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Source: Arctic, Antarctic, and Alpine Research, 38(3) : 465-476

Published By: Institute of Arctic and Alpine Research (INSTAAR), University of Colorado

URL: https://doi.org/10.1657/1523-0430(2006)38[465:ACROAN]2.0.CO;2

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Arctic, Antarctic, and Alpine Research, Vol. 38, No. 3, 2006, pp. 465–476

Are Current Rates of Atmospheric Nitrogen Deposition Influencing Lakes in the Eastern Canadian Arctic?

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Abstract

Although arctic lakes rank among the most pristine ecosystems remaining on Earth, widespread paleoecological analyses have revealed rapid recent changes in lake ecology that largely surpass Holocene natural variability and that are generally attributed to climate warming since the end of the Little Ice Age. However, the possibility that climate is only one dimension of these unprecedented ecological shifts remains an untested possibility, especially given that current warming may not yet exceed maximum, naturally mediated, postglacial warmth. In this paper, we assess whether increased anthropogenic nitrogen (N) deposition from distant sources is contributing to directional changes in the biogeochemistry and ecology of two remote lakes on Baffin Island in the eastern Canadian Arctic. Paleolimnological analyses, including diatom assemblages and a suite of biogeochemical proxies (total organic matter, biogenic silica, organic N and C contents, and stable isotopic ratios) reveal a complex suite of progressive changes that are coherently expressed in both lakes. Diatom assemblages began to change as early as the mid-19th century, but major inflections in the biogeochemical proxies occurred significantly later, being most pronounced after 1950. Among these changes are increases in sediment organic matter, depletions of 2‰ in sediment $\delta^{15}N$, and decoupling of $\delta^{13}C$ and $\delta^{15}N$ signatures. It seems likely that climate warming, subsequently coupled to anthropogenic N deposition, is synergistically driving these ecosