determine the contribution of atmospheric deposition of strong acids (NO_3- and SO_4^{-2}) to the acid-base chemistry of the region's lakes. Many researchers now believe that naturally acidic, brown water systems are even more sensitive to anthropogenic acidification than oligotrophic water bodies, in strictly an acid-base sense, although the same may not necessarily be true for the susceptibility of biological communities (Jones et al. 1986). The insular Newfoundland region is thought to be demonstrating the initial effects of anthropogenic acid deposition, that is the depletion of buffering capacity in sensitive, low alkalinity lakes, without concurrent chronically depressed pH levels and corresponding biological perturbations (Scruton 1983; 1986). Parts of insular Newfoundland, the southwest coast in particular, could be on the threshold of a potential acidification problem (Scruton and Taylor 1987). However, the absence of any historical water chemistry data for sensitive, low order lakes precludes any comparison through time as additional evidence of progressing acidification.

There is evidence that long-term acidification is a natural process in lakes underlain by resistant bicarbonate-poor bedrock, however, analyses of diatom microfossils in lake sediments from northwestern Europe and eastern North America have shown rapid pH declines over the last 100 to 150 years (Battarbee 1984). Consequently, the fossil record from freshwater lake sediments has become an increasingly valuable tool to assess the possible effects of the deposition of atmospheric pollutants on dilute, acid-sensitive lakes (Merilainen 1967; Davis and Berge 1980; Del Prete and Schofield 1981; Charles 1984; Renberg and Hellberg 1982; Flower and Battarbee 1983; Delorme et al. 1984; Dickman et al. 1984). Contemporary relationships between diatoms and water chemistry (pH) is quantifiable and useful in interpretation of present and past aquatic environments (Battarbee 1979). A method using the relationship between preserved diatom assemblages in recently deposited sediment and contemporary lake pH has been widely used to infer pH histories (Brakke 1984; Davis and Anderson 1985). There are two distinct approaches to relating diatom sub-fossil data to pH. The first method uses diatom floristic data to determine a quantitative index, the $\log_{10}$ transformation of which is related to pH by linear regression (Nygaard 1956; Renberg and Hellberg 1982). The second method develops a direct relationship between the diatom assemblage data and lake pH by means of multiple regression analysis, principal components analysis, or related ordination techniques (Davis et al. 1980; Davis and Berge 1980; Charles 1985; Davis and Anderson 1985). These calibration equations, or transfer functions, are then used to infer historical pH of lakes by applying these equations to diatom remains deeper in the sediments (Davis and Berge 1980; Renberg and Hellberg 1982). Deeper sediments in the lakes can be dated by ^{210}Pb, ^{137}Cs, pollen (largely by *Ambrosia* sp.), and charcoal analysis to put a time scale on pH changes and onset of acidification (Davis et al. 1983; Charles and Whitehead 1985).

In the first part of this report series (Scruton and Elner 1986), relationships between fossil diatoms in recent sediments (top 1 cm) and contemporary water chemistry (pH) were explored using two widely-applied paleolimnological indices. The two indices, Index alpha (α) (Nygaard 1956) and Index B (Renberg and Hellberg 1982), were regressed (Log_{10}) with contemporary pH to yield equations, or transfer functions, for computation of inferred pH, unique to the Island of Newfoundland. Index B is considered superior to Index alpha (α) by several authors (Charles 1985; Davis and Anderson 1985) because it incorporates more information into its calculation and is not as sensitive to variations in numbers of alkaliphilous taxa. In addition, if no alkaline taxa are present in a sample, as in extremely acidic lakes, Index alpha (α) cannot be calculated. Index alpha (α) is therefore biased in its ability to infer the pH of very acid lakes (or strata). The Index B transfer function for insular Newfoundland was statistically superior to the Index alpha (α) equation (better regression coefficient, lower standard error, fewer outliers) (Scruton and Elner 1986) and, for these combined reasons, the Index B transfer function has been adopted as the preferred method to calculate inferential pH in downcore sediment strata.

STUDY OBJECTIVES AND APPROACH

The objectives and rationale for paleoecological study of lake acidification in insular Newfoundland have been previously outlined in Part 1 of this report series (Scruton and Elner 1986). The time frame associated with past/ongoing/future paleoecological research and projected goals are similarly outlined.

Due to the time frame involved in this research program, the data arising from the study is being published as a report series. The first report in this series, entitled "Part 1: Relationships between Diatom Sub-Fossils in Surface Sediments and Contemporary pH from Thirty-Four Lakes" has been published

(Scruton and Elner 1986). This report, entitled "Part 2: Downcore Diatom Stratigraphies and Historical pH Profiles for Seven Lakes" represents the second in the series. The third report will deal with spatial and temporal (inferred historical) variation in major cations and metals in lake sediments, particularly with respect to evidence for atmospheric deposition and mobilization of these elements from the watershed. Information obtained in future studies will be published as a continuation of the report series and/or as summary publications in the primary literature.

MATERIALS AND METHODS

LAKE SELECTION

Seven lakes were selected for downcore collection and subsequent diatom analysis for determination of historical (inferential) pH stratigraphies. Five lakes (#'s 8, 12, 204, 219, 660) were selected from the Department of Fisheries and Oceans' (DFO's) acid rain lake monitoring network while two lakes (Stephensons Pond, Aides Pond) were included from Environment Canada's (Inland Waters Directorate, Water Quality Branch, Moncton, N.B.) LRTAP monitoring network. Stephensons Pond has been previously studied in DFO's lake inventory program (in 1981) and had been assigned lake #206. Lakes were selected from the two geographical regions, the south coast of Newfoundland and eastern side of the Northern Peninsula, where susceptibility to the effects of acid precipitation is acknowledged to be greatest (Scruton 1983). Lakes selected were acidic (pH generally less than 5.5, excepting Aides Pond) and low in alkalinity (0-56 μeqL^{-1}), had simple morphometry and hydrology (six of the seven lakes were headwater or second order ponds), and represented the lower range of organic influence characteristic of most insular Newfoundland lakes (colour values of 10-50 TCU, organic anion concentrations from 30-50 μeqL^{-1}; Tables 1 and 2). Excess sulphate values at the time of core collection were also relatively low (10-31 μeqL^{-1}, Table 2). Sulphate values (as an indicator of anthropogenic acid deposition) and water colour/$COOH^-$ values (as an indicator of natural acid production) are expected to be near annual lows due to considerations of seasonality (see Scruton and Elner 1986 for an explanation). Lake #204, previously assessed as demonstrating the effects of anthropogenic acidification on the basis of the cation/sulphate/pH considerations as expressed by Henriksen's nomograph (Scruton 1983), was included in the study to see if the paleoecological record supported the apparent chemical evidence for acidification.

SAMPLE COLLECTION

Cores were collected from the seven lakes selected for downcoring from August 14 to 20, 1984. Lakes were accessed by Bell Jet Ranger 206B helicopter and coring operations were completed from the floats of this aircraft. Coring sites were established over the point of apparent maximum depth (mid-lake, maximum depth was not determined by bathymetry) and the depth of the site was recorded. Cores were collected using a 10-cm diameter, light-weight Williams and Pashley (1979) corer, designed for use in unconsolidated deposits. The corer was modified (shortened in length) for safe operation from a helicopter.

One core per lake was obtained and all cores retained for analysis had an undisrupted sediment/water interface.

Cores were extruded and sectioned immediately at the lake side so as to minimize disturbance of the highly flocculent sediment. The extrusion device used hydraulic water pressure to upwardly (vertically) extrude the core. A collar on the extrusion device shaved off the outer 1-cm diameter of core, so as to remove any sediment that may have experienced smearing along the inside of the core tube. One centimetre horizons were removed from each core by spoon (in very watery sediment) or by spatula (in more-consolidated sediment). Cores were sectioned at 1 cm intervals for the first 25 cm of core length, and subsequently a 1 cm horizon was obtained at 5 cm intervals for the remainder of the core, until the extrusion device was observed to interfere with the consolidation of the core. Each horizon was transferred to pre-labelled "whirl-pak" bags and frozen upon return to the field laboratory.

WATER AND SEDIMENT SAMPLE ANALYSIS

Water sample collection methods and parameters analyzed have been described in Part 1 of this report series. Analytical methods followed those described by Environment Canada (1979) and the American Public Health Association et al. (1975). Details on analytical procedures and detection limits are given in Scruton (1983). The samples were analyzed for the following parameters:

pH	Sulphate
Alkalinity	Chloride
Dissolved Organic Carbon	Orthophosphate as P
Colour	Nitrate as N
Turbidity	Aluminum
Calcium	Iron
Magnesium	Manganese
Sodium	Total hardness (calculation)
Potassium	Bicarbonate (calculation)

Total inflection point (or Gran) alkalinity and pH (field) were determined at a field laboratory within 24 hr of sample collection. The field measured pH value, used in this paleolimnological study as contemporary pH, was the initial pH reading taken in the acidimetric titration for alkalinity determination.

Non-marine concentrations of cations and sulphate were calculated after Thompson (1982). Organic anions (as $COOH^-$ in ··eq L^{-1}) were calculated from pH and dissolved organic carbon (DOC) after Oliver et al. (1983).

Quality assurance was determined by ion balance (after Environment Canada 1979), comparison of measured and calculated conductivity, blind batching and spiking of samples and through participation in round-robin interlaboratory quality assurance check program.

Procedures used in laboratory preparation of sediment samples (strata), cleaning of fossil diatoms by acid digestion, and preparation of slide for taxonomic identification and enumeration are detailed in Part 1 of this report series.

Each sediment sample was thawed and mixed thoroughly prior to being divided into three aliquots for the required analyses. One aliquot was reserved for diatom identification and counts, a second for chemical analyses, and a third for determination of wet/dry weights and for dating by ^{210}Pb analysis.

Diatoms were cleaned by an acid digestion technique. Small aliquots of sediment (0.5 ml) were heated to boiling in a 5:1 mixture of concentrated $H_2SO_4.HNO_3$ acid. Cleaned samples were cooled, centrifuged, the supernatant discarded and the diatom pellet cleaned in deionized water. The diatom suspension was diluted with deionized water, mounted in "Hyrax" on slides, and ringed for permanence.

DIATOM TAXONOMY AND ENUMERATION

Diatom identification was based in the most part on the floras of Cleve-Euler (1951-1955), Hustedt (1930), Molder and Tynni (1967-1973), Patrick and Reimer (1966, 1975) and Tynni (1975-1981) and also included Renberg (1977). Taxa identified for each stratum were assigned to pH tolerance groups based on the Hustedt (1937-1939) classification as follows:

1) Alkalibiontic (Alkb): occurring at pH values > 7;

2) Alkaliphilous (Alk): occurring at pH about 7 with widest distribution at pH 7;

3) Indifferent or Circumneutral (C): equal occurrences on both side of pH 7;

4) Acidophilous (Ac): occurring at about pH 7 with widest distribution at pH < 7;

5) Acidobiontic (Ab): occurring at pH values < 7, optimum distribution at pH 5.5 and under.

The pH tolerance category of each taxa was based on a review of the ecological requirements of each species from the pertinent literature (references as above and also including Beaver 1981; Lowe 1974; Merilainen 1967; and others). A list of the species identified (with taxonomic reference) and assigned pH tolerance category is contained in Appendix 1 of Part 1 of the report series. A minimum of 300 frustules were counted for each sediment stratum.

DETERMINATION OF pH STRATIGRAPHIES

The inferred (historical) pH of each strata was calculated on the basis of the Index B- contemporary pH transfer function established in Part 1 of this report series. Index B for each stratum (after Renberg and Hellberg 1982) was calculated from the following equation:

$$\text{Index B} = \frac{\%C + 5 \times \%\,Ac + 40 \times \%\,Ab}{\%C + 3.5 \times \%\,Alk + 108 \times \%\,Alkb} \qquad (1)$$

where C, Ac, Ab, Alk, and Alkb correspond to pH tolerance groups of Hustedt (1937-39). Inferrential pH was computed from Index B (above) after the following equation (Scruton and Elner 1986):

$$\text{pH (inferred)} = 6.63 - 0.87 \log_{10} \text{Index B} \qquad (2)$$
$$(r^2 = 0.79,\ S_e = \pm\ 0.36,\ n = 34)$$

Cluster analysis of the diatom frequency data for each stratum (only selected taxa, see Part 1 of this report series) was completed using a Biomedical Computer Programs (BMDP) package, (Dixon and Brown 1979). The distance matrix (amalgamation distance) in the cluster analysis is based on a chi-square statistic.

DATING OF SEDIMENT STRATA

A chronology was attempted for each of the seven sediment cores, using ^{210}Pb radioisotope analysis of the uppermost twelve - one cm horizons. Lead (^{210}Pb) is a naturally occurring radionuclide, subject to decay, and is useful as a continuous time marker (Binford 1984). Lead (^{210}Pb) in each sediment stratum was determined by alpha-ray spectoral analysis of the daughter decay product ^{210}Po, (in the ^{238}U decay series) with a pre-processing spike of ^{208}Po added for the purposes of calibration (Eakins and Morrison 1978; Flynn 1968).

Dates were determined from the ^{210}Pb spectoral analysis by the Constant Rate of Supply (CRS) or Constant Flux (CF) model of Appleby and Oldfield (1978). This model assumes that the flux of ^{210}Pb to sediments is a constant, and is independent of the rate of sedimentation, while the sedimentation rate may vary over time.

RESULTS

In Part 1 of this report series, relationships were developed between fossilized diatom remains in the surface sediments (0-1 cm) and contemporary water pH in 34 lakes of wide ranging acidity (4.86 to 7.72). Calibration equations for calculation of inferred pH were determined using two widely applied paleolimnological indices, Index alpha (α) (Nygaard 1956) and Index B (Renberg and Hellberg 1982). The strengths and weaknesses of these two indices, and index methods in general, in calculating historical pH were evaluated. The transfer functions (calibration equations) computed from the two indices were compared, and subsequently evaluated with respect to equations developed for pH-diatom relationships from other global regions. The Index B transfer function is considered superior to that developed from Index alpha (α) (Renberg and Hellberg 1982; Davis and Anderson 1985; Charles 1985). For insular Newfoundland lakes, the pH-log Index B relationship had better regression statistics and fewer outliers than did the pH-log Index alpha (α) equation. In this report, inferred pH histories of seven lakes have been developed using the Index B transfer function and diatom assembledge data for 1 cm horizons obtained from 25 to 60 cm long sediment cores. A lead-210 (^{210}Pb) chronology was attempted on each core to put a time scale on observed pH changes.

For each of the seven study lakes the following information is presented and evaluated:

- A stratigraphic representation of the most commonly occurring diatom taxa in each core (Figs. 3, 6, 9, 13, 16, 19, and 22).

- A stratigraphic representation of the frequencies of different pH tolerance groups (acidobionts, acidophils, etc.) with a reconstructed (inferred) pH profile using the Index B transfer function (Figs. 4, 7, 10, 14, 17, 20 and 23). For cores where lead (^{210}Pb) dating was successful, extrapolated dates are indicated beside the inferred pH profile. Extrapolated dates are the average for each 1 cm horizon based on polonium (^{208}Po) activity and sedimentation rate.

- A cluster analysis of the diatom assemblage data for all horizons from each core (Figs. 5, 8, 12, 15, 18, 21 and 24).

- A regression of Index B inferred pH versus core depth (horizon depth) for each lake, using both simple linear regression (SR) and a multiplicative regression (MR), which emphasizes apparent trends (Table 3, Fig. 25).

It is initially important to note that the regression of inferred pH on core depth is intended to demonstrate the significance of acidity trends throughout the entire lake history covered by the core length. This of course does not address significance of acidity changes in recent strata, which are considered to cover the period of increases in anthroprogenic acid deposition (~50 yr b.p.). Inferred pH is not expected to change in a linear fashion with depth, as pH is log intensity function of hydrogen ion (H^+) activity. Additionally, owing to the nature of the acidmetric titration of bicarbonate, more rapid pH declines in response to acid deposition are expected over the pH range 6.5 to 5.5, than on either side of this range (Stumm and Morgan 1981). In evaluating response to acidic deposition the uppermost strata (top 5 cm in particular) are of greatest interest.

LAKE 8

The sediment core collected from Lake 8 was 30 cm in length (sectioned length) and had a moisture content that varied between 94% and 98% water. The sediment of the entire core was a uniform, dark brown color and had the texture of organic matter. A low cumulative dry weight (0.52 g cm^{-2} at a depth of 12 cm) and the low and constant polonium (^{210}Po) activities throughout the core prevented calculation of a sediment accumulation rate and subsequent dating of this core. Consequently, no time scale can be put on the inferred pH history.

An average of 61 taxa were identified from each horizon of the Lake 8 sediment core and between 500 and 600 diatom valves were identified and enumerated from each stratum. The diatom flora of this lake was dominated by acidobiontic and acidophilous taxa throughout the time period encompassed by the core. The acidobionts Semiorbis hemicyclus (Ehr.) Patr. and Tabellaria binalis (Ehr.) Grun. increase in frequency towards the top of the core (Fig. 3). Tabellaria quadricepta Kunds. showed an increase in numbers in association with Tabellaria binalis. An increase in the acidophil Cyclotella glomerata Bach., from horizons 9 and 10 to the surface, was accompanied by a corresponding decrease in Melosira distans (Ehr.) Kütz., also ranked as acidophilous (Fig. 3).

A cluster analysis of the floristic data from each stratum indicates a clear division into two major clusters of horizons according to inferred pH (Fig. 5). Cluster I contains those horizons within the pH range 5.3 to 5.5 (sediment horizons 1 through 9) while cluster II contains horizons within the pH range 5.5 to 5.8 (horizons 10 through 30). Sediment horizon 13 did not cluster well.

A pH of 5.34 was recorded for the water sample collected from Lake 8 in August, 1984, which is comparable to an inferred pH value of 5.3 calculated for the surface sediment sample from the core (Fig. 5). The histogram of diatom occurrence, as pH tolerance groups (Fig. 4), shows a gradual increase in the numbers of acidobionts in the upper 9 or 10 horizons of core 8, with a corresponding decline in the proportion of alkaliphilous and acidophilous taxa. The inferred pH profile demonstrates a rather stable pH history (pH of 5.5 to 5.6) from horizon 30 to horizon 13, followed by an appreciable rise in pH to 5.8 at horizon 12. The inferred pH then declines to 5.3 at horizon 9 and remains below pH 5.5 for the upper 9 cm of the core. The lowest inferred pH is 5.2 at horizon 5. A decline in pH of 0.3 of a unit occurred from horizon 25 to the core surface sample (pH of 5.3). The maximum inferred difference (decline) between historical and present day pH is 0.5 of a unit (from strata 12, pH 5.8, to pH 5.3 at the surface). Including horizons 25 and 30, and despite the general "noise" in this pH profile, the increase in acidity (pH decline) to present day is statistically significant ($r = 0.56$, $S_e = \pm 0.11$, $P = 0.010$, $n = 20$) (Table 3).

LAKE 12

The sediment core collected from Lake 12 was 25 cm long (sectioned length) and had a water content of between 90% and 99%. The sediment was similar in color and texture to that of Lake 8. The cumulative dry weight of this sediment core was even lower (0.41 g cm^{-2} at 12 cm horizon) than the core from Lake 8 and, with the low and constant polonium (^{210}Po) activities for the top 12 horizons, no dating was possible.

An average of 56 taxa were identified from the 20 strata and counts of diatom valves varied between 300 and 400 for each horizon. Again, the diatom flora was dominated by acidobiontic and acidophilous taxa (Fig. 7). The distribution of species is generally uniform throughout the lower 20 cm of core, although some changes in floristic composition are apparent in the top

5 cm of core. In this top 5 cm section, there are increases in the frequency of Asterionella formosa Hass., the Fragilarias, Tabellaria flocculosa (Roth.) Kütz. and Coscinodiscus lacustris Grun., with corresponding decreases in Cyclotella glomerata and Melosira distans (Fig. 6). A cluster analysis of these floristic data did not divide the sediment profile into distinct groups of horizons according to similarity in pH (Fig. 8).

A pH of 5.13 was recorded for the water sample from Lake 12 in August, 1984 and a pH of 4.9 in summer-fall of 1981, as compared to a surface sediment inferred pH value of 5.4 obtained for this core. The Index B inferred pH for the surface stratum does not correspond well with the measured water pH. There was very little change in the frequency of occurrence of diatoms in each pH tolerance group from the 25 cm horizon to the 5 cm horizon. Over the top 5 cm, there is a decrease in alkaliphilous taxa and corresponding increase in the circumneutral and acidobiontic groups (Fig. 7). The inferred pH profile demonstrates an initial increase in pH, from 5.5 (horizon 25) to 5.7 (horizon 22), followed by stable inferred pH from horizon 22 to horizon 4 (pH of 5.6 to 5.7). A pH decline of 0.3 of a unit was evident from horizon 4 (pH of 5.7) to the surface (pH of 5.4). Although the inferred pH profile demonstrated an increase in acidity in the top few cm of sediment, the relationship between horizon depth and inferred pH for the core as a whole was not statistically significant ($r = 0.31$, $S_e = \pm 0.08$, $p = 0.19$, $n = 20$) (Table 3).

LAKE 204

The Lake 204 sediment core was 60 cm in length (sectioned length) and the water content varied from 89% to 96%. The unusually constant polonium (^{210}Po) activities throughout the length of the core did not permit calculation of a sediment accumulation rate, and subsequent dating of strata.

An average of 65 taxa were identified in each horizon and between 500 and 700 diatom valves were counted for each sample. It is readily apparent that there is an increase in the occurrence of the acidobiontic taxa, Actinella punctata Lewis, Semiorbis hemicyclus and Tabellaria quadricepta, in the upper 7 to 8 cms of the core from Lake 204 (Fig. 9). These acid indicator species are generally insignificant (present at less than 1%) throughout the rest of the sediment core. The alkaliphilous species Fragilaria construens (Ehr.) Grun., F. construens v. venter (Ehr.) Grun., F. construens v. exigua (W. Sm.) Schulz. and F. pinnata Ehr., present in the lower core horizons, start to decline in frequency from 10 cm to become absent at the 5 cm horizon. There is also a decline in the planktonic centrics, Cyclotella glomerata and Melosira distans, towards the upper horizons of the core (Fig. 9). Cluster analysis of these floristic data produced three main groups of core horizons (Fig. 11). Group I, comprising horizons 1-7, is characterized by the inferred pH range 5.2-5.4. The second cluster, II, represents an inferred pH range from 5.3 to 5.8 and encompasses horizons 9 through 30. The remaining horizons, 40 to 60, fall into cluster III and have a range of inferred pH values from 5.8 to 5.9. The floristic data from horizons 14, 1, and 8 did not fit well into any of these pH cluster groups.

An inferred pH value of 5.3 was calculated for the surface sediment of the core from Lake 204 which is slightly higher than the value of 5.1 recorded for lake water in August, 1984. The pH of Lake 204 has been measured a total of six times between August 1981 and October, 1984 in inventory and monitoring studies, and has varied from 5.00 to 5.49 ($\bar{x}$ = 5.13).

The profile of pH tolerance groups is characterized by a very evident decrease in alkaliphilous and circumneutral species towards the top of the core (Fig. 10). Correspondingly, there is an increase in acidophilous taxa from horizon 60 to 14. At about horizon 14 the acidophilous group also start to decline in importance at which point there is a sharp increase in the numbers of acidobionts, a trend which continues to the top horizon (Fig. 10). The inferred pH profile (Fig. 10) and the distinct cluster groupings (Fig. 11) demonstrate a clear trend to declining pH over the length of the core. The maximum pH decrease to present day is 0.6 pH unit from horizon 40 (pH of 5.9) to pH 5.3 (at the surface). The lowest inferred pH is 5.2 for horizons 6 and 7. The relationship between inferred pH and depth for the entire core demonstrated a very highly significant decrease in pH from older to the more recent sediments ($r = 0.87$, S_e ±0.12, $P = 0.001$, $n = 20$) (Table 3).

Figure 11 presents the inferred pH profiles for Lake 204 as generated from Index B transfer functions determined by simple linear regression (SR; pH = 6.63 - 0.87 log Index B, $r^2 = 0.77$, $S_e = \pm 0.33$, $P < 0.001$) and by geometric mean regression (GMR; pH = 6.70 - 0.98 log Index B, $r^2 = 0.77$, $S_e = \pm$ 0.38, $P < 0.001$). In Part I of this report series the "difference plots" of surface diatom inferred pH versus measured water pH, using both the SR and GMR transfer functions, indicated a better correspondence between inferred and measured pH values using the GMR transfer function. The "difference plot" using the inferred pH values calculated by the GMR equation had a marginally higher regression coefficient (0.77 vs 0.76) and lower standard error (0.22 vs 0.27) while the slope of the equation was much closer to the 1:1 line (1.01 vs 0.89) than the "difference plot" for inferred pH values determined from the SR equation. A comparison of the statistics of the "difference plots" suggested the GMR transfer function was superior to the SR transfer function for computation of inferred pH for reconstruction of downcore pH stratigraphies. A comparison of the pH profiles developed using both transfer functions is made for Lake 204 only, and not for all other lakes, because the GMR approach has not been used in other regional studies of fossil diatom inferred evidence of lake acidification. The value and validity of using the GMR derived equation will be considered along with other refinements in development of transfer functions using index methods, multiple regression (MR), stepwise variable selection (SWVS) and principal components analysis (PCA) in a separate publication.

LAKE 219

The core from Lake 219 was 50 cm in length (sectioned length) and had the lowest water content of any of the DFO downcore lakes (81 to 97%). The sediments were a mid-brown color and had a fine grained texture. Lake 219 had a good sediment accumulation rate (3.78 mg cm^{-2} y^{-1}, $r^2 = 0.99$) determination for the top 6 horizons which represented a time period of about 100 years.

The sediments below 6 cm demonstrated background polonium (^{210}Po) activities. The surface sediment horizon from 0 to 1 cm, dated at 1976 A.D. This and the other dates are an average of the accumulation for each 1 cm horizon.

The stratigraphies of the major species identified in this core (Fig. 13) showed little change in floristic composition through time (with depth). Lake 219 was one of the few lakes to contain frequencies of the acidobiont Anomoeoneis serians (Breb. ex Kutz.) Cl. greater than 1 or 2 percent. Cluster analysis of the 18 dominant taxa divided the core into four groups of horizons (Fig. 15). The horizons in group I, 1 and 2, fell within the pH range of 5.4 to 5.6. All the other clusters are made up of groups of horizons with a pH range of 5.3 to 5.5, thus showing no change in lake acidity through the time period represented by the core. However, clusters I and II do tend to include many of the horizons from the top 10 cm of the core and clusters III and IV, those horizons occurring below the 10 cm level.

A lake water pH of 5.62 was recorded in August, 1984 and this is identical to the index B inferred pH value (5.6) calculated for the surface sediment sample from this core. The pH of lake 219 has been measured on five occasions, from August 1981 to October 1986, with the pH ranging from 5.62 to 6.18 ($\bar{x}$ = 5.96). The mean lake water pH for the period of record is considerably higher than the pH recorded in the during paleolimnological study (5.62) and the diatom inferred pH for the surface sediment (5.6).

There has been little change in the proportion of diatoms in each of the pH tolerance groups in the 50 cm long Lake 219 sediment core (Fig. 14). The top 5 cm of sediment did exhibit a slight increase in circumneutral taxa and corresponding decrease in acidophilous and acidobiontic species. The inferred pH profile demonstrated considerable variability and no distinct trends in pH over the top 20 cm of core. In the top 5 cm, a pH decline from 5.6 (horizon 5, circa 1895) to 5.4 (horizon 2, circa 1963) is evident followed by a rise to pH 5.6 at the surface (circa 1976). The lowest inferred pH is 5.3 at horizon 45 cm. There is no significant relationship between inferred pH and depth of sediment strata ($r = -0.23$; $S_e = \pm 0.09$, $P = 0.331$, $n = 20$) for the length of the core (Table 3).

LAKE 660

The core collected from Lake 660 was 60 cm in length (sectioned length) and was composed of mid-brown sediment that dried to a light grey-brown color. The water content of the core varied from 83 to 98%. A sediment accumulation rate of 12.6 mg cm^{-2} y^{-1} ($r^2 = 0.88$) was obtained for the top 9 sections of the core which represented about a 65 year period. The surface sediment horizon (0-1 cm) dated at 1983 A.D.

An average of 52 taxa were identified in each horizon and between 500 and 600 valves were enumerated for each sample. The occurrence of major taxa in relation to depth is presented in Fig. 16. The principal alkalibiont found in the upper section of this core was Pinnularia viridis v. commutata (Grun.) Cl.. A very high frequency of occurrence of the acidobiont, Semiorbis hemicyclus, was also noted throughout this profile.

Cluster analysis of the floristic data divided the horizons into 3 main clusters, representing the pH ranges 5.6-5.8 (Cluster I), 5.6-5.7 (Cluster II) and 5.5-5.9 (Cluster III), respectively (Fig. 18). The sediment horizons did not cluster in chronologically similar groups. It should be noted that horizons from both the top and bottom of the core occur in group III. These are horizons that include higher proportions of acidobiontic and acidophilous taxa.

The inferred pH value calculated for the surface sediment (circa 1983) was 5.7 which is also the pH measured for the lake water in July, 1983. A lower pH value of 5.46 was recorded for the water of this lake in August, 1984. The mean pH for Lake 660, sampled four times between July 1983 and October 1984, was 5.60 (5.46-5.71). The diatom stratigraphies demonstrate an increase in circumneutral, alkaliphilous and alkalibiontic taxa, however, there is a reversal of this trend in the top 2-3 cm (~ circa 1966-1976) at which point the alkalibiontic taxa disappear and notable increases in acidobionts occur (Fig. 17). Correspondingly a decline in inferred pH from 5.9 to 5.7 (0.2 pH units) is apparent. The maximum inferred pH difference is 0.4 of a unit, increasing from pH 5.5 (horizons 60 and 50) to pH 5.9 (horizons 6 and 3). The relationship between inferred pH and depth throughout the core is significant ($r = -0.70$, $S_e = \pm 0.09$, $P = 0.001$, $n = 20$) (Table 3). The negative correlation coefficient (r) indicates an overall increase in pH from the bottom to the surface of the core, which is particularly evident from horizon 18 to horizon 16.

STEPHENSONS POND (LAKE 206)

The Stephensons Pond core was 60 cm long (sectioned length) and had a water content of between 94% and 99%. In this core, only the top two horizons had polonium (^{210}Po) activity above background levels. A dry sediment accumulation rate of 0.44 mg cm^{-2} yr^{-1} can be derived for this lake from dating of the top two horizons. The surface sediment horizon dated circa 1961 and the second horizon circa 1938. This accumulation rate is extrapolated to strata lower in the sediment core, with the top ten strata covering a time period of about 900 years

An average of 60 taxa were identified in diatom counts of between 400 and 600 valves for each horizon in the Stephensons Pond core. From the bottom of the core (60 cm horizon) to the 7 cm horizon, the floristic composition of the core is fairly stable (Fig. 19). Above sediment horizon 7 cm there is a decline in the frequency of Cyclotella compta (Ehr.) Kütz. This and other alkaliphilous taxa are replaced by increases in the typical acidic species, Anomoeoneis serians v. brachysira (Breb. Kutz.) Hust., Eunotia species, Frustulia rhomboides v. saxonica (Rabh.) De T. and Navicula subtilissima Cl.. The circumneutral species Navicula radiosa Kutz. also increased in frequency during the upper 7 horizons of this core.

A lake pH of 5.8 was recorded for Stephensons Pond in June 1984 (DOE unpublished data), while a value of 5.74 was recorded in August 1984 at the time of core collection, both of which compare well with a surface sediment inferred pH value of 5.9 obtained for this core. Stephensons Pond has been sampled a total of 8 times (September 1981 to September 1985) and the lake pH ranged from 5.62 to 6.26 ($\bar{x}$ = 5.95). The reconstructed pH profile for Stephenson's Pond suggests that there has been a slight increase in acidity (approx. 0.2 pH units) throughout the time period covered by the core (Fig. 20). The proportion of alkaliphilous taxa decreases towards the top of the core, most notably in the top 7 horizons. An increased abundance of species typical of more acidic waters also occurred in this top 7 cm of core (an extrapolated time span of 570 years). Cluster analysis of the diatom floristic data did not produce any meaningful cluster groupings in relation to inferred pH or by chronology (Fig. 21). The maximum inferred pH difference (decrease) is 0.3 of a unit from pH 6.1 (at horizons 50 and 21) to pH 5.8 (at horizons 7, 4, and 2). Both the inferred pH history in recent strata, and contemporary water chemistry, suggest little pH change. The regression of inferred pH versus depth was significant at $P < 0.05$ ($r = 0.55$, $S_e = \pm 0.07$, $P = 0.011$, $n = 20$) (Table 3), but not at $P < 0.01$.

AIDES POND

The Aides Pond sediment core was 60 cm in length (sectioned length) and had a water content that varied between 89 and 92%. A good sediment accumulation rate (8.43 mg cm^{-2} yr^{-1}, $r^2 = 0.98$) was obtained for the top 7 horizons of this core, representing approximately 50 yrs of sediment accumulation. The surface sediment horizon from 0-1 cm, dated at 1980 A.D., however, this and the other dates are averages for the accumulation of the whole 1 cm section.

An average of 60 taxa were identified in each horizon of the Aides Lake sediment core and between 500 and 800 diatom valves were identified and enumerated in each sample. The diatom flora of Aides Lake has been dominated by two species Tabellaria flocculosa and Cyclobella compta which, when combined, account for between 50% and 60% of each sample stratum (Fig. 22). Much of the increase in pH noted in the upper 7 cm of the inferred pH profile is accompanied by high frequencies of the alkaliphilous C. compta and reduced numbers of T. flocculosa, an acidophil.

A pH of 6.3 was measured for Aides Pond in June, 1984 (DOE unpublished data), which is comparable with an inferred pH value of 6.4 calculated for the surface sediment sample for the core. A lake pH of 6.02 (DFO data) was measured at the time of core collection (Table 2).

The inferred pH profile developed for Aides Lake can be divided into three main zones (Fig. 22). The first of these zones, including the sediment horizons from 60 cm to 30 cm, covers a time period during which the inferred pH increased by 0.2 units. It is evident from the histograms of pH tolerance groups that increases in pH in zone 1 is a consequence of increases in the proportion of alkaliphious diatoms and coincident decreases in circumneutral taxa. The proportion of acidophilous diatoms remained fairly constant through this zone. In the second zone of the Aides Pond core, from 25 cm to 8 cm, the inferred pH fluctuated around a value of 6.2. A decrease in pH occurred from the 10 cm horizon (extrapolated date circa 1913) to a depth of 8 cm (extrapolated date circa 1931) by which time a notable decrease in alkaliphilous taxa (20%) had occurred. There is an abrupt increase in inferred pH (0.3 pH units) from horizon 8 cm to the 7 cm horizon and the start of the final zone. The inferred pH of zone 3 (circa 1936 to circa 1980), fluctuates from 6.2 to 6.4 pH units. There is a marked increase in the proportion of alkaliphilous taxa and decrease in acidophilous diatoms from sediment horizon 8 cm to horizon 7 cm, a floristic change which is sustained throughout the remainder of the core. A cluster analysis of the diatom floristic data, however, did not produce any meaningful cluster groupings in relation to sediment horizon pH (Fig. 24). The maximum inferred pH difference (increase) is 0.5 of a pH unit from pH 5.9 (horizons 8) to pH 6.4 at the surface. The relationship between inferred pH and strata depth, as determined by linear regression, was not significant ($r = -0.38$, $S_e = \pm 0.11$, $P = 0.096$, $N = 20$) (Table 3).

DISCUSSION

Historical pH profiles have been developed for the seven study lakes using the Index B transfer function calculated in Part 1 of this report series. The strengths and limitations of the index approach to inferring pH histories have been evaluated, and the index B transfer function for insular Newfoundland compares favourably to equations developed from other geographical locales. Using a regional index B transfer function to assess acidity changes in insular Newfoundland should therefore be a valid approach and permit comparisons to other diatom inferred lake acidification studies in Northern Europe and North America. Some of the weaknesses in the index methods are associated with occasional misclassification of diatoms as to pH preference, and taxonomic difficulties in some of the key pH indicator groups (Battarbee 1984). Davis and Anderson (1985) and Charles (1985) have demonstrated the ability to develop more refined transfer functions by multiple regression, principal components analysis, and other ordination techniques for application to inferring acidity chronologies. Charles (1985) is also convinced of the need to use regional contemporary pH-diatom occurrence data to assign pH tolerance groupings because of the apparent regional variations in species pH preferences and also because of some errors in the literature. Similar approaches are being examined for the insular Newfoundland diatom/contemporary lake pH data and will be discussed in a separate publication.

Unfortunately, problems were experienced in obtaining chronologies (^{210}Pb dates) for three of the cores; those from lakes #8, #12, and #204. There are three possible scenarios that may account for the inability to date these cores. Firstly, the sediment accumulation rate for these lakes could be extremely low. For example, one of the successfully dated lakes, Stephensons Pond, had an exceedingly low sediment accumulation rate (0.44 mg cm^{-2} yr^{-1}) and only two strata had ^{210}Po activities above background. A second possibility is that the sedimentation rate for these ponds is reasonably high but the accumulating sediments are composed of material low in ^{210}Po content. While the organic content of the sediment strata was not determined, field observations of colour and texture suggested a very high organic content for the sediments of these undated lake cores. A third possibility is that the sediments are thoroughly mixed, through physical processes, bioturbation, and organic decomposition, resulting in the ^{210}Po content remaining essentially constant throughout the core length, or at least for the horizons where dating was attempted. An analysis of the cation and metal (Pb, Zn, Fe, Al, Mn, As, Ti, Ca, Mg, Na, K, Cu, Hg, Ca) profiles for the five DFO lakes that were cored may provide some insight into the problems experienced in obtaining chronologies by ^{210}Pb dating (to be evaluated in Part 3 of this report series).

In evaluating the Index B inferred pH profile for Lake 8, it is apparent that the inferred pH value for the surface sediment was identical to the measured value for the lake water in August, 1984. This would suggest that, although it was not possible to date this core, the surface sediments are intact. The evidence presented for Lake 8 indicates that, although the lake has been acidic for the entire length of the core, the pH has decreased through time, with a pronounced increase in acidity starting at about horizon 9 cm to 12 cm and continuing to the surface. There is a corresponding and fairly steady increase in the proportion of acidobionts over the top 9-11 cm.

It is possible that the increase in pH seen at the bottom of this core, from about pH 5.4 (horizon depth 30 cm) to pH 5.7 (horizon depth 20 cm), and the return to pH 5.4 by depth 15 cm, is the result of a forest fire in the Lake 8 watershed. Watershed disturbances such as fires and logging operations characteristically result in an increase in pH of runoff water from the deforested area (Viro, 1974; Dickman and Fortescue, 1984). The pH gradually decreases until a new equilibrium is established. The recovery phase from fires and logging is distinctive in a pH profile and can be separated from decreases in pH associated with acid rain (Dickman and Fortescue, 1984). Currently, the Lake 8 catchment is about 40% forested, consisting mostly of scrub spruce-fir trees with barrens-bog-heath accounting for the other 60%. Lake 8 seems to have entered a fairly stable phase at about horizon 15 cm to 9 cm, where acidification seems to have begun again. A decrease in pH of 0.3 units, from pH 5.5 at 15 cm to 5.2 at the surface, seems to have occurred in Lake 8, although a greater decrease (0.5 unit) from pH 5.8 to 5.3 has occurred over the last 12 cm. This latter phase, represented by horizons in cluster I on the dendrogram (Fig. 5), could be the result of the effects of acid precipitation. It is difficult to interpret the causes for these pH changes without a knowledge of the watershed history and the time frame associated with each stratum.

It is also noteworthy that Lake 8 represents one of the many lakes in this study in which a clear division of *Tabellaria flocculosa* and *T. quadriceptata* is of importance, owing to the different pH preferences of these two taxonomically similar species. Figure 3 reveals that *T. quadriceptata* is not a significant contributor to the acidobiontic totals.

For Lake 12, the inferred surface sediment pH (5.1) is 0.3 pH units lower than the 1984 lake water pH value (5.4). This could indicate that the sedimentation rate in this lake is so low that a 1 cm horizon incorporates many years of diatom accumulation and the surface horizon thus represents an average for a long time interval. The profile presented for Lake 12 (Fig. 7) shows little change in the diatom community or inferred pH through the majority of the core. However, the top 5 cm of core does show a decrease in inferred pH. An increase in circumneutral taxa, principally *Coscinodiscus lacustris* Grun., and a reduction in the frequency of alkaliphilous taxa has also occurred in this upper zone of the sediment core (Fig. 7). During the time period represented by the top 5 cm of core, there is also a decline in some acidophilous taxa, notably centrics, and a corresponding increase in the frequency of other acidophilous taxa including members of the family Fragilariaceae (notably Fragilaria virescens Ralfs and F. lata (Cleve-Euler) Renberg and Tabellaria flocculosa (Roth) Kütz. (Fig. 6). Lake 12 is highly colored (50 T.C.U.) and acidic, and might be expected to support populations of the acidobiont Asterionella ralfsii v. americana Korner., common to many of the study lakes, and not an increasing population of A. formosa Hass., an alkaliphil. Again it is difficult to speculate on the causes for this apparent pH decline in recent strata without a knowledge of the watershed history and the time frame associated with this change. The Lake 12 catchment is 100% forested and the trees in the spruce-fir forest of the neighbouring Cat Arm watershed have been aged at between 69 to 130 ($\bar{x}$ = 103) years old (Hunter and Associates 1980). This would suggest no major watershed disturbance over the last century.

Lake 204, a highly colored lake (50 T.C.U.), also has become increasingly acidic throughout the time period covered by the core. An initial decline of about 0.3 units (pH 5.8 to 5.5) occurred between horizons 60 and 20. A possible cause for the acidification trend in these lower, older strata might be progressive humification of the watershed. In the second zone of this core, 10-20 cm, the lake appears to have reached an equilibrium with fluctuation about pH 5.5. (range of 5.4-5.6). In the final phase, the pH declines from 5.6 (10 cm) to 5.3 (at the surface). In this phase there is an obvious decline in the acidophilous taxa (Cyclotella glomerata, Melosira distans for e.g.) coupled to a corresponding increase in the circumneutral (all Navicula radiosa for e.g.) and acidobiontic (Actinella punctata, Semiorbis hemicyclus and Tabellaria quadriceptata, for e.g.) taxa. The pH of Lake 204, recorded in August 1984 was 5.13 and the mean pH for this lake, from samples collected over a 4 year period (1981-85) was 5.21 (5.00-5.49). The decrease in pH in the recent strata (top 10 cm), possibly due to anthropogenic acidic deposition, is about 0.3-0.4 of a unit. Lake 204 has previously been identified as demonstrating acidification effects, on the basis of chemical interpretation, from both natural (organic) and anthropogenic (sulphate) sources (Scruton 1983). This evidence is supported by a clear acidification trend in the inferred pH profile both over the long term (60 cm of core length) and in recent strata (top 10 cm). The lake has demonstrated an alkalinity deficit from 31.9 to 50.1 μeq L^{-1} ($\bar{x}$ = 38.5), while excess sulphate values have varied from 15.8 to 59.8 μeq L^{-1} ($\bar{x}$ = 30.2). Recently, employing the Oliver et al. (1983) method of computing organic anions ($COOH^-$), it has been determined that the dominant anion in Lake 204 is organics.

The inferred pH profiles for Lake 204 using the SR and GMR transfer functions are quite similar in shape, and both demonstrate the clear trend to increasing acidity over the core length, while the GMR profile is shifted about 0.1 of a pH unit to the left (lower pH) for many of the strata (Fig. 11). As a consequence, the minimum inferred pH is lower (5.1, strata 6 and 7) and the maximum pH decline over the core length is greater (0.8, from strata 40 to strata 6 and 7) for the GMR inferred profile. This is consistent with the statistics of the GMR and SR "difference plots" for inferred versus measured pH. The SR "difference plot" had a slope of less than 1.0 (0.89) suggesting that, in the lower pH ranges, the inferred pH value was being slightly overestimated.

Unfortunately, attempts to date the Lake 204 core (using ^{210}Pb) were unsuccessful. A recent review of historical water chemistry (sulphate export) for rivers in Atlantic Canada has suggested that sulphate loading in the region was at a maximum in the early 1970s (1972 and 1973 in Newfoundland) and has declined since (to about 1983) (Thompson 1986; Howell and Brooksbank 1987). A similar temporal trend is also apparent in North American sulphur emissions and the decline from the 70s to the 80s has largely been ascribed to a down-turn in North American economies (Martin and Brydges 1986). If stratum 6 and/or 7 had dated at circa ~ 1970 then the inferred pH profile would have been consistent with apparent regional trends in acid loading. However, without a time scale on the pH profile, it is difficult to speculate on the slight pH recovery evident in the upper 6 cm of the Lake 204 core.

The sedimentation rate for Lake 219 was 3.78 mg cm^{-2} y^{-1} which roughly translates to a low accumulation rate of 0.04 cm yr^{-1} for the top 10 cm of the core. Extrapolating this accumulation rate to the 50 cm horizon, these sediments would have been deposited about 1,250 years ago. Data for each of the 1 cm horizons represents, on an average, a 25 year period. Evidence presented for the Lake 219 sediment core from (Fig. 14) suggests that little change in acidity has occurred in the lake since at least 1,200 B.P. Throughout this period, the pH of the lake has fluctuated around 5.5 and within the standard error established for the inferred pH data. In the top 5 cm there is an increase in circumneutral taxa with a slight decrease in acidobiontic taxa, but there is no clear pH trend in the top 5 cm (since circa 1895). The Index B inferred pH for the surface sediment (5.6) and lake pH recorded in August 1984 (5.62) are appreciably lower than the mean pH obtained from five sampling periods through August 1981 to October 1984 (5.96). The overall trend throughout the length of the core suggested a slight increase in pH with time, but this was not significant ($r = -0.23$, $p = 0.331$, $n = 20$).

For Lake 660, a sedimentation rate of 0.16 mg cm^{-2} yr^{-1} was obtained for the top 10 cm of core, yielding a much higher accumulation rate than Lake 219. A 1 cm horizon from the Lake 660 sediment core represents an average accumulation of 6 years of sediment. From an extrapolation of this sedimentation rate to the lowest section of the core, it is estimated that the 50 cm horizon is composed of sediments dating from 375 B.P.

The pH of Lake 660 seems to have remained close to pH 5.6 from around circa 1600 A.D. (horizon 60 cm) to just prior to circa 1913 A.D. (horizon 12 cm) at which time Lake 660 started to gradually increase in pH (Fig. 17). The increase in pH is similar to that discussed for Lake 8 and may have resulted from a period of deforestation in the watershed. A return to more acidic conditions in Lake 660 began at about circa 1946 A.D. (horizon 6 cm) and continued right up to the surface sediment horizon (circa 1983). This represents a decline in pH of 0.2 pH units from horizon 6 (circa 1946) to the pH value of 5.7 established for the surface sediment sample (circa 1983) and for the lake water (July 1983). A pH of 5.54 was recorded in the paleolimnological survey of August, 1984 and the mean pH for this lake (n = 5) is 5.66. A more detailed analysis of the diatom data, in conjunction with chemical data, is required to determine if the acidification recorded for Lake 660 represents a natural return to an equilibrium after a disturbance in the watershed, or if the decline in pH in recent strata and in the lake is, at least in part, a response to acidic deposition.

Stephensons Pond had an extremely low rate of sedimentation, 0.044 mg cm^{-2} yr^{-1} for the top 2 cm of sediment, giving the second horizon a mean date of circa 1939. Consequently, the past 50 yrs the period during which much of the anthropogenic acidic deposition has occurred, is represented by only the top two strata in the historical pH profile (Fig. 20). The inferred pH for horizon 2, circa 1939, is 5.8 and a value of pH 5.9 is calculated for horizon 1, circa 1961. It must be remembered each horizon represents an average for a 25 year period so it is not possible to detect changes within these broad time periods. However, the inferred pH values for the top two horizons are close to contemporary water pH (5.8 measured in August 1984) so there is no evidence of progressing acidification in this lake.

Extrapolating the sedimentation rate to the lower horizons of the Stephensons Pond core, it is evident that the historical pH profile for the lake covers a time period as long as 5,000 yrs. Throughout this time period Stephensons Pond has shown a trend towards increasing acidity but by a maximum of only 0.3 pH units. The increase in pH noted at horizon 20 could be the result of a forest fire and resultant loss of tree cover in the watershed, although it is not possible to see the characteristic fire signature in the pH profile due to the long time periods between inferred pH values. The Stephensons Pond watershed presently is largely non-forested (~95%) dominated by bog heathland with some scrub spruce-fir trees. One possible scenario for the small acidification trend over the long-term (5000 yrs) could be progressing humification since the last period of glaciation, as suggested by Renberg and Hellberg (1982) for Swedish lakes. Alternatively, progressive shallowing and extension of the littoral zone in Stephensons Pond is another possible explanation as many acidophilous taxa occupy littoral habitats (Vaughan et al. 1982, 1986). Species shifts may demonstrate an apparent chemical change when in effect the response is to changing lake morphometry.

There is "noise" observed in the upper 8 cm of this pH profile which may be due, in part, to a possible mis-classification of Semiorbis hemicyclus as discussed in Part I of this report series (Scruton and Elner 1986). In addition much of the Stephensons Pond sediment core had higher than average amounts of broken frustules, which will contribute to the apparent variations between samples.

For the Aides Pond core, a sedimentation rate of 9.32 mg cm^{-2} yr^{-1} was obtained for the top 7 cm, which translates to about 0.16 cm of accumulation per year. Each 1 cm horizon thus represents, on an average, approximately a 6 year period. Extrapolating the sedimentation rate of 0.16 cm yr^{-1} to the zone 1 section (0-30 cm) of sediment, it is estimated that the first 30 cm of core represents a time period in excess of 180 yrs.

The inferred pH profile for Aides Pond (Fig. 23) can be divided into three distinct zones. In the first of these zones (horizon depths 60 cm-30 cm), there is evidence that the lake underwent a period of gradually increasing pH. The major floristic change during this time was an increase in *Cyclotella compta*, an alkaliphil, towards the 30 cm horizon. Zone 2 (horizons 25-8 cm) has lower inferred pH values than the other two zones of the core. There is an apparent increase in pH of 0.3 pH units between horizons 8 and 7. If the sedimentation rate of 0.16 cm yr^{-1} is extrapolated to horizon 8 cm, the apparent increase in pH between horizons 8 and 7 occurred during a period of only 6 years. The third zone of the core (top 7 cm) represents a period of gradually increasing pH (top 4 strata) to a contemporary (inferred) pH of 6.4. This zone is characterized by an increase in the proportion of alkaliphilous taxa (*Cyclotella compta* in particular) with a concurrent decline in acidophilous taxa (*Tabellaria flocoulosa* in particular). Discontinuity was noted between horizons 7 and 8 in the supported ^{210}Po profile. The concentration of ^{210}Po at horizon 7 was 8.4 $DPMg^{-1}$ and had reduced to a background level of 1.2 $DPMg^{-1}$ at horizon 8 cm. The evidence suggests that a section of sediment below horizon 7 cm has resulted from slumping or other sediment movement and may therefore represent an erroneous segment in the core chronology. However, the ^{210}Pb profile indicates that the top 7 cm of the core have been undisturbed.

The 7 cm horizon has been dated (circa 1936) and this and the upper section of core covers the era of increases in anthropogenic airborne pollution in North America (Bangay and Riordan 1983). The Aides Pond core shows no historical evidence of recent increases in acidity of the lake due to acid precipitation. The inferred pH profile for this lake demonstrates an increase in pH in recent years to approach the value measured for lake water in 1984.

Evidence of recent lake acidification from inferred pH profiles is presented for three, possibly four, of the seven study lakes (Lakes 8, 12, 204, and possibly 660). The inferred pH decline in these four lakes is from 0.2 to 0.3 of a unit, and contemporary water chemistry for Lakes 12 and 204 suggests that further declines in pH may have occurred in these two lakes. In Lakes 12 and 204 (and the other lakes) the surface sediment strata represent an average of many years of sediment accumulation (6-25 years in some cases) which may obscure recent pH trends. Lake 660, although demonstrating no significant pH trend throughout the core length, does demonstrate a pH decline of 0.2 pH unit over the last 6 cm of the core (circa 1946), and this trend is supported by a lower contemporary water pH in recent years as compared to the inferred pH in the surface stratum. Stephensons Pond demonstrates a significant pH decline throughout the length of the core ($r = 0.55$, $p = 0.011$, $n = 20$), although there is no definitive trend in the recent sediment horizons (top 8-12 cm). There is no apparent trend in the Lake 219 core while Aides Pond demonstrates a trend to increasing pH, particularly over the top 10 cm (70+ years).

There is ample evidence in the literature of fossil diatom inferred pH profiles being used to demonstrate long-term and recent lake acidification. A number of these examples are presented in Table 4. Berge (1975) investigated fossil diatoms in a core from Langtern in Norway covering approximately 800 years. Diatom analysis suggested the lake was highly acidic (pH $\sim$ 5.0) throughout the entire core. Berge (1979) presented clearer evidence of lake acidification in 5 lakes from Agden and Hordaland Counties in southern Norway. In Övre Malmesvatn a pH decline of 0.6 of a unit (from ~ 5.2 to 4.6) was evident in the top 6 cm of the core (from 1927-37 to 1977). Berge, however, used the index ω of Nygaard (1956) which appeared to produce inaccurate pH values.

Renberg and Hellberg (1982) examined the post glacial pH history of two Swedish lakes (Gardsjon and Harsvalten) using their own newly developed index, index B. The diatom inferred pH's indicated a long-term natural acidification from pH 7.0 to 6.0 over the last 12,500 years, attributed to progressive oligotrophication, humification, soil weathering, etc. In recent years (since 1950), rapid pH declines of 1.5 unit (6.0-4.5) in Gardsjon and of 1.7 unit (5.9-4.2) in Harsvalten have been attributed to the era of anthropogenic acid deposition. A third lake, Lysevalten, demonstrated a similar rapid pH decline until this decline was reversed by liming in 1974.

The pH chronologies of three Finnish lakes were developed by Tolonen and Jaakkola (1983) using Index alpha (α) and B. The lakes demonstrated pH declines from 0.7 to 1.4 of a unit to contemporary pHs (inferred) ranging from 4.9 to 5.6. The onset of acidification in one lake (Hanklampi) was determined to be relatively recent, circa 1961-64.

Battarbee et al. (1985) used index B to infer the pH history of Loch Enoch in the Galloway Region of Scotland. The pH of this lake has declined from 5.20 (1840) to a contemporary (inferred) pH of 4.3, a change of 0.9 unit. Two other lochs in the region, Loch Grannoch and Round Loch of Glenhead, demonstrated pH declines of 0.9 to 1.0 unit from pH 5.5 to contemporary pHs at 4.5-4.6. The onset of acidification was 1910 for Loch Grannoch and 1850 for Round Loch of Glenhead (Flower and Battarbee 1983). A fourth lake in the Galloway Region (Loch Dee) has also demonstrated an Index B inferred pH decline of 0.5 unit (pH 6.1 to 5.6) with the onset of acidification about 1890 (Battarbee 1984).

Davis et al. (1983) have reconstructed the pH history of nine Norwegian and six New England (USA) lakes using multiple regression methods. Three very acidic (pH < 5.0) Norwegian lakes demonstrated pH declines from 0.5 to 0.8 of a unit to contemporary pH's of 4.6 to 4.8. The onset of acidification was determined to vary from 1890 to 1927. Four lakes with contemporary pHs from 5.0 to 5.7 demonstrated lesser to no change (0.0 to 0.3 pH unit decline) while two lakes with pHs greater than 5.7 demonstrated no acidification trend. The six New England lakes all demonstrated pH declines in the range of 0.2 to 0.4 unit to present day pH's of 4.7 to 5.9. This magnitude of pH change is very similar to that demonstrated by the four Newfoundland study lakes showing recent declines in pH. The onset of acidification in New England lakes was determined to be 1920-1940. Four additional New England lakes (Branch Pond, Mountain Pond, Unamed Pond, Cone Pond) have also been studied (Norton et al. 1985; Davis and Anderson, unpublished data as cited in Charles and Norton 1986; Ford 1985) and in only one lake (Branch Pond) was a significant pH change apparent (0.3 unit decrease with the onset of acidification about 1930). Both watershed disturbance and acid deposition have been suggested as the cause for the pH decline.

Del Prete and Schofield (1981) studied three Adirondack lakes, of which only one (Honnedaga) showed a significant pH decrease of 0.6 unit in the uppermost core strata. The authors speculated this decrease occurred at least 10 years prior to coring (1966 or earlier). Del Prete and Galloway (1983) studied diatom inferred pH changes in cores from 3 additional Adirondack lakes (Panther, Sagamore, Woods) and found slight pH decreases (0.3-0.5 unit) in two of the lakes. The authors caution that these changes are close to the standard error of the inferrence techniques. No dating of the cores was attempted. Charles (1984) analyzed a core from Big Moose Lake in the Adirondacks using a variety of methods (Index α and B, multiple regression). The lake demonstrated a diatom inferred pH decline of about 1.0 unit (from pH 5.7 to 4.7) over a 10-year period commencing about 1950. Charles (unpublished data as cited in Charles and Norton 1986) also examined four additional Adirondack lake cores (Deep Lake, Lake Arnold, Upper Wallace Pond, Little Echo Pond). An inferred pH change of about 0.7 unit was evident in Deep Lake with the onset of acidification in the 1940s to 1950s while an inferred change of 0.4 of a unit, commencing in the 1930s to 1940s, was detected in Upper Wallace Pond.

Dickman and Fortescue (1984) examined eight lakes north of Lake Superior in Ontario, and found the Index alpha (α) inferred pH change to range from 0.20 to 0.8 of a pH unit. This change, however, was the difference between inferred pH maximum and minimum over the length of each core and was not necessarily reflective of recent change due to anthropogenic acidification. Only one lake (Beaver Lake) had a statistically significant reduction in diatom inferred pH over the last 30 years (Dickman et al. 1985). Dickman et al. (1984) also reconstructed the pH of two other lakes in this region (Lakes B and CS), and found Index α inferred pH histories to demonstrate a decline of 1.0 pH unit (6.2-5.2) in Lake B over the last 20 years and a decline of 1.7 pH unit (7.1-5.2) in Lake CS over the last 30 years. Delorme (unpublished data as cited in Charles and Norton 1986) examined a core from Bachawana Lake (Turkey Lakes Watershed Study) and found no obvious trend in diatom inferred pH.

Vaughan et al. (1982) studied surficial diatoms in four lakes in the Halifax area of Nova Scotia collected over a 9 year period (1971 and 1980). These lakes all demonstrated similar shifts towards more acid tolerant diatom assemblages which is supportive of other studies demonstrating changes in lake chemistry (Watt et al. 1979) in response to atmospheric deposition of acids. However, in rural Nova Scotian lakes experiencing lower rates of acidic deposition, the chemical and biological responses were complex (Vaughan et al. 1986). In clear water rural lakes, acidification has proceeded at a slower rate, with changes observed over a 30-50 year period in clear rural lakes resembling changes observed in urban Halifax lakes over a ten year period (the 1970 decade). In rural brown water lakes, organic acids appear to dominate pH variation and the rate of atmospheric deposition is insufficient to dominate this trend. A comparison of diatom responses in rural and urban Nova Scotian lakes suggests the trend to more acid tolerant diatom assemblages and simplification of communities is not only dependent on pH but the rate of change as well. Elner and Ray (1987) examined diatom inferred pH changes in four New Brunswick lakes and four Nova Scotian lakes using Nygaard's (1956) Index alpha (α). Inferred pH declines of 0.2 and 0.8 of a unit were evident in two Nova Scotian lakes (Big Indian, Kinsac) commencing 1939 and 1917, respectively, while declines of 0.4 to 0.5 of a unit were evident in two New

Brunswick lakes (Emigrant, Lily). The onset of acidification in Emigrant Lake was determined to be 1912.

Battarbee (1984), in reviewing the bulk of the fossil diatom/lake acidification research to that date, has concluded that diatom assemblages can be confidently used to predict pH changes within a standard error of ±0.25 to ±0.5 of a pH unit. In this study the standard error of the Index B transfer function for insular Newfoundland is ±0.33 of a pH unit and the inferred pH decline for the three lakes showing a significant pH decline is mostly in the range of 0.2 to 0.3 of a pH unit. Consequently, the diatom inferred pH change is largely within the limits of accuracy for the methodology. In order to be confident that the inferred pH changes are realistic we will be attempting to refine the diatom assemblege/inferred pH relationships using principal components analysis (PCA), stepwise multiple regression (SWMR), etc. to develop transfer functions with a smaller standard error. In addition more lakes will be cored and pH profiles generated so that the "weight of evidence" can be used to support the validity of these slight pH declines.

Battarbee et al. (1985) have reviewed several alternative hypotheses to acidic precipitation as the cause for freshwater acidification of lakes in Galloway, Scotland Region including changing land use patterns, aforestation (tree plantations on moorlands), and long-term (post glacial) natural change. All alternative hypotheses were rejected and a strong case is made for deposition of acidic precipitation as the primary cause for lake acidification.

In insular Newfoundland a similar review of alternative hypotheses can be made. As all lakes selected for coring were remote and aircraft accessible only, the changing land use and aforestation hypothesis can be rejected immediately. In addition, three of the lakes (#204, 660, and Stephensons Pond) were characterized by no forest development in their catchment and, therefore, the influence of forest regeneration (succession or forest fires) on the paleolimnological record is not likely. There is no certainty, however, that these lake catchments were non-forested over the period covered in the paleolimnological record. The two lakes (#'s 8, 12) on the Northern Peninsula contained some forestation in their catchments (40%, 100%, respectively). Lake 12 is near the Cat Arm watershed where mature fir-spruce forests with trees of ages 69-130 years (mean age of 103) are predominant. It is unlikely there have been any changes in the Lake 12 catchment over the last century. Lake 8 is located more northerly and is characterized by dwarf scrub forest (40%) and barrens, bogs, and rock heath in the catchment. The vegetation in this catchment, and the eastern side of the Northern Peninsula north of Harbour Deep, is typical of tundra barrens. Lake 219, located on the eastern side of the south coast, is typified by softwood scrubs forest with bogs and some open barrens in the catchment. The forest cover in this area is discontinuous and it is unlikely that the Lake 219 catchment has been subjected to recent forest fires. The Lake 660 catchment is mostly open barrens with about 15% of scrub forests. Conversely, the trend to increasing pH in Aides Pond may be linked to logging activity in the watershed. Bowater (Nfld.), now Kruger Inc., have been harvesting pulpwood in the Upper Humber River watershed (Aides Pond drainage) for in excess of 70 years.

The pH profiles for Lakes 8 and 660 do suggest some watershed disturbance in the deeper older strata, 20-30 cm and 6-12 cm (circa 1913-1946) respectively, which may be characteristic of forest fires. Damman (1983) suggests that forest fires have played a major role in determining the landscape and vegetation cover in southern Newfoundland. Poor regeneration and marginal climatic conditions for tree growth has resulted in the region remaining essentially barren after fires. The dwarf shrub-heath vegetation that has replaced the forest cover has a thick raw humus surface horizon which helps contribute a natural acidity to the surface waters. The long-term trend to humification as indicated by Renberg and Hellberg (1982) may be accentuated by forest fires, which characteristically result in an initial increase in pH, followed by vegetation shifts to forms that encourage moisture retention and organic accumulation and decomposition.

The question of long-term changes can be addressed by looking at the deeper strata in the cores where sectioning was accomplished (at 5 cm intervals) down to 50 or 60 cm. The most recent glaciation in insular Newfoundland seems to have receded about 10,100 to 13,700 years ago (Rogerson 1983). For the successfully dated cores the 50-60 cm strata cover a time period of 1250, 375, 5000, and 400 years for Lakes 219, 660, Stephensons Pond and Aides Pond, respectively, and obviously do not cover the whole post glacial period. The older strata (30 cm and greater) in Lake 660 and Aides Pond demonstrate a trend to increasing pH, while in Lake 219 and Stephensons Pond, there is little apparent trend. For Lake 204, which is undated, the trend to increasing acidity throughout the core length is also evident in the older (30-60 cm) strata. Longer cores, covering the full post-glacial period, would be necessary to address long-term changes reflecting oligotrophication, humification, soil weathering, progressing littoralization, changing climatic conditions, etc.

Four lakes demonstrated pH declines in recent sediment strata, and unfortunately of these, only Lake 660 was successful dated by ^{210}Pb. A decline of 0.2 of a unit was evident over the top 6 cm of the Lake 660 core corresponding to a time period of about 40 years (onset circa 1946). Battarbee (1984) has reported on the wide range in dates for the onset of recent (anthropogenic) acidification for different countries and within countries. For example the onset of lake acidification has been dated at 1890 to 1930 in Norwegian lakes (Davis et al. 1983) and even earlier in Scottish lakes, at about 1850 to 1920 (Flower and Battarbee 1983). The chronology of lake acidification in Sweden Renberg and Hellberg 1982) and Finland (Tolonen and Jaakkola 1983) suggests the phenomenon to be a relatively recent event (~ 1950 to 1960).

In the northeastern United States, the onset of acidification has been dated from 1920 to 1940 in New England lakes (Davis et al. 1983) and in the 1930s to 1950s in the Adirondacks (Charles 1985; unpublished data as cited in Charles and Norton 1986). In Ontario, the onset of acidification from diatom inferred pH chronologies was determined to be in the 1950s and 1960s; 20 to 30 years prior to core collection (Dickman et al. 1984). In New Brunswick and Nova Scotia the onset of pH declines was determined to vary from 1912 to 1939.

CONCLUSIONS

The Index B inferred pH profiles for the seven study lakes suggest a recent pH decline of about 0.2 to 0.3 of a pH unit in three, possibly four, of the lakes (Figs. 25 and 26). In two of these lakes contemporary water chemistry suggests further declines in pH that are not evident in the paleolimnological record. The primary causes for these pH declines are postulated to be atmospheric acidic deposition in recent sediment strata and possibly watershed disturbances (most likely forest fires) in the older strata in two of the lakes. Progressing humification of lake catchments over the period covered by the paleoecological record of each core is also a possibility.

The slight pH declines evident in these lakes are supportive of other conclusions with respect to the extent of lake acidification in insular Newfoundland (Scruton 1983 1985; Scruton and Taylor 1987). Many of the region's lakes are extremely sensitive to potential damage from the long range transport of air pollutants and lakes are demonstrating the initial effects of acidification, that is loss of alkalinity with concurrent slight declines in mean pH (with some episodic excursions). Chronically acidic Newfoundland lakes tend to be dominated by organic acidity, although this and other studies have identified acidity trends and current pH levels that cannot be totally ascribed to natural acidity. There is a growing body of evidence (as summarized in Jones et al. 1986) to suggest that organic influenced systems may be more sensitive to anthropogenic acidification than clear-water systems, although the same may not be true for biological responses. The suggested target levels for reduction of sulphate deposition (to 20 kg ha^{-1} yr^{-1}) may not be sufficient to protect acidic brown water systems from progressing acidification.

Currently the relationships between contemporary lake water pH and preserved diatom assemblages for insular Newfoundland (as discussed in Part I of this report series) are being refined through the following:

- re-assignment of taxa to pH tolerance categories on the basis of occurrence in insular Newfoundland lakes (as opposed to basing the assignment on autoecological data presented in the literature).

- transfer functions are being developed (after the above reassignment) using index methods (Index B and Index alpha), multiple regression (MR) stepwise variable selection (SWVS), and principal components analysis (PCA).

- separate transfer functions are being developed for clear water and brown water lakes.

Historical pH profiles for five of the seven study lakes will be re-evaluated using the above refinements in transfer functions. This information will be discussed and presented in a separate publication.

RECOMMENDATIONS

An interpretation of the pH history in the study lakes is hampered by the inability to date (with ^{210}Pb analysis) three of the four lake cores demonstrating an acidification trend, which precludes putting a time scale on the inferred trends. Further, the low sedimentation rate obtained for the dated cores indicates that a 1 cm horizon can cover a wide time period (6-25 years), which further complicates an assessment of effects related to anthropogenic deposition (which is considered a relatively recent phenomenon in North America).

Further lake sediment coring studies (August 1986) have been conducted with a view to minimizing the above mentioned problems. Study lakes were pre-selected on the basis of basin and hydrological characteristics that suggested reasonable rates of sedimentation. A larger suite of lakes (11) was cored (and sectioned) and a ^{210}Pb chronology of all cores was attempted. The lake cores demonstrating adequate rates of sedimentation and good vertical distribution of ^{210}Pb were chosen for detailed downcore diatom and metals analysis. In addition, cores were sectioned at finer intervals (0.5 cm strata) to try to improve the resolution of the time scale associated with acidity changes. Recreation of historical pH profiles for these lakes will also incorporate the refined transfer functions currently under development.

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Table 1. Morphometric and physical characteristics of the 7 downcore lakes.

Lake No./Name	Latitude	Longitude	Lake surface area (ha)	Watershed area (ha)	Ratio (WA:LSA)	Lake order	Apparent maximum depth (m)	Secchi Transparency (m)	Elevation (m)	Distance from coast (km)	Water colour (TCU)
8	50°43'	56°25'	104	617	5.9	1	4.0	3.0	297.2	14.5	10
12	50°18'	53°36'	25	167	6.7	1	12.0	1.0	403.9	7.0	50
204	47°49'	58°23'	23	257	11.2	1	20.0	2.0	312.4	4.5	50
219	47°52'	55°22'	177	1,027	5.8	1	2.2	2.5	167.6	14.5	17.5
660	47°52'	56°32'	164	752	4.6	2	8.0	NA	266.7	27.5	15
Stephensons Pond (206)	47°54'	57°30'	120	874	7.3	2	5.0[a]	2.8	362.7	22.0	25
Aides Pond	44°25'	57°17'	2,280	46,535	20.4	5	50.0	NA	76.2	32.0	30

[a]Actual maximum depth as confirmed by bathymetric survey (DeGraaf and Chaput 1986).

NA - not available.

Table 2. Physical/chemical data for the 7 downcore lakes (collected August 14-21, 1984).

Lake No./Name	pH	Alk.	Cond.	Hard.	Colour	Turb.	Ca^{+2}	Mg^{+2}	Na^{+2}	K^{+}	Cl^{-}	SO_4^{-}	HCO_3^{-}	$COOH^{-}$	Al
8	5.34	14	14	39	10	0.31	20	19	61	4	59	23	17	30	17
12	5.13	5	14	36	50	0.38	19	18	48	4	48	19	6	48	14
204	5.13	0	16	50	50	0.36	32	19	65	1	54	31	0	46	15
219	5.62	11	19	64	175	0.32	39	26	87	6	85	27	13	42	9
660	5.46	11	14	42	15	0.41	26	16	57	3	56	19	13	31	7
Stephensons Pond (206)	5.64	11	12	45	25	0.31	29	16	52	3	42	10	13	37	15
Aides Pond	6.02	56	18	105	30	0.31	79	26	57	4	48	13	68	50	15

All data in $\mu eq\ L^{-1}$ except pH, colour (T.C.U.), turbidity (J.T.U.) and aluminum ($\mu g\ L^{-1}$).

Table 3. Regression equation and statistics for pH (index B inferred) versus core depth for the seven study lakes. Two regressions were computed including a simple linear regression (SR) and a multiplicative regression model (MR) using natural log of depth as the dependent variable.

	Regression type	Equation	r	S_e	P
Lake 8	SR	pH = 0.010 depth + 5.35	0.56	0.11	0.010[b]
	MR	pH = 0.017 depth + 5.26	0.61	0.02	0.004[b]
Lake 12	SR	pH = 3.53 depth + 5.58	0.31	0.08	0.190
	MR	pH = 0.009 depth + 5.53	0.53	0.01	0.015[a]
Lake 204	SR	pH = 0.11 depth + 5.30	0.87	0.12	0.001[c]
	MR	pH = 0.03 depth + 5.10	0.81	0.03	0.001[c]
Lake 219	SR	pH = -1.49 depth + 5.48	-0.23	0.09	0.331
	MR	pH = -0.004 depth + 5.53	-0.27	0.02	0.252
Lake 660	SR	pH = -0.004 depth + 5.78	-0.70	0.09	0.001[c]
	MR	pH = -0.012 depth + 5.87	-0.70	0.01	0.001[c]
Stephensons Pond	SR	pH = 0.003 depth + 5.89	0.55	0.07	0.011[a]
	MR	pH = 0.009 depth + 5.81	0.63	0.02	0.003[b]
Aides Pond	SR	pH = -0.003 depth + 6.24	-0.38	0.11	0.096
	MR	pH = -0.009 depth + 6.30	-0.55	0.02	0.013[a]

[a]Significant ($P < 0.05$).
[b]Highly significant ($P < 0.01$).
[c]Very highly significant ($P < 0.001$).

Table 4. Acidification history of lakes from northeastern Europe and North American as inferred by paleocological reconstruction using fossil diatoms (adapated from Battarbee 1984; Charles and Norton 1986).

Country/Region	Lake	Contemporary pH (observed)	Contemporary pH (diatom inferred)	Pre-acidification pH (diatom inferred)	pH change	Onset of acidification	Method of pH[a] reconstruction	Reference
Norway								
Buskerud	Langtjern	4.68-5.16	<5.5	4.3-6.2	-	-	Index ω	Berge 1975
West Agder	Øvre Malmesvatn	4.53	<5.2	4.4-6.5	0.5	1927-1937	Index ω	Berge 1979
Hordaland	Øvre Botnatjönn	4.74	<5.1	<5.0	-	-	Index ω	Berge 1979
Hordaland	Rödlivatn	6.23	5.2-6.9	5.1-6.8	-	-	Index ω	Berge 1979
East Agder	Högleivvatn	4.47	<6.3	4.7-6.6	-	-	Index ω	Berge 1979
East Agder	Risvatn	5.66	5.4-7.1	5.7-7.2	-	-	Index ω	Berge 1979
Near Bergen	Blavtn	5.1	5.1	5.2	0.1	1930	MR	Davis and Berge 1980
West Agder	Holmvatn	4.7	4.46-4.51	4.8-5.2	0.6	1927	MR	Davis et al. 1983
West Agder	Nedre Malmesvatn	4.6	4.9-5.3	4.1-4.5	0.8	1927	MR	Davis et al. 1983
West Agder	Hovvatn	4.4	3.9-4.4	4.8-5.1	0.6	1890	MR	Davis et al. 1983
West Agder	Dorsvatn	5.0	5.0	5.0	-	-	MR	Davis et al. 1983
West Agder	Brarvatn	5.2	5.2-5.3	5.3-6.1	0.3	1850	MR	Davis et al. 1983
West Agder	Botnavatn	5.7	5.7	5.9	0.2	1920	MR	Davis et al. 1983
West Agder	Opplejosvatn	5.8	5.8	5.8	-	-	MR	Davis et al. 1983
West Agder	Gronlivatn	6.5	6.5	6.5	-	-	MR	Davis et al. 1983
Sweden								
West Coast	Stora Skarsjön	4.5	4.5	6.0	1.5	-	Index ω	Almer et al. 1974
Bohuslän	Gardsjon	4.6	4.5	6.0	1.5	1950	Index B	Renberg and Hellberg 1982
Bohuslän	Harsvatten	4.4	4.2	5.9	1.7	1950	Index B	Renberg and Hellberg 1982

Table 4. (Cont'd.)

Country/Region	Lake	Contemporary pH (observed)	Contemporary pH (diatom inferred)	Pre-acidification pH (diatom inferred)	pH change	Onset of acidification	Method of pH[a] reconstruction	Reference
Finland								
N. Espoo	Hauklampi	4.75-4.90	5.4	6.0-6.4	1.0	1961-1964	Index α/B	Tolonen and Jaakkola 1983
N. Espoo	Orajärvi	4.70-4.80	4.9	6.3	1.4	1961-1964	Index B	Tolonen and Jaakkola 1983
N. Espoo	Häkläjärvi	4.88-5.10	5.6	6.3	0.7	1961-1964	Index B	Tolonen and Jaakkola 1983
Scotland								
Galloway	Round Loch of Glenhead	4.6 (4.5-5.0)	4.8	5.5	0.9	1850	Index B	Flower and Battarbee 1983
Galloway	Loch Grannoch	4.4-4.9	4.5	5.5	1.0	1910	Index B	Flower and Battarbee 1983
Galloway	Loch Enoch	4.4-4.7	4.3	5.2	0.9	1840	Index B	Battarbee et al. 1985
Galloway	Loch Dee	4.9-5.9	5.6	6.1	0.5	1890	Index B	Battarbee et al. 1985
United States								
New York (Adirondacks)	Honnedaga	4.7-4.8	5.2	6.1	0.9-1.1	1966	Index α	Del Prete and Shofield 1981
New York (Adirondacks)	Woodhull	5.1-5.3	6.0	6.1	-	-	Index α	Del Prete and Shofield 1981
New York (Adirondacks)	Seventh	6.4-7.0	6.5	6.6	-	-	Index α	Del Prete and Shofield 1981
New York (Adirondacks)	Big Moose P	4.7	4.7	5.7	1.0	1950	MR/Index α/B	Charles 1984
New York (Adirondacks)	Panther L.	6.2	6.1	6.4	-	-	Index α	Del Prete and Galloway 1983
New York (Adirondacks)	Sagamore L.	5.6	6.3	6.1	-	-	Index α	Del Prete and Galloway 1983

Table 4. (Cont'd.)

Country/Region	Lake	Contemporary pH (observed)	Contemporary pH (diatom inferred)	Pre-acidification pH (diatom inferred)	pH change	Onset of acidification	Method of pH[a] reconstruction	Reference
<u>United States</u>								
New York (Adirondacks)	Woods L.	4.7	4.8	5.2	0.3-0.5	-	Index α	Del Prete and Galloway 1983
New York (Adirondacks)	Deep L.	4.7	4.3	5.0	0.7	1940-1950	Index α/B;MR,TC	Charles (unpub. data as cited in Charles and Norton 1986)
New York (Adirondacks)	Lake Arnold	4.8	4.5	4.8	-	-	Index α/B;MR,TC	Charles (unpub. data as cited in Charles and Norton 1986)
New York (Adirondacks)	Upper Wallace P.	4.9-5.0	4.7	5.1	0.4	1930-1940	Index α/B;MR,TC	Charles (unpub. data as cited in Charles and Norton 1986)
New York (Adirondacks)	Little Echo P.	4.3	4.1-4.7	4.7	-	-	Index α/B;MR,TC	Charles (unpub. data as cited in Charles and Norton 1986)
New England (Vermont)	Branch P.	4.7	4.5	4.8	0.3	1930	Index α,MR,B PCA,TAX	Norton et al. 1985
New England (Maine)	Mountain P.	6.1	5.0	5.0-5.1	-	-	Index α,MR,B PCA,TAX	Norton et al. 1985
New England (Maine)	Unnamed P.	4.8	5.1	5.1	-	-	Index α,MR,B PCA,TAX	Norton et al. 1985
New England (N.H.)	Cone P.	4.5-4.8	4.6-4.7	4.5-4.8	-	-	Index ω,PCA,MR	Ford 1985
New England (N.H.)	Solitude P.	4.8	4.8	5.1	0.3	1920	MR	Davis et al. 1983
New England (Maine)	Speck P.	4.7	4.7-5.0	4.9-5.2	0.2	1920	MR	Davis et al. 1983
New England (Maine)	Ledge P.	4.5	4.5	4.9	0.4	1905	MR	Davis et al. 1983
New England (Maine)	Tumbledown P.	4.8	4.8	5.05	0.25	1970	MR	Davis et al. 1983
New England (Maine)	E. Chairback P.	5.0	5.0	5.2	0.2	1960	MR	Davis et al. 1983
New England (Maine)	Klondike P.	5.9	5.9	6.1	0.2	1945	MR	Davis et al. 1983

Table 4. (Cont'd.)

Country/Region	Lake	Contemporary pH (observed)	Contemporary pH (diatom inferred)	Pre-acidification pH (diatom inferred)	pH change	Onset of acidification	Method of pH[a] reconstruction	Reference
United States								
Colarado	Emerald L.	6.2	6.9	7.2	-	-	Index B	Beeson 1984
Colarado	Lake Halyaka	6.4	6.8	6.8	-	-	Index B	Beeson 1984
Colarado	Lake Husted	6.9	6.8	6.6	-	-	Index B	Beeson 1984
Colarado	Lake Louise	6.8	6.9	6.9	-	-	Index B	Beeson 1984
Canada								
Ontario	B	5.20	4.7	6.2	1.5	1962	Index α	Dickman et al. 1983
Ontario	Cs	5.20	6.4	7.1-7.3	0.8	1954	Index α	Dickman et al. 1983
Ontario	WW1	6.8	6.8	6.5	0.5[b]	-	Index α	Dickman et al. 1985
Ontario	WW2	6.7	6.8	6.5	0.5[b]	-	Index α	Dickman et al. 1985
Ontario	Fenton	7.5	7.2	7.2	0.5[b]	-	Index α	Dickman et al. 1985
Ontario	Logger	4.9-5.1	4.9	0.2	0.5[b]	-	Index α	Dickman et al. 1985
Ontario	Beaver	5.2-5.3	5.2	5.8	0.6[b]	1954	Index α	Dickman et al. 1985
Ontario	Doc Grieg	5.6	5.8	5.4	0.45[b]	-	Index α	Dickman et al. 1985
Ontario	Crozier	7.3-7.5	7.2	6.7	0.6[b]	-	Index α	Dickman et al. 1985
Ontario	Crawford	7.8	7.8	7.6	0.8[b]	-	Index α	Dickman et al. 1985
Ontario	Batchawana L.	6.1	6.2	6.0	-	-	Index α	Delorme et al. 1985
New Brunswick	Emigrant	6.1	6.1	6.5	0.4	1912	Index α	Elner and Ray (unpub. data) (cited in Charles and Norton 1986)

Table 4. (Cont'd.)

Country/Region	Lake	Contemporary pH (observed)	Contemporary pH (diatom inferred)	Pre-acidification pH (diatom inferred)	pH change	Onset of acidification	Method of pH[a] reconstruction	Reference
Canada (Cont'd.)								
New Brunswick	Tomoowa	5.3	6.4	6.4	-	-	Index α	Elner and Ray 1987
New Brunswick	St. Patrick's	6.7	6.8	6.8	-	-	Index α	Elner and Ray 1987
New Brunswick	Lily	5.6	5.7	6.2	0.5	-	Index α	Elner and Ray 1987
Nova Scotia	Big Indian	-	5.3	6.1	0.8	1939	Index α	Elner and Ray 1987
Nova Scotia	Kinsac	5.5	6.1	6.3	0.2	1917	Index α	Elner and Ray 1987
Nova Scotia	Round	4.6	6.2	6.2	-	-	Index α	Elner and Ray 1987
Newfoundland	Lake 8	5.34	5.3	5.5-5.6	0.2-0.3	-	Index B	This study
Newfoundland	Lake 12	5.13	5.4	5.7	0.3	-	Index B	This study
Newfoundland	Lake 204	5.13	5.3	5.6	0.3	-	Index B	This study
Newfoundland	Lake 219	5.62	5.6	-	-	-	Index B	This study
Newfoundland	Lake 606	5.46	5.7	5.9	0.2	1946	Index B	This study
Newfoundland	Stephenson's Pond (206)	5.64	5.9	-	-	-	Index B	This study
Newfoundland	Aldes Pond	6.02	6.4	-	-	-	Index B	This study

[a]Index α (Nygaard 1956).
Index ω (Nygaard (1956).
Index β (Renberg and Hellberg 1982).
MR - multiple regression of pH tolerance categories.
TC - multiple regression of floristic data after cluster analysis (taxa clusters).
PCA - multiple regression of principal component of floristic data.
TAX - multiple regression of common taxa.
[b]Difference between minimum/maximum pH in the core.

Table 5. A summary of acidification indicators determined from recent water chemistry data and inferred from fossil diatoms in lake sediments for the 7 study lakes.

	Water Quality Indicators				Fossil Diatom Inferred Indicators				
	Contemporary pH (Aug. 1984)	$\bar{x}$ pH (n)	$\bar{x}$ Alkalinity Deficit (μeqL^{-1})	$\bar{x}$ Excess sulphate (μeqL^{-1})	Contemporary Inferred pH (Index B)	Preacidification Inferred pH (Index B)	pH decline	Approximate onset of acidification	Possible cause of acidification
8	5.34	5.36 (2)	23.0	10.2	5.3	5.5-5.6	0.2-0.3	~10 cm	Acid deposition Watershed/forest disturbance (older strata)
12	5.13	5.02 (2)	36.6	28.7	5.4	5.7	0.3	~4 cm	Acid deposition
204	5.13	5.13 (6)	32.3	31.1	5.3	5.6	0.3	~7 cm	Acid deposition
219	5.62	5.96 (5)	42.7	18.9	5.6	-	-	-	-
660	5.46	5.60 (4)	29.5	15.1	5.7	5.9	0.2	~6 cm crica 1946	Acid Deposition Watershed/forst disturbance
Stephensons	5.64	5.95 (8)	20.5	26.7	5.9	-	-	-	-
Aldes	6.02	6.02 (1)	44.9	18.3	6.4	-	-	-	-

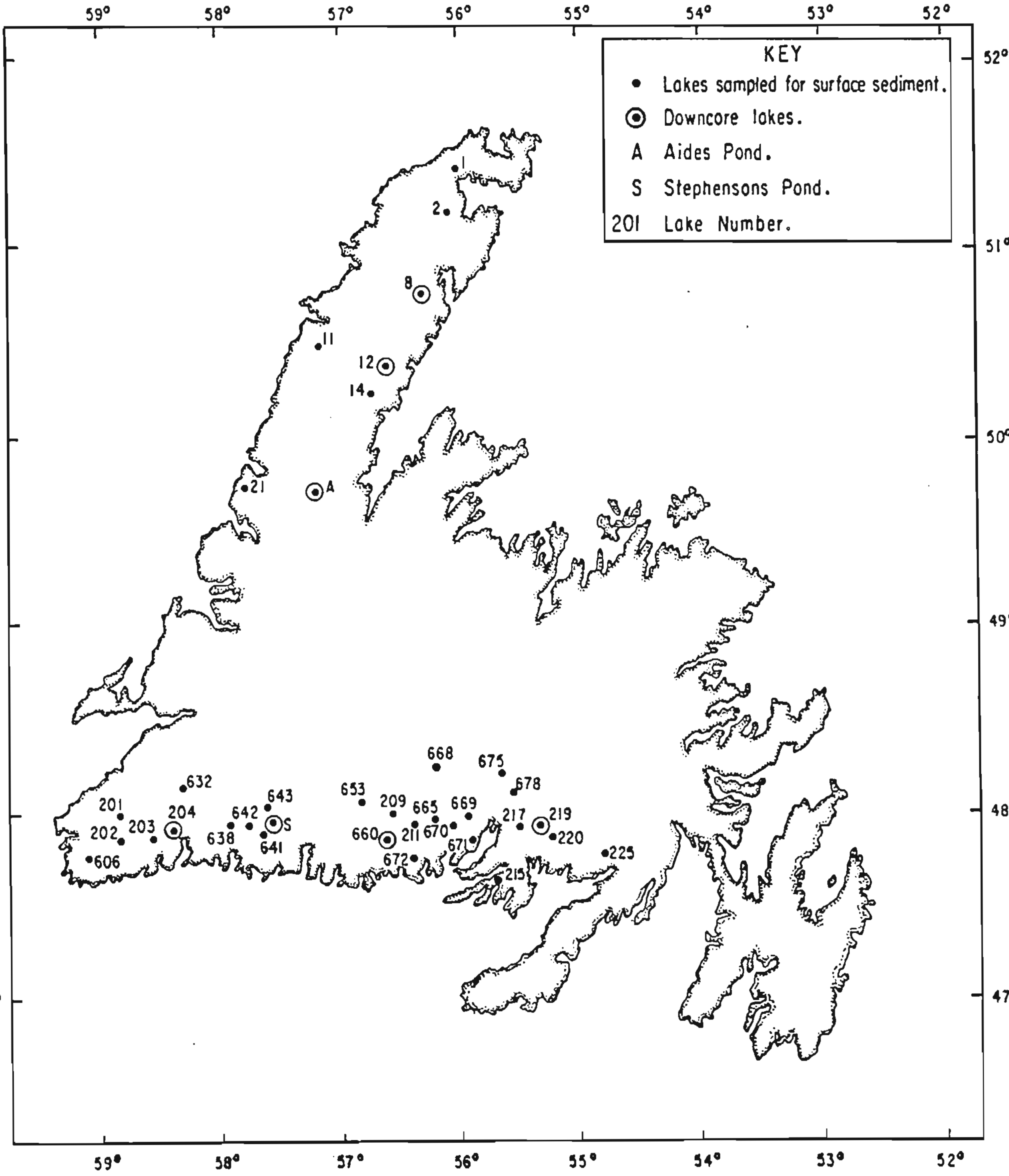

Fig. 1. Map of Insular Newfoundland showing lakes sampled (both downcore lakes and lakes sampled for surface sediments only) in the paleolimnological survey, August, 1984.

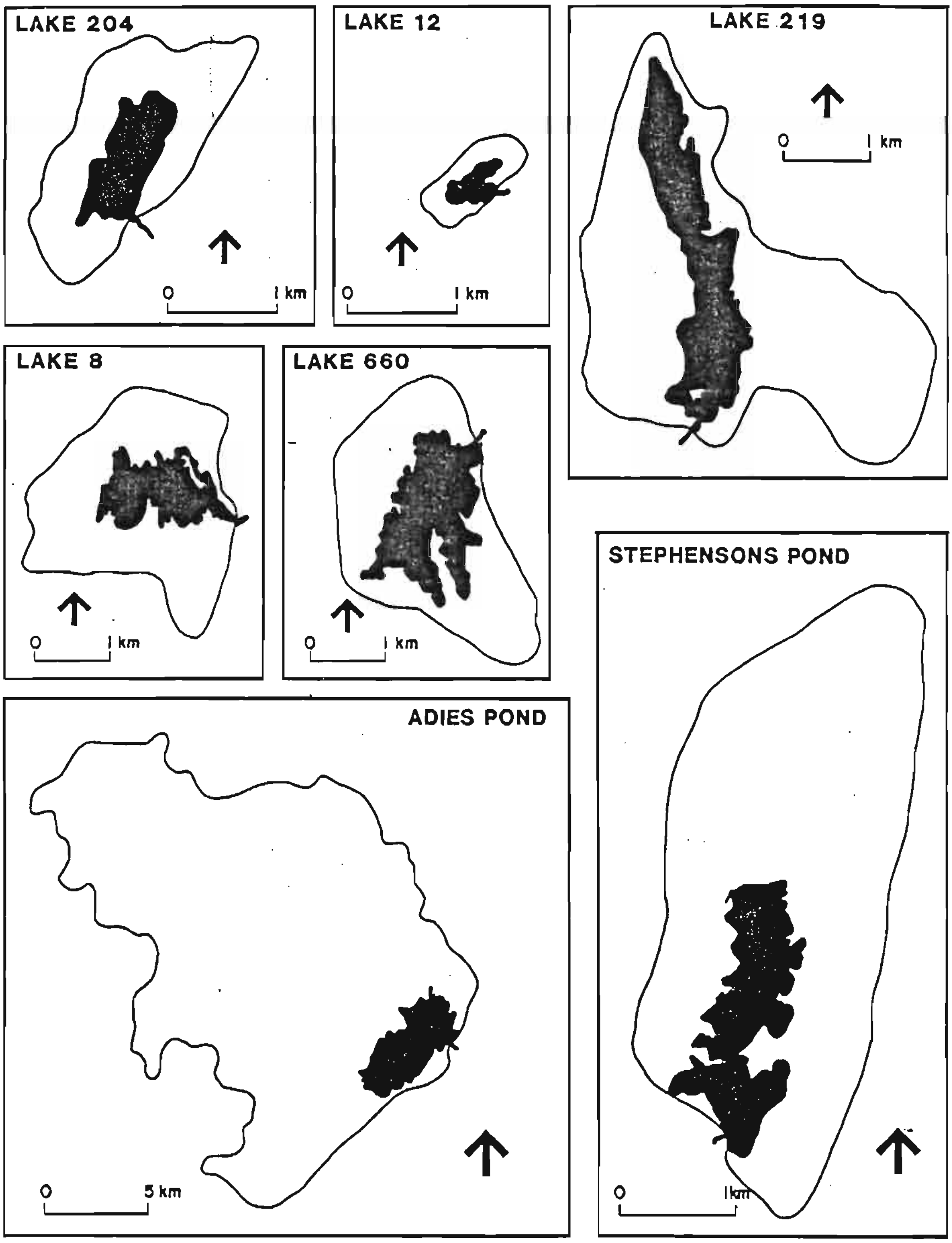

Fig. 2. General configuration and catchment area of lakes selected for downcoring.

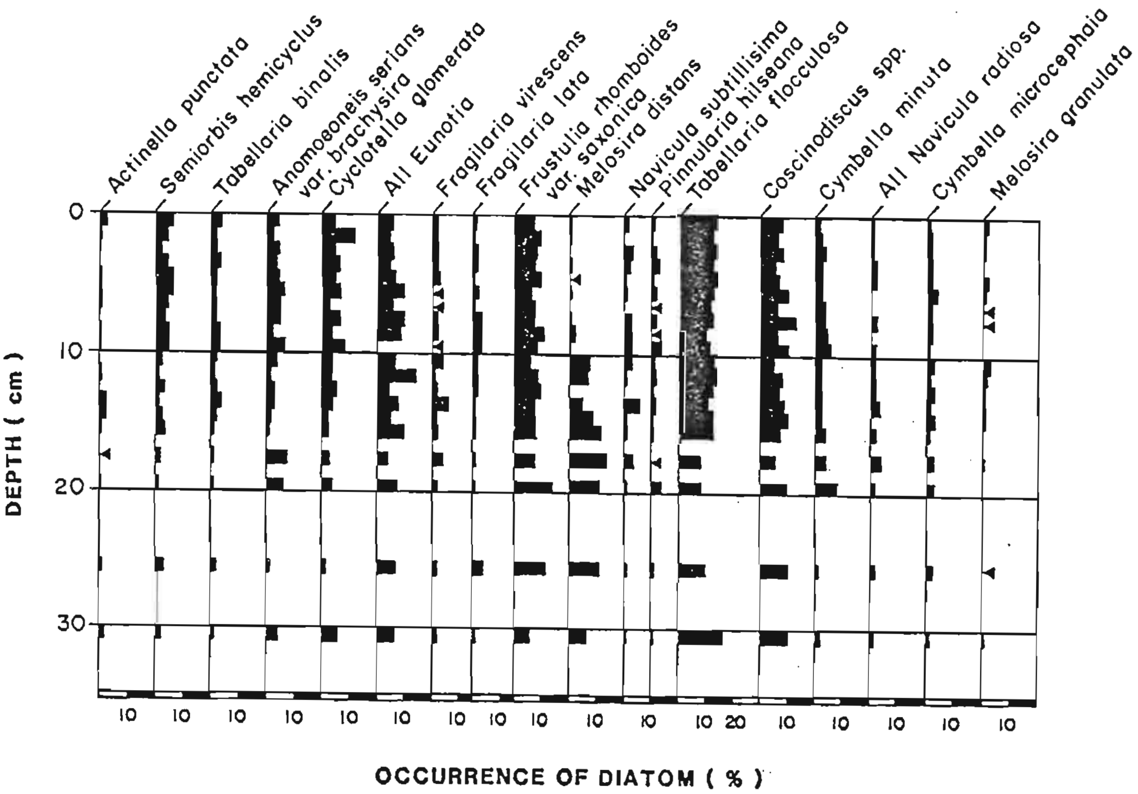

Fig. 3. Downcore profile of the frequencies (% occurrence) of selected key diatom taxa for lake 8.

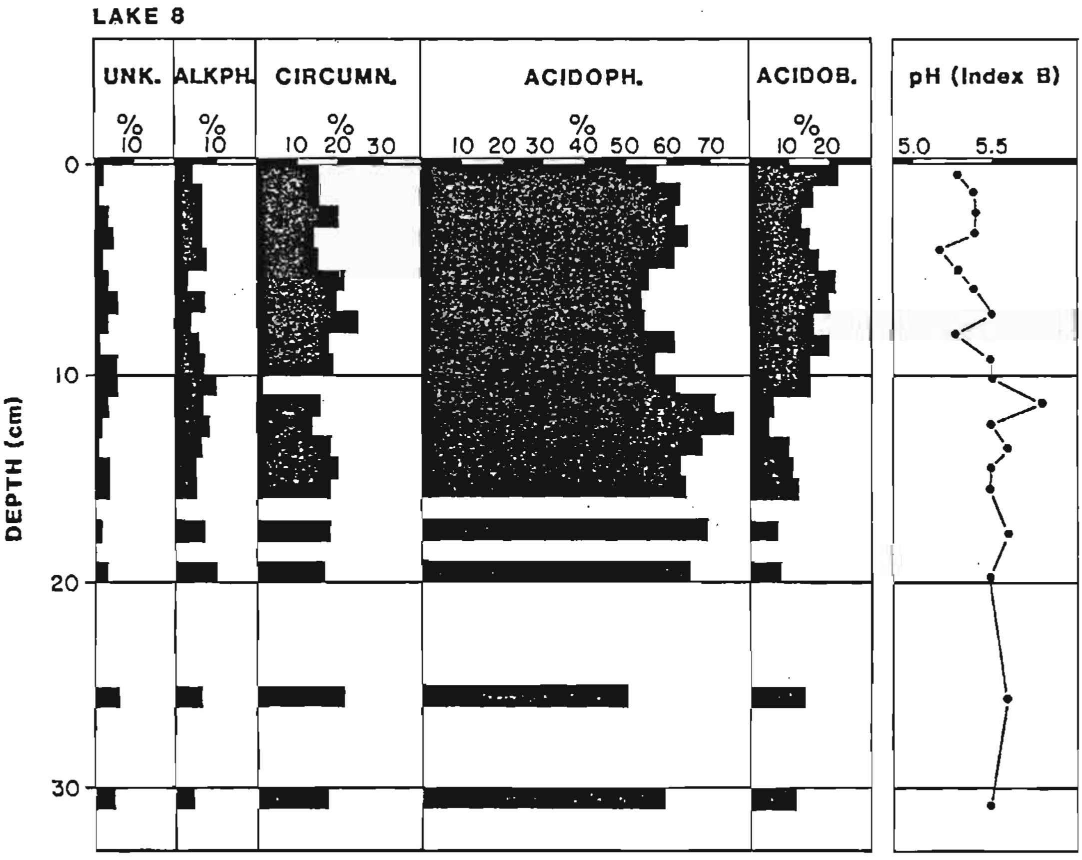

Fig. 4. Downcore profile of the frequencies (% occurrence) of diatoms in pH tolerance categories and Index B inferred pH for lake 8.

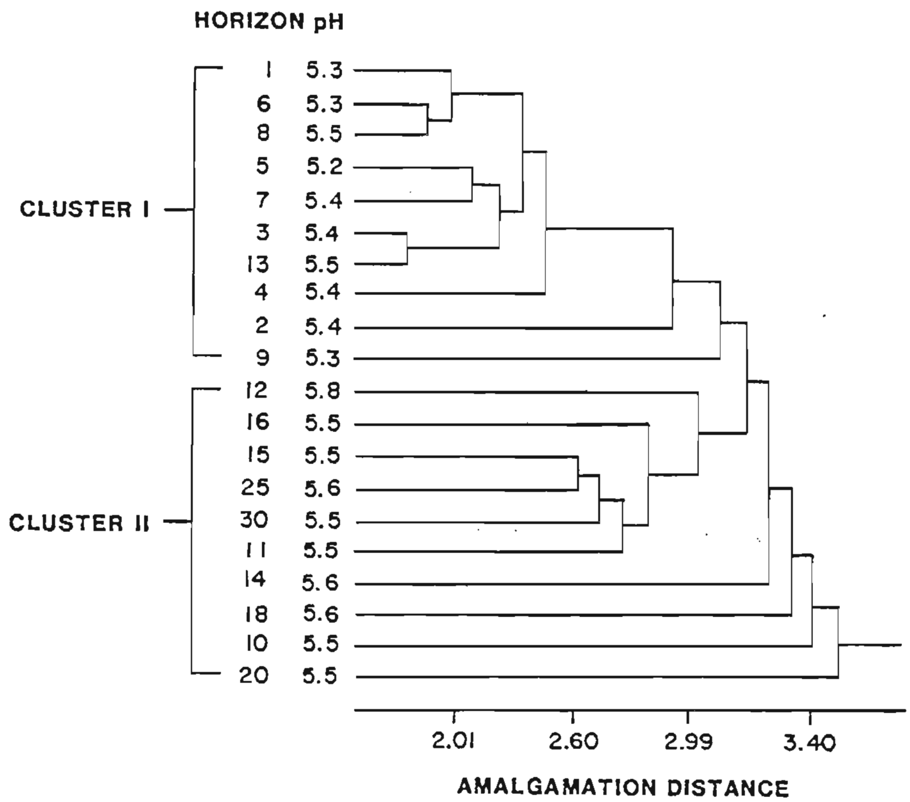

Fig. 5. Cluster analysis of selected diatom stratigraphies (% occurrence) for lake 8. pH is Index B inferred pH and the amalgamation distance is derived from a chi-square statistic.

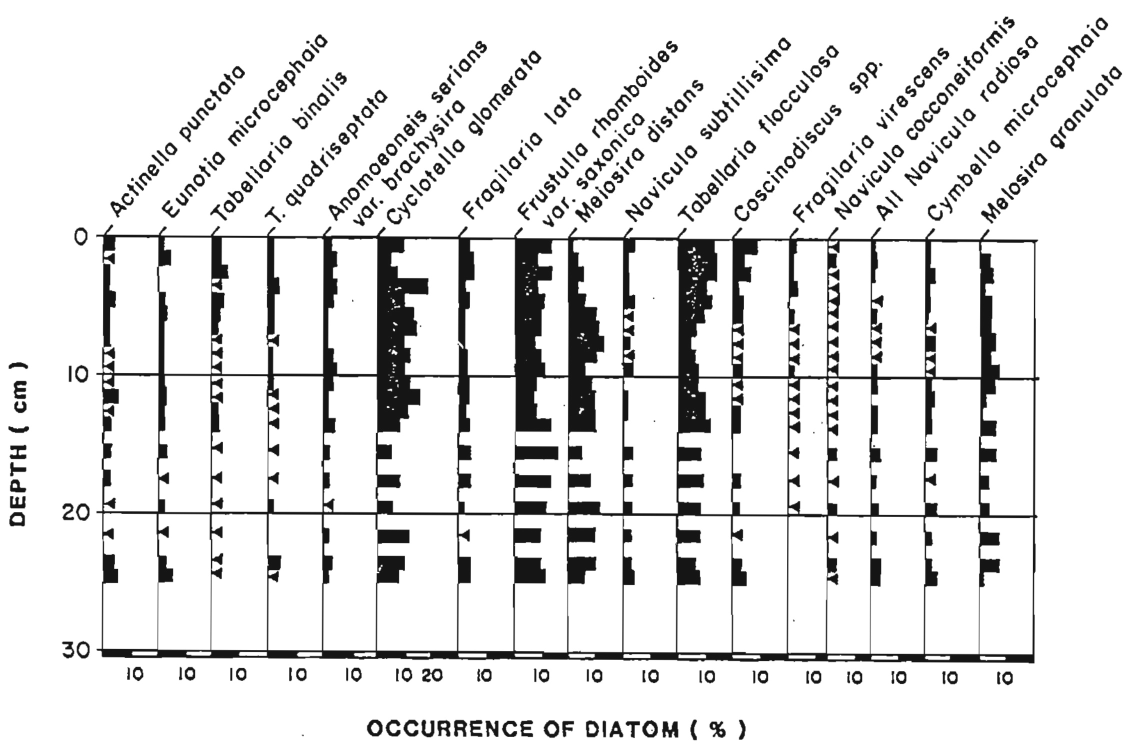

Fig. 6. Downcore profile of the frequencies (% occurrence) of selected key diatom taxa for lake 12.

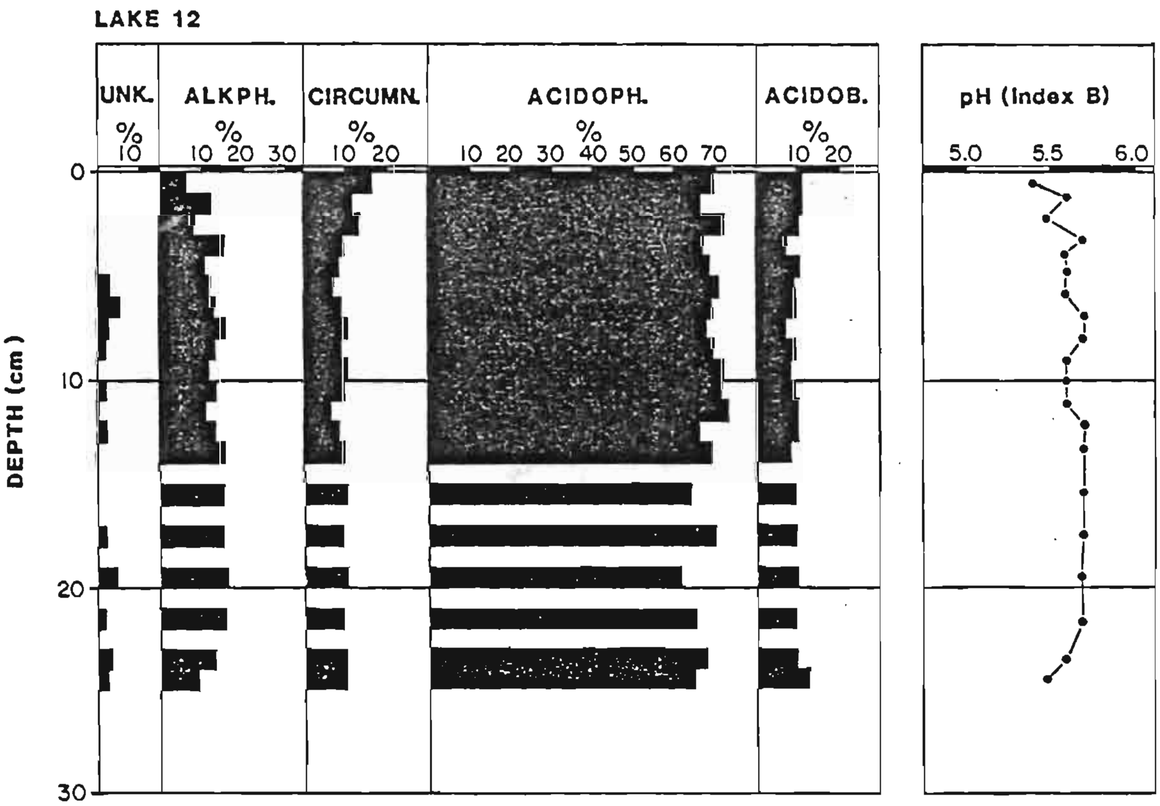

Fig. 7. Downcore profile of the frequencies (% occurrence) of diatoms in pH tolerance categories and Index B inferred pH for lake 12.

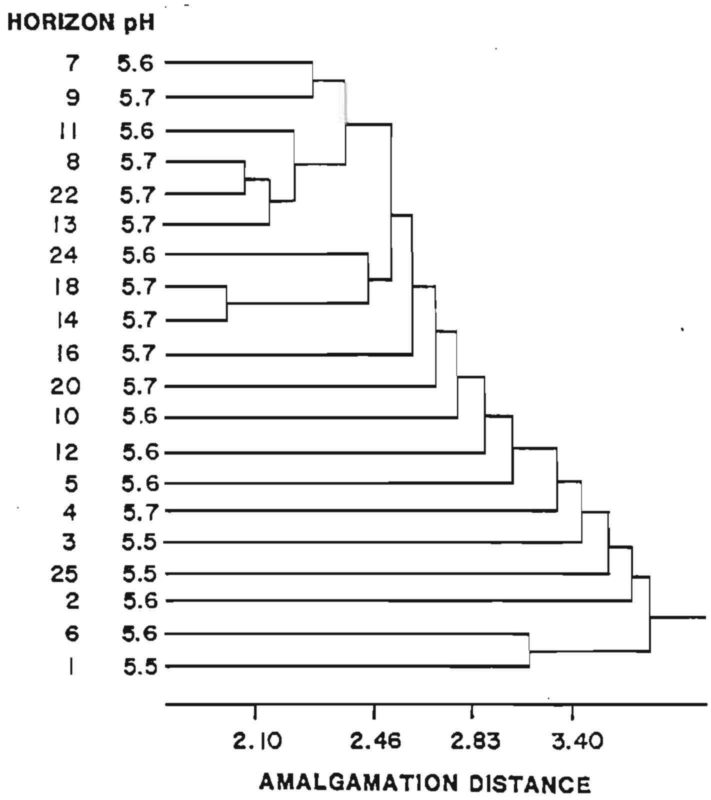

Fig. 8. Cluster analysis of selected diatom stratigraphies (% occurrence) for lake 12. pH is Index B inferred pH and the amalgamation distance is derived from a chi-square statistic.

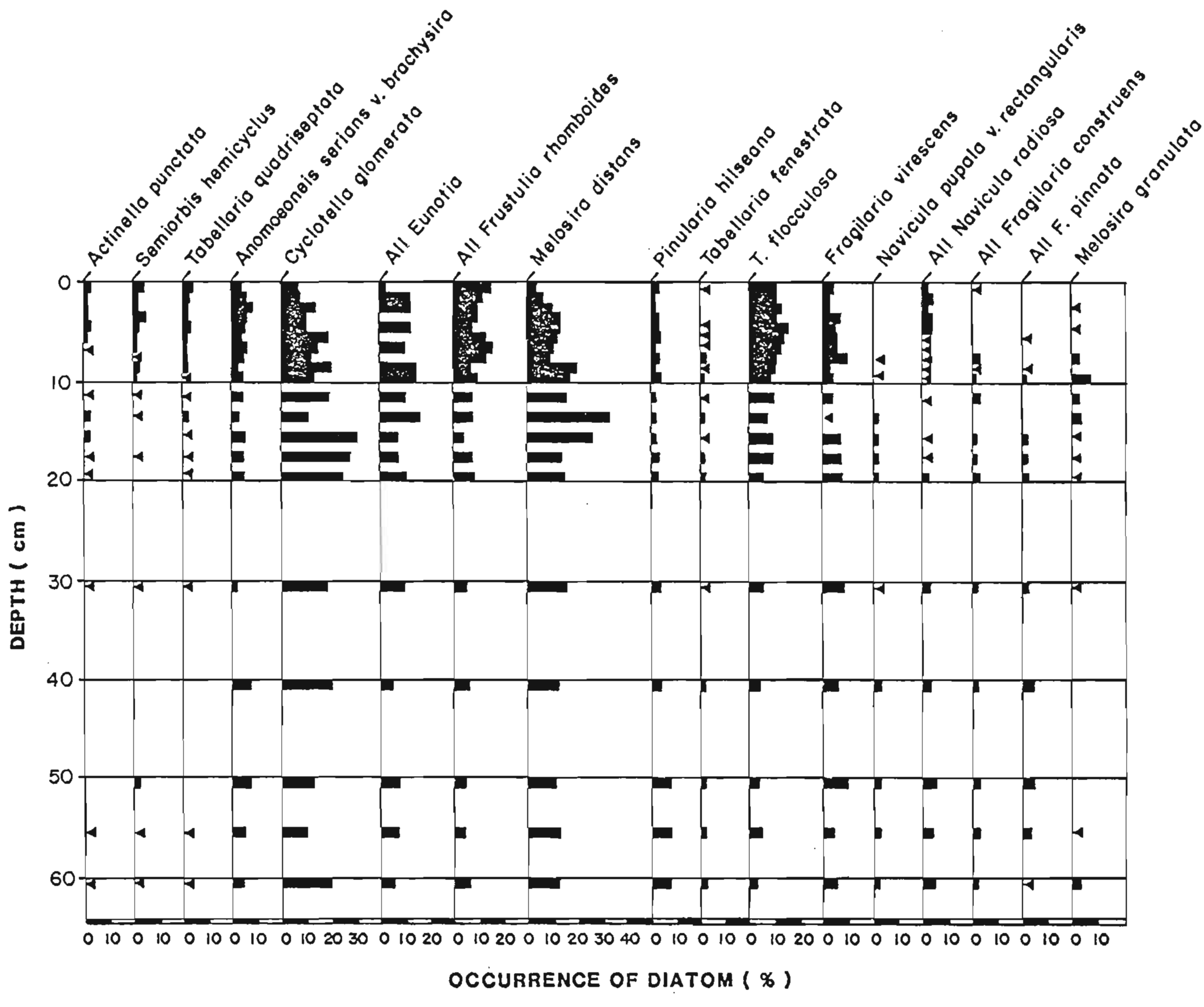

Fig. 9. Downcore profile of the frequencies (% occurrence) of selected key diatom taxa for lake 204.

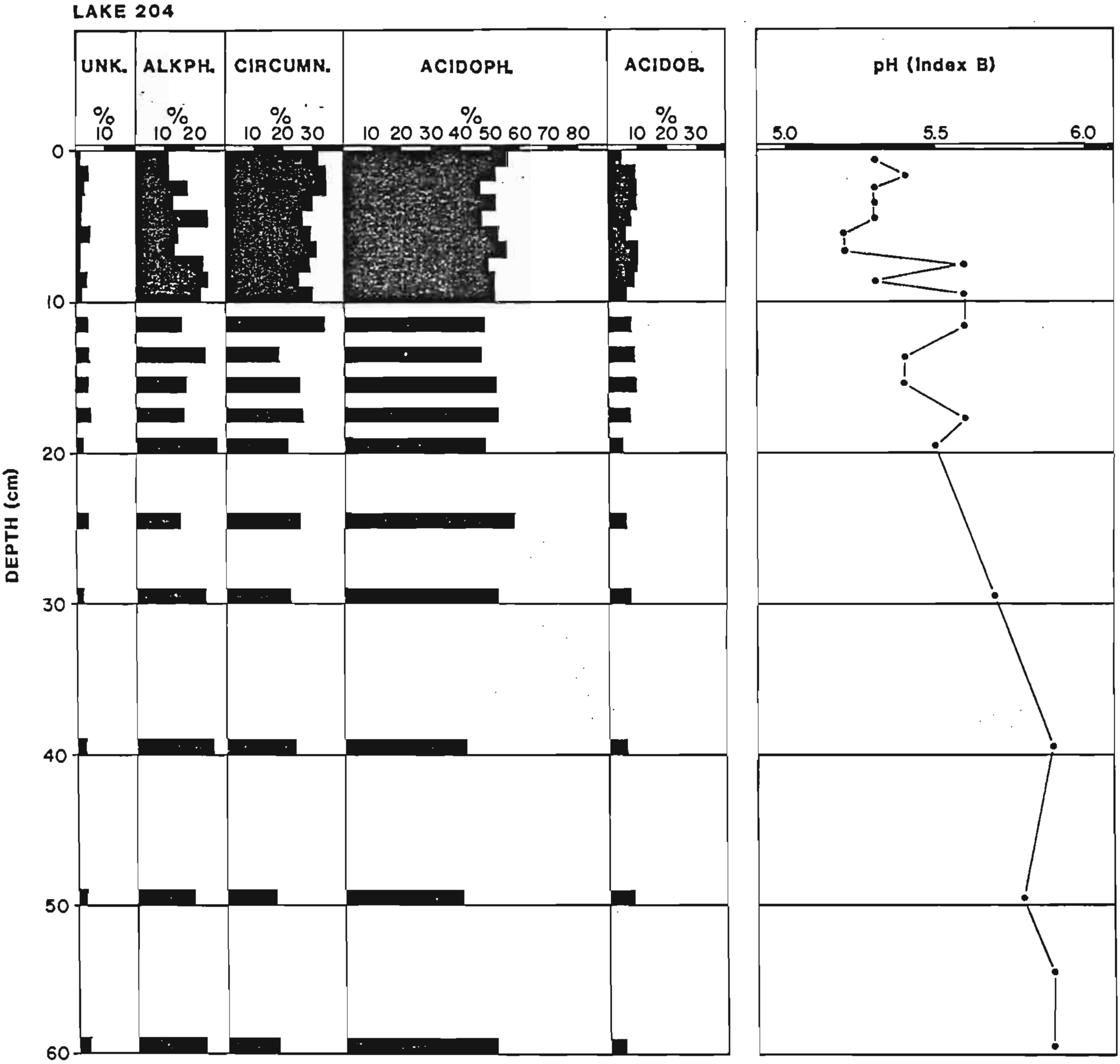

Fig. 10. Downcore profile of the frequencies (% occurrence) of diatoms in pH tolerance categories and Index B inferred pH for lake 204.

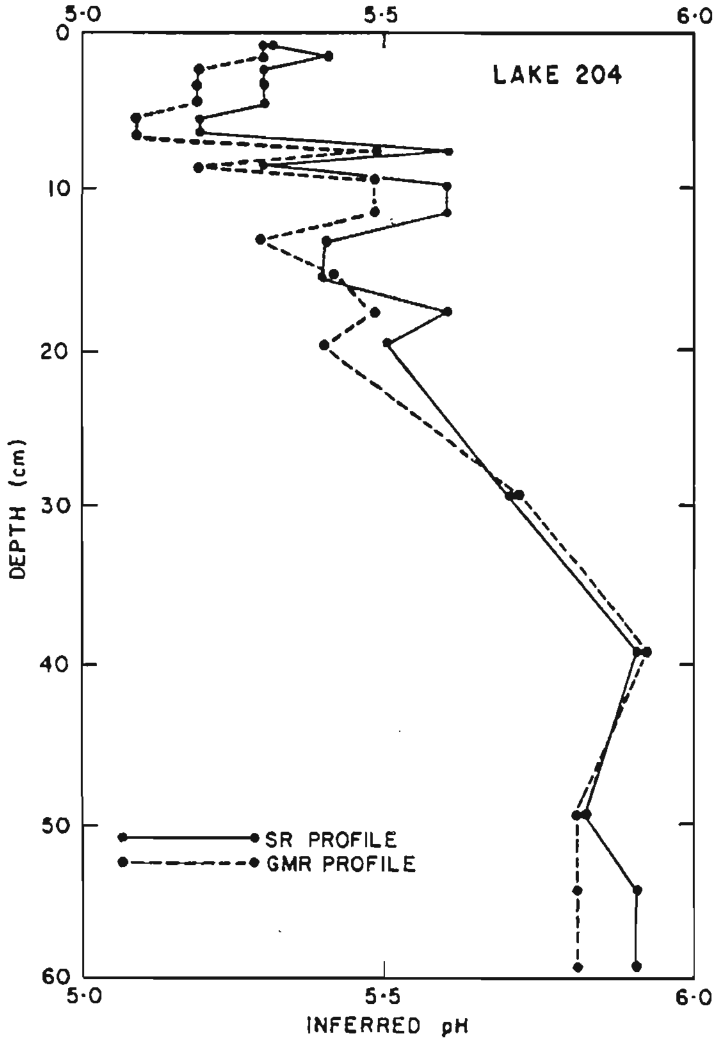

Fig. 11. A comparison of inferred pH profiles for lake 204 using transfer functions obtained by simple linear regression (SR; pH = 6.63-0.87 log Index B) and geometric mean regression (GMR; pH = 6.70-0.98 Log Index B).

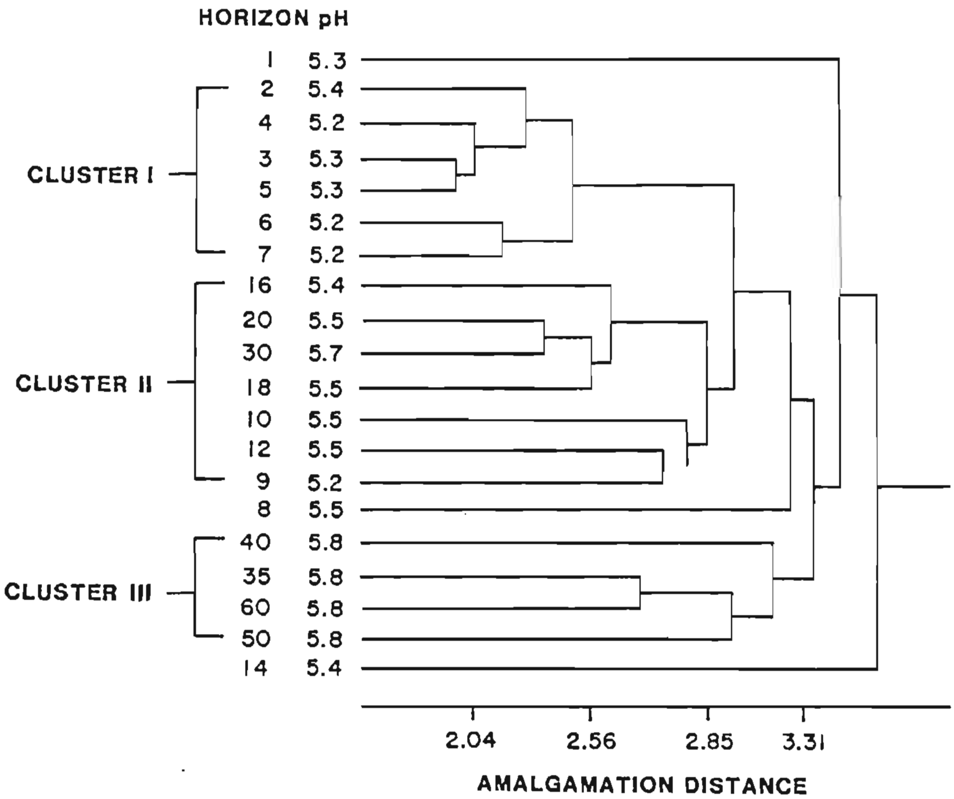

Fig. 12. Cluster analysis of selected diatom stratigraphies (% occurrence) for lake 204. pH is Index B inferred pH and the amalgamation distance is derived from a chi-square statistic.

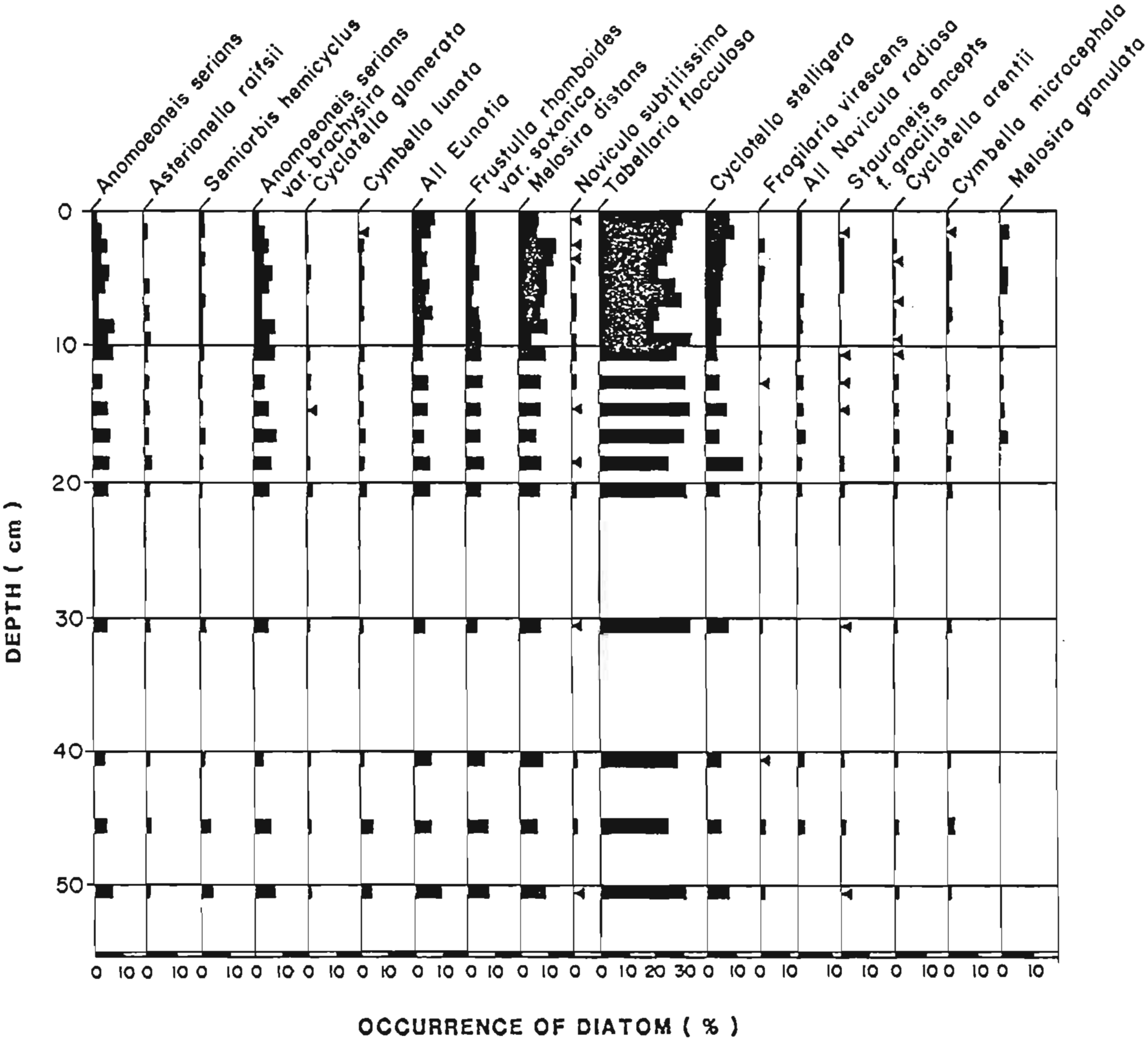

Fig. 13. Downcore profile of the frequencies (% occurrence) of selected key diatom taxa for lake 219.

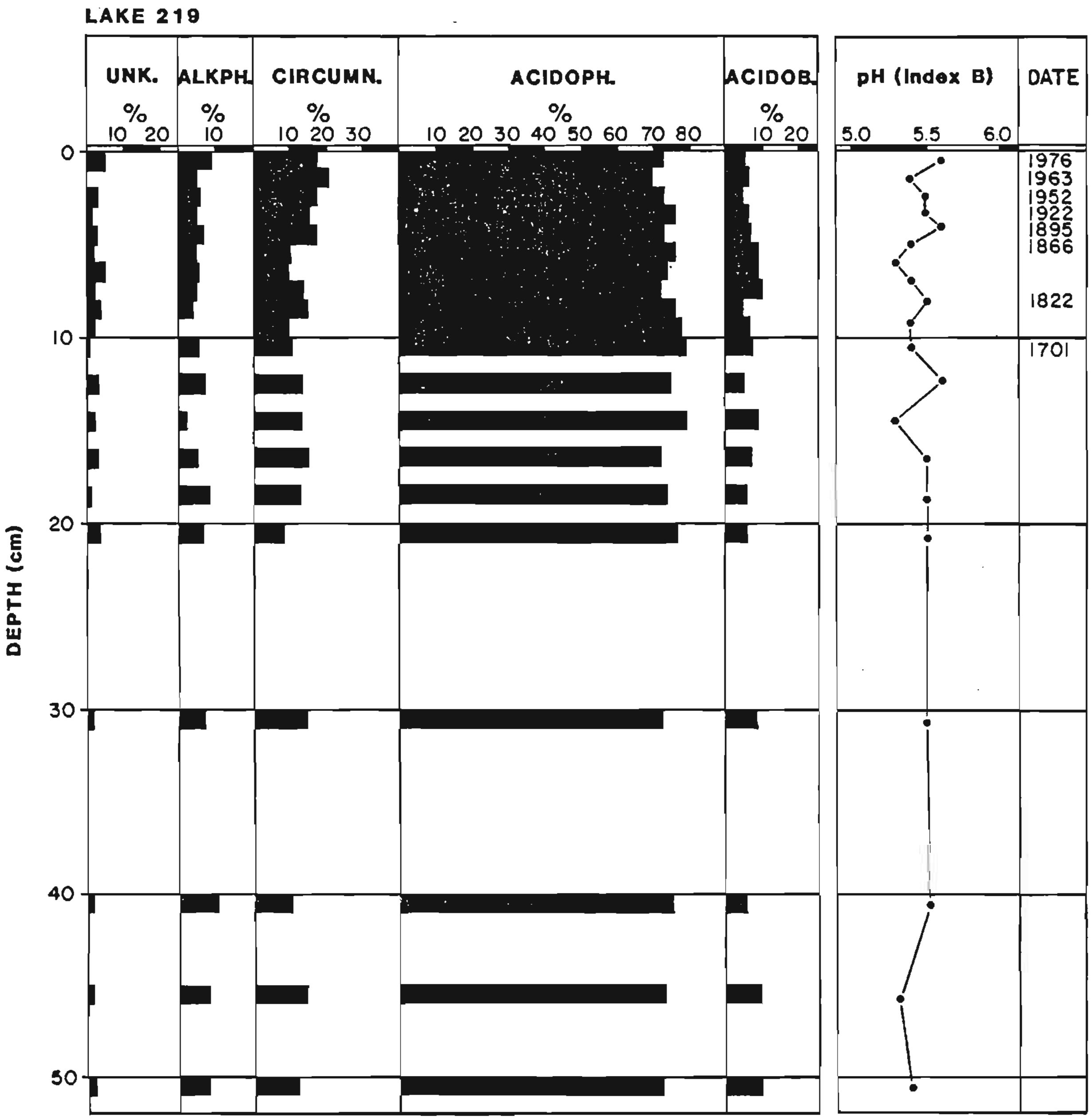

Fig. 14. Downcore profile of the frequencies (% occurrence) of diatoms in pH tolerance categories and Index B inferred pH for lake 219.

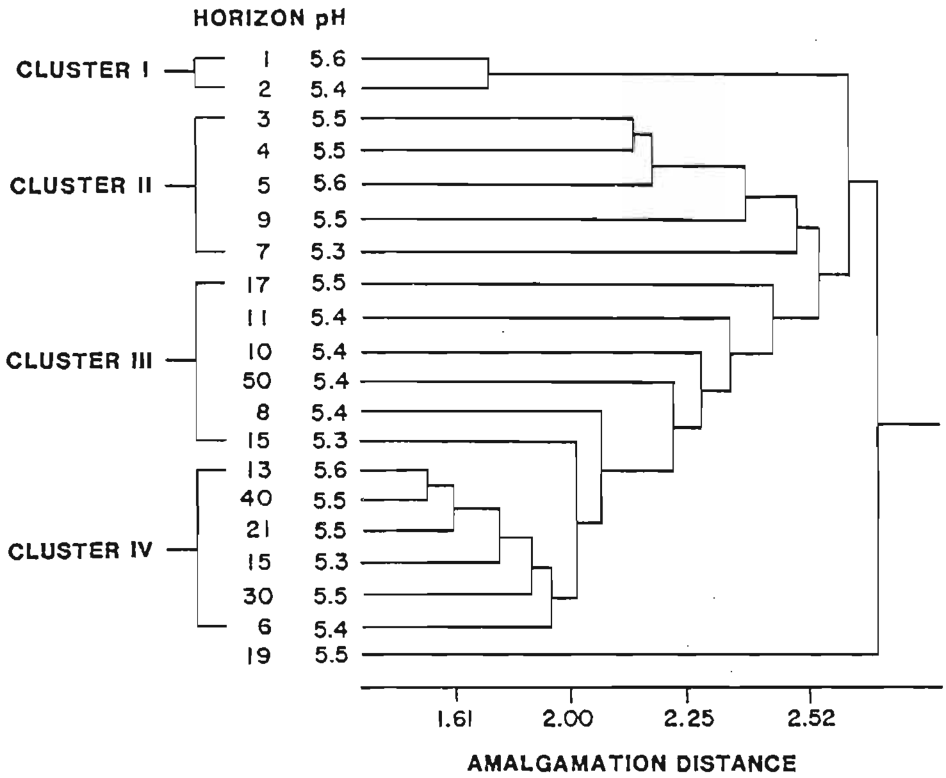

Fig. 15. Cluster analysis of selected diatom stratigraphies (% occurrence) for lake 219. pH is Index B inferred pH and the amalgamation distance is derived from a chi-square statistic.

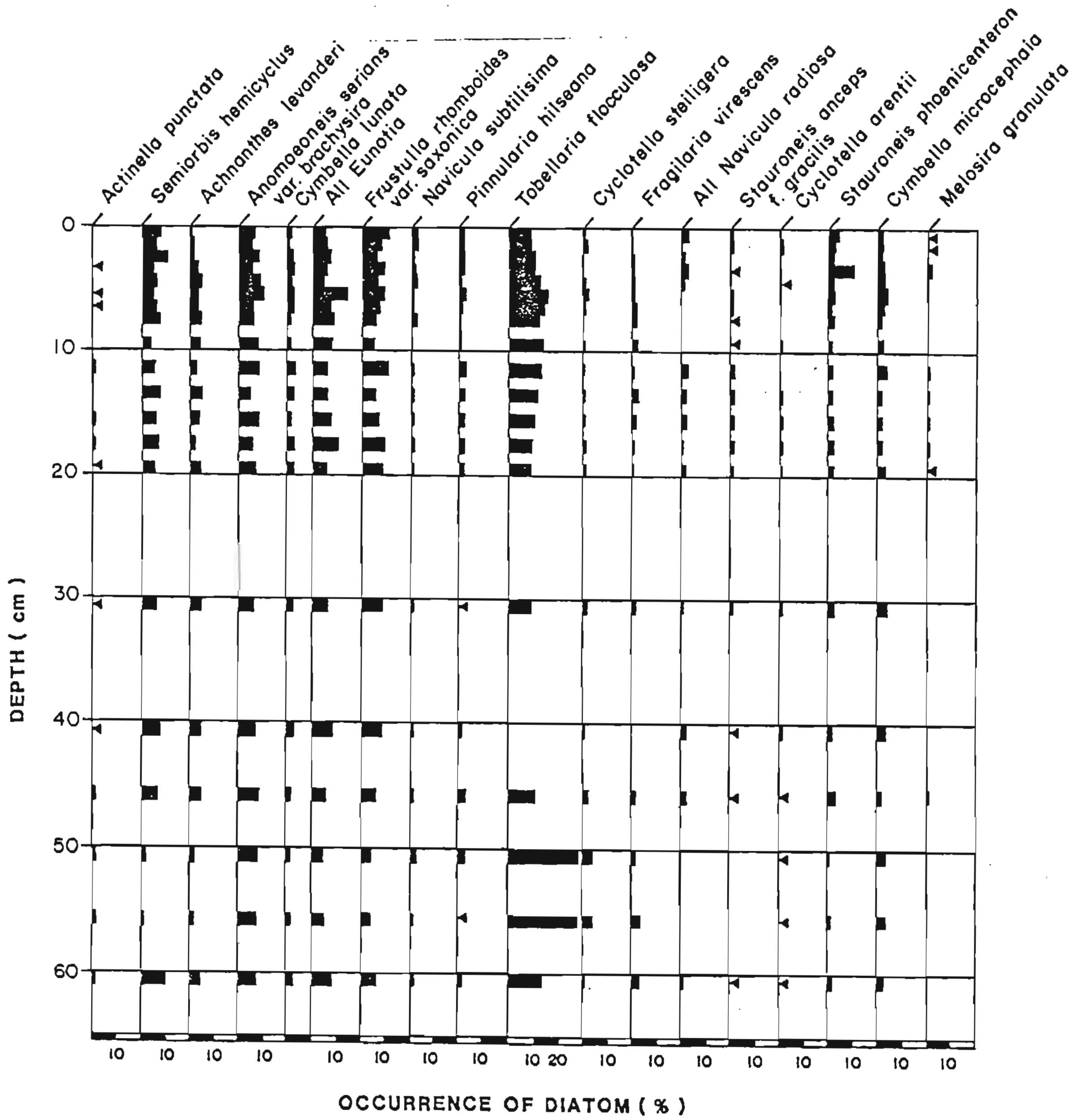

Fig. 16. Downcore profile of the frequencies (% occurrence) of selected key diatom taxa for lake 660.

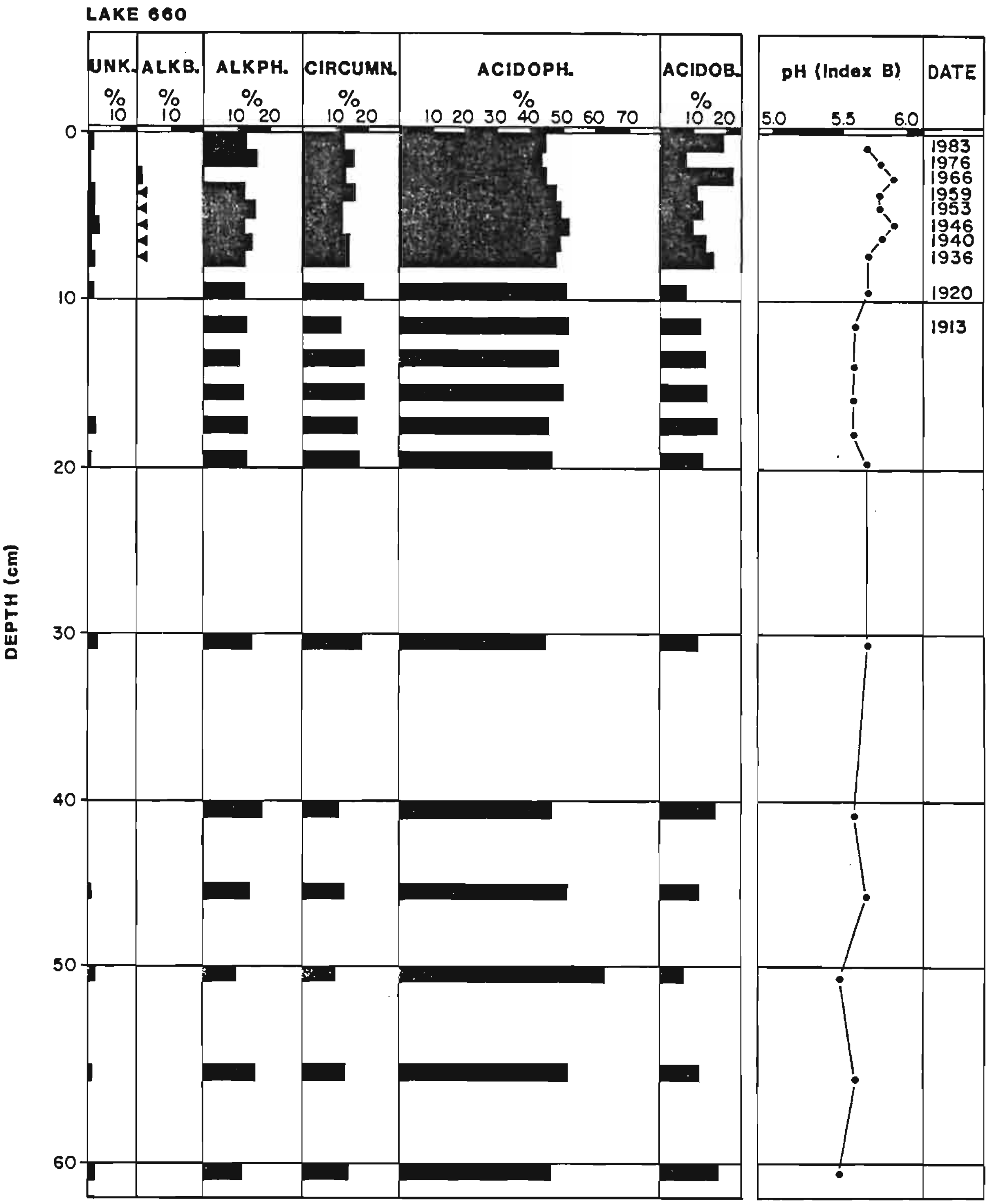

Fig. 17. Downcore profile of the frequencies (% occurrence) of diatoms in pH tolerance categories and Index B inferred pH for lake 660.

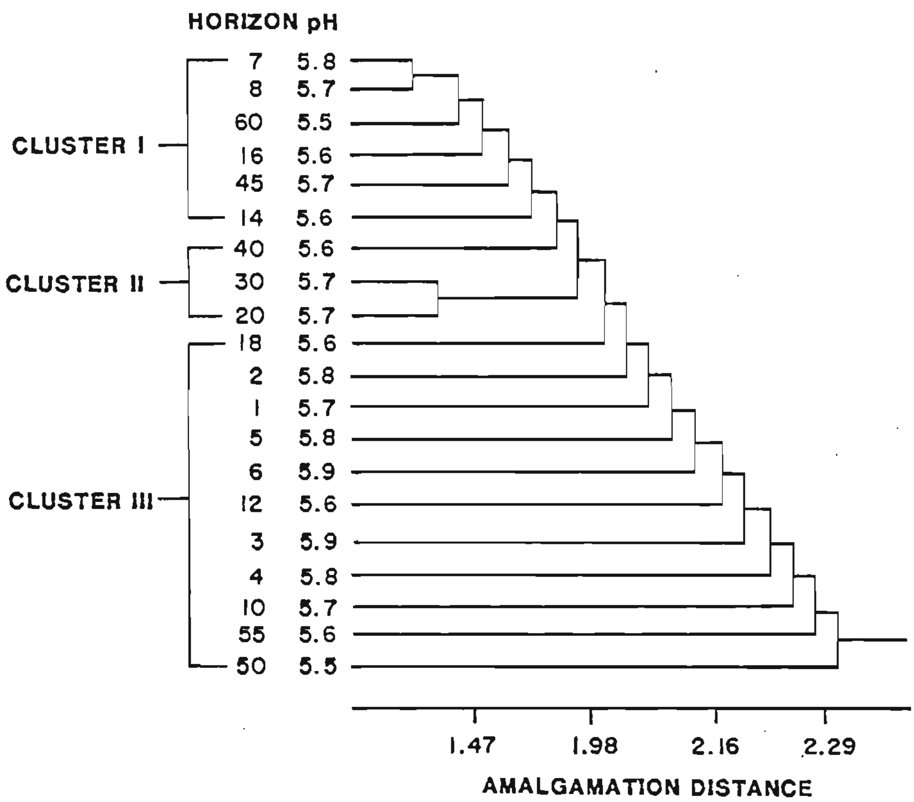

Fig. 18. Cluster analysis of selected diatom stratigraphies (% occurrence) for lake 660. pH is Index B inferred pH and the amalgamation distance is derived from a chi-square statistic.

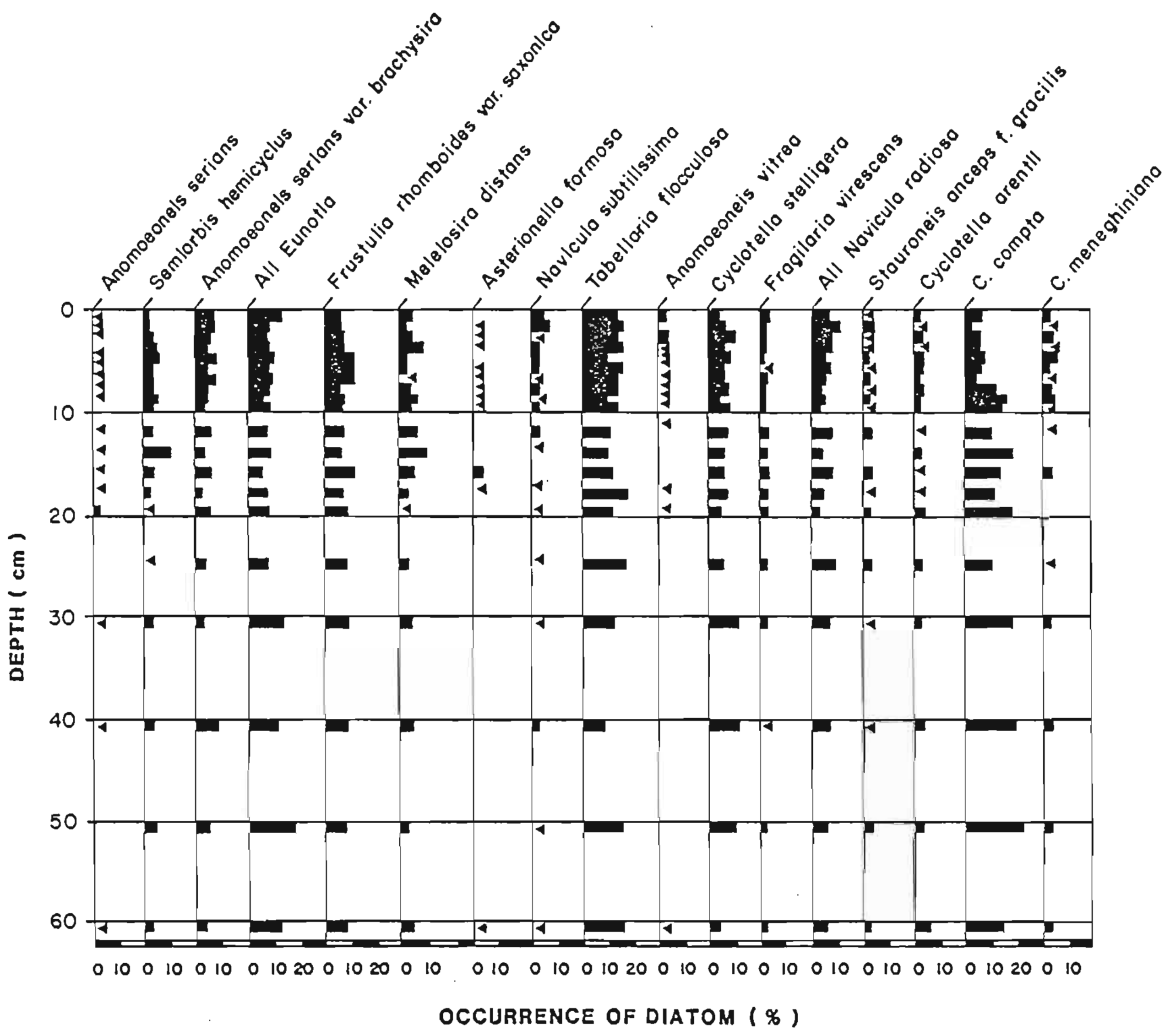

Fig. 19. Downcore profile of the frequencies (% occurrence) of selected key diatom taxa for Stephenson's Pond (lake 206).

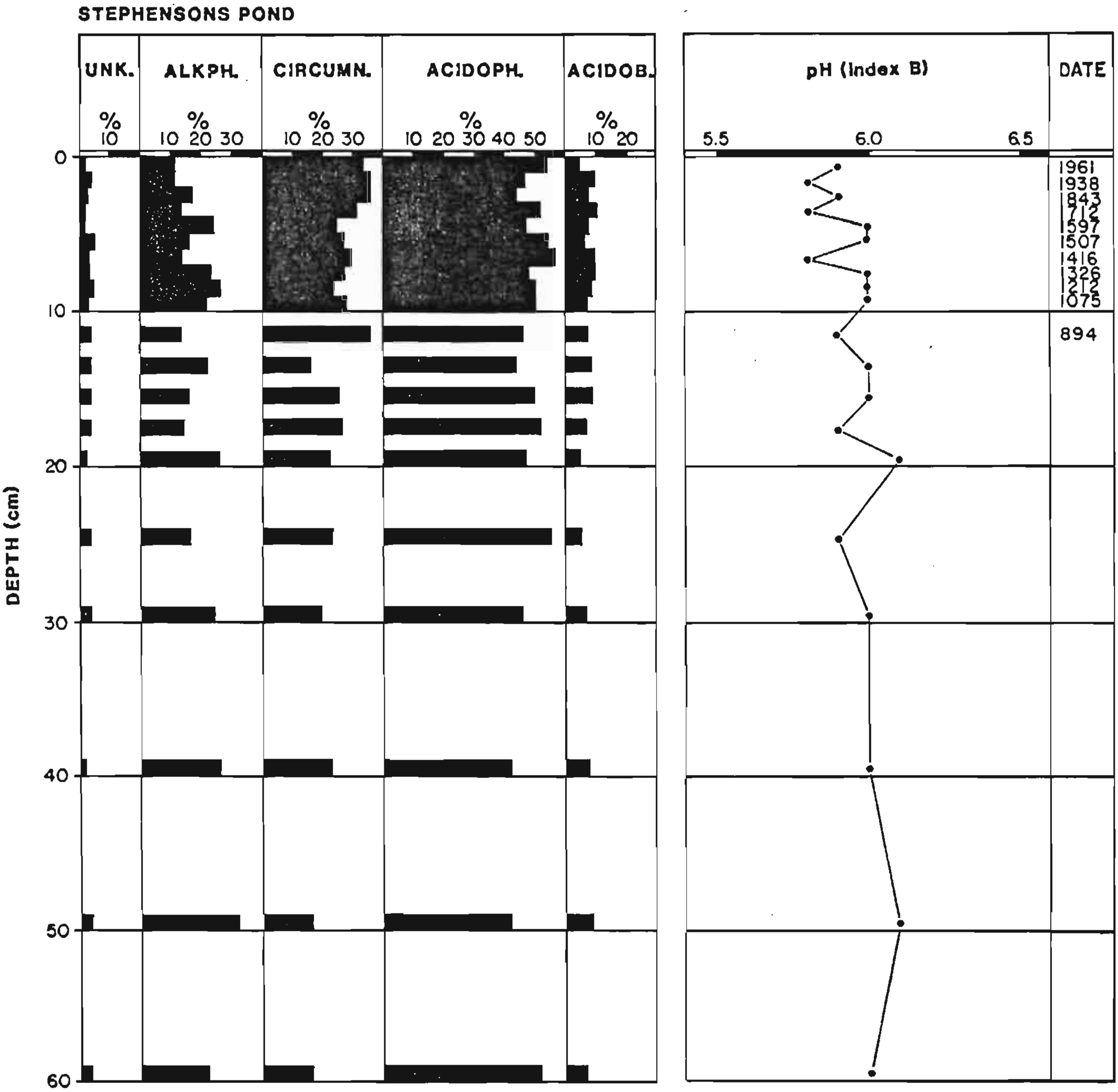

Fig. 20. Downcore profile of the frequencies (% occurrence) of diatoms in pH tolerance categories and Index B inferred pH for Stephenson's Pond (lake 206).

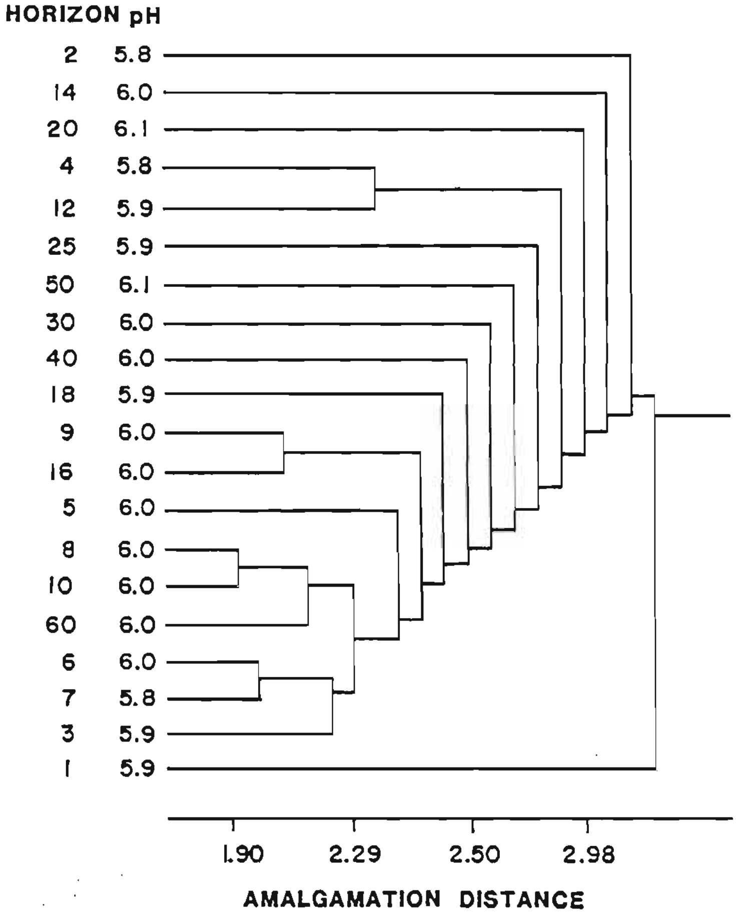

Fig. 21. Cluster analysis of selected diatom stratigraphies (% occurrence) for Stephenson's Pond (lake 206). pH is Index B inferred pH and the amalgamation distance is derived from a chi-square statistic.

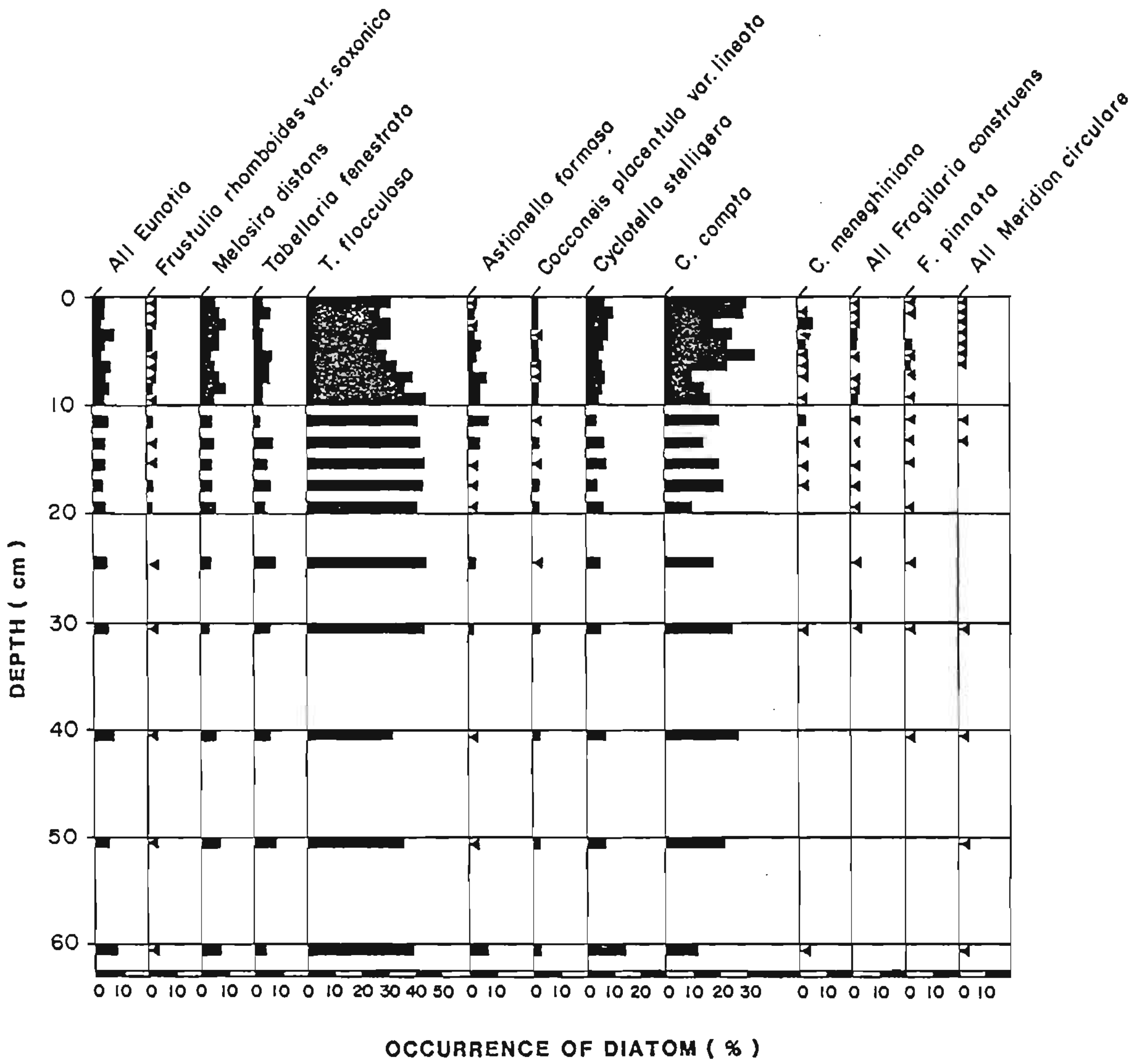

Fig. 22. Downcore profile of the frequencies (% occurrence) of selected key diatom taxa for Aides Pond.

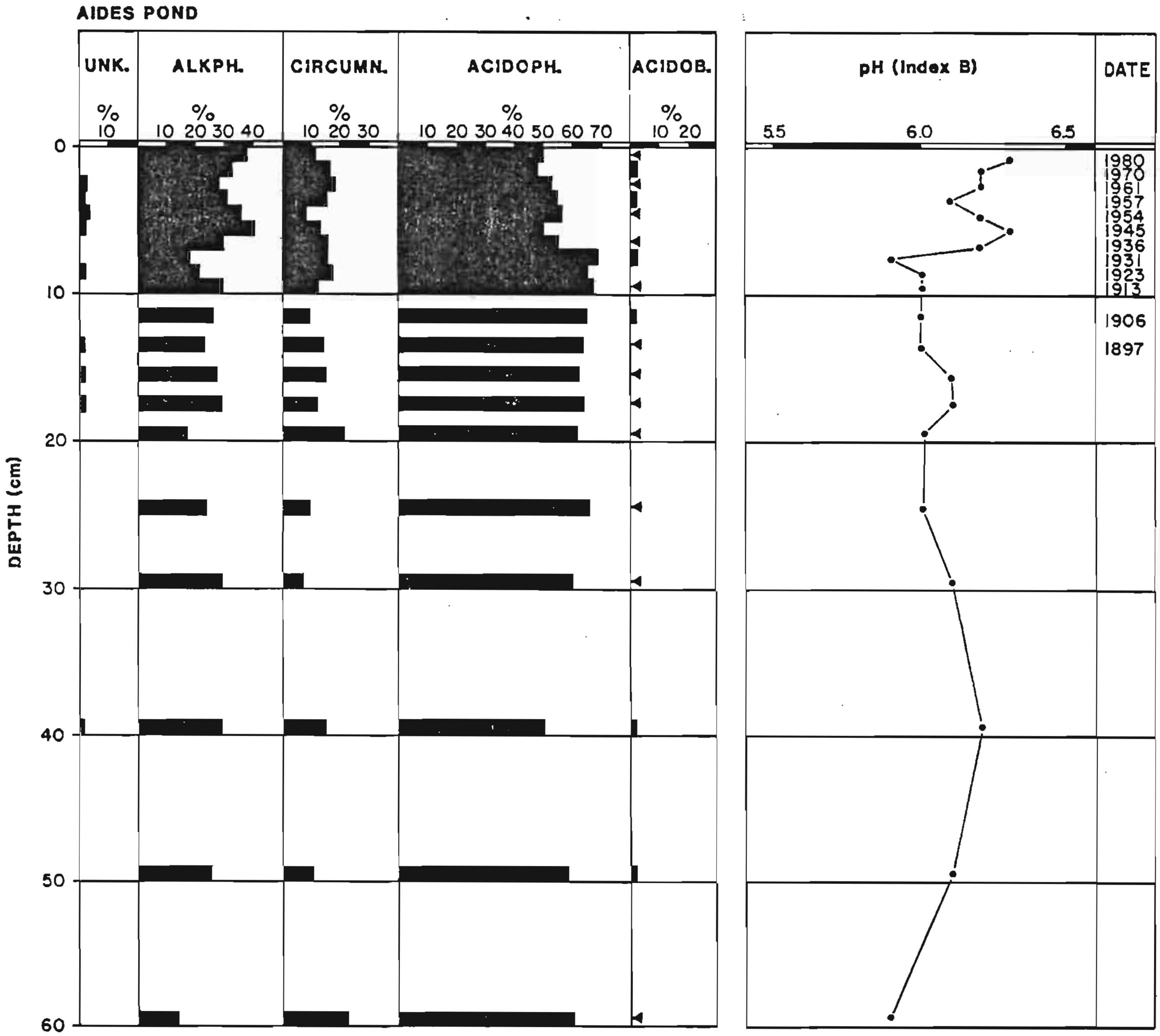

Fig. 23. Downcore profile of the frequencies (% occurrence) of diatoms in pH tolerance categories and Index B inferred pH for Aides Pond.

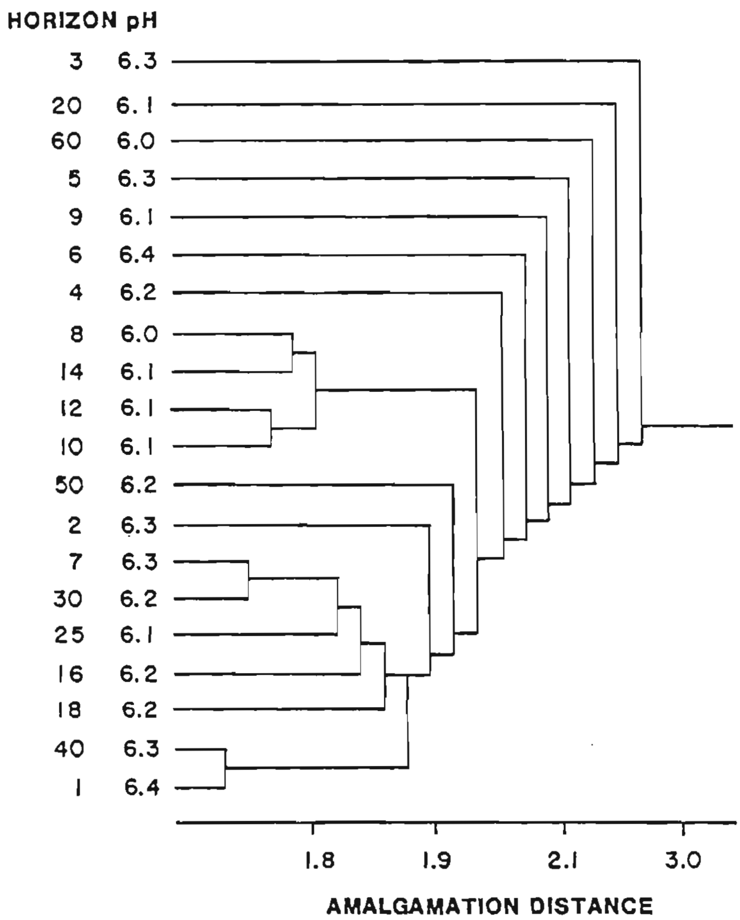

Fig. 24. Cluster analysis of selected diatom stratigraphies (% occurrence) for Aides Pond. pH is Index B inferred pH and the amalgamation distance is derived from a chi-square statistic.

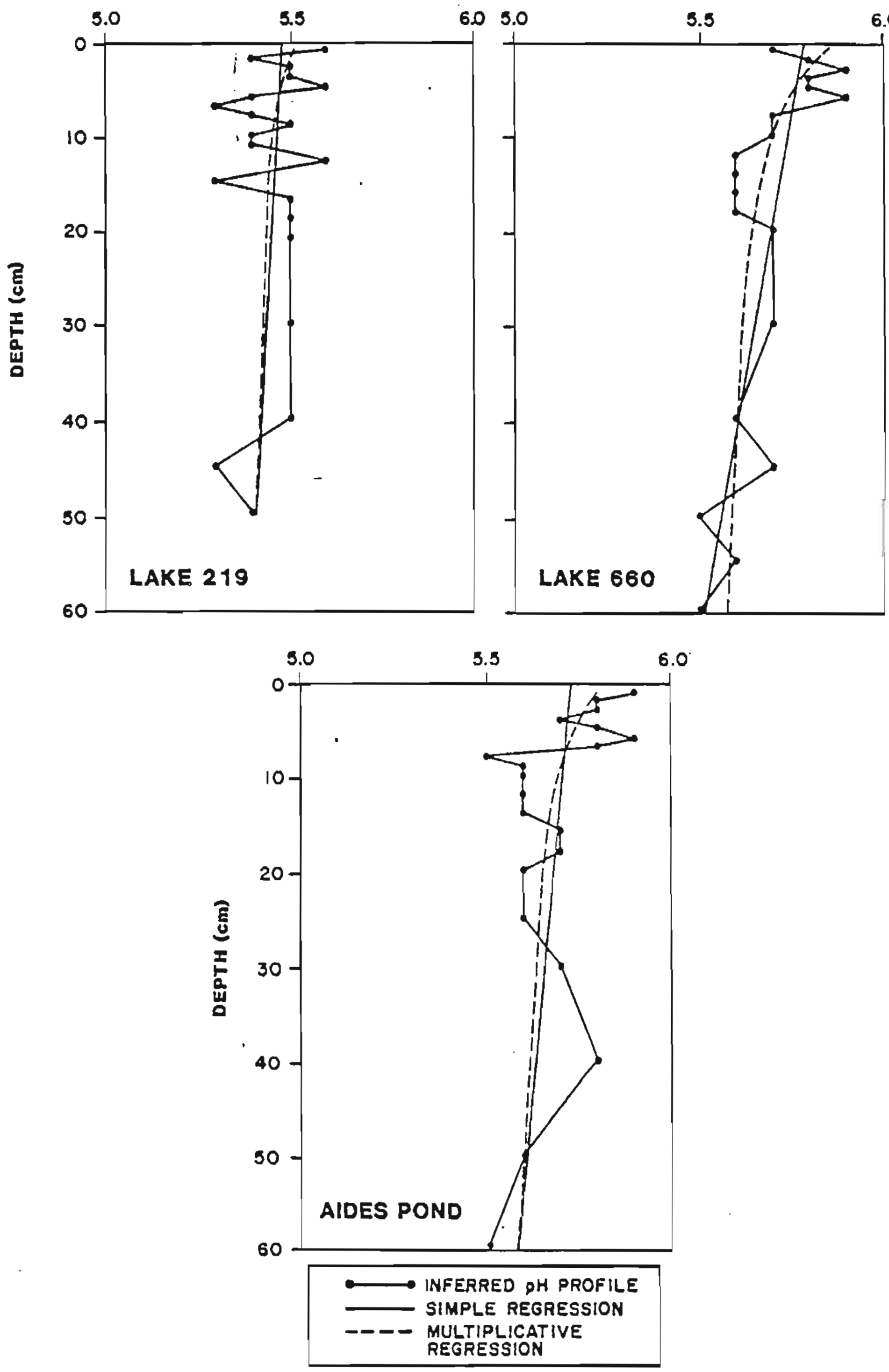

Fig. 25. Plot of Index B inferred pH profiles with regression lines (simple, SR, and multiplicative, MR) of pH vs. sediment depth for lakes 8, 12, 204 and Stephenson's Pond. Refer to Table 3 for regression equations and significance of regression.

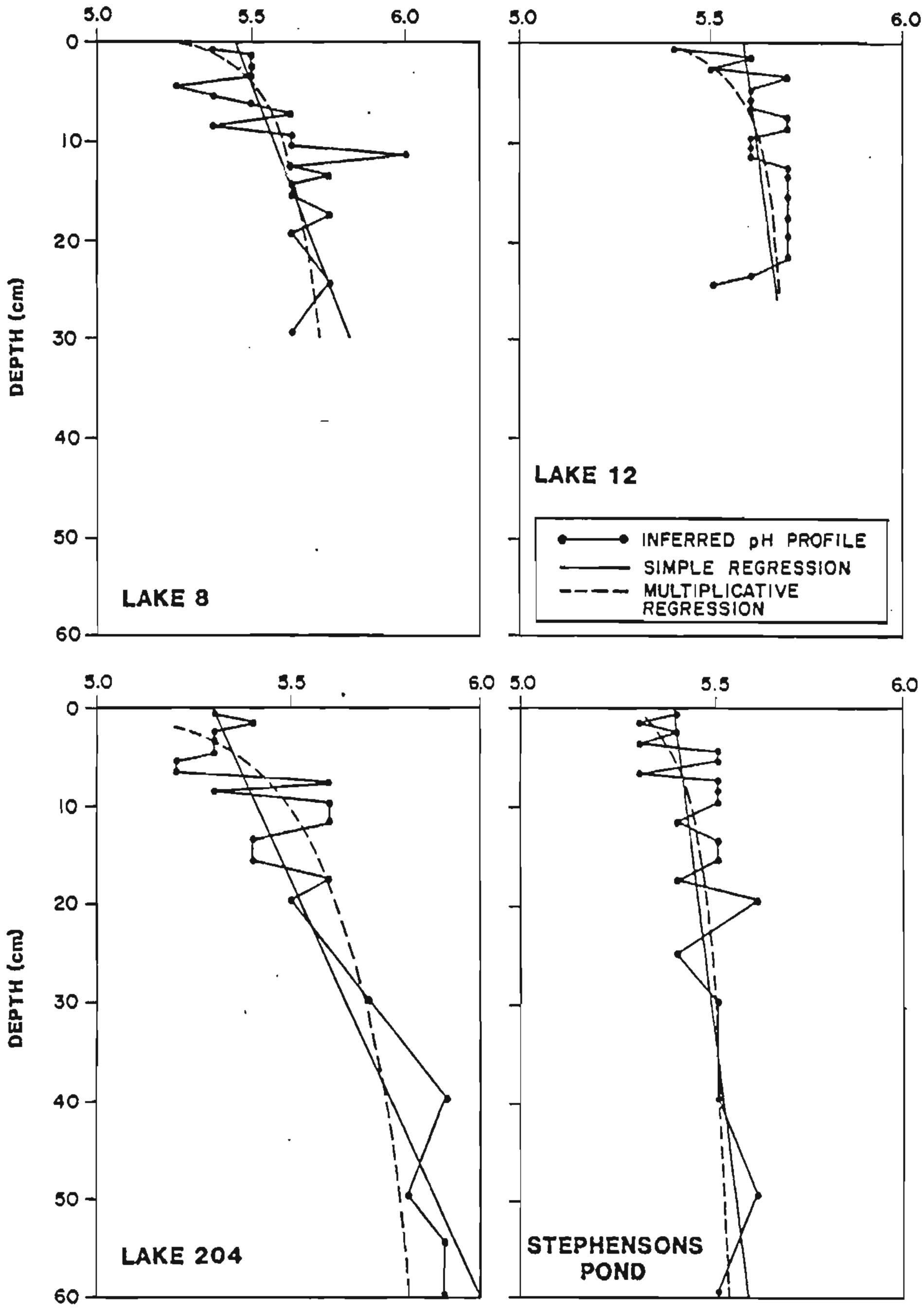

Fig. 25 (Cont'd.). Plot of Index B inferred pH profiles with regression lines (simple, SR, and multiplicative, MR) of pH vs. sediment depth for lakes 219, 660, and Aides Pond. Refer to Table 3 for regression equations and significance of regression.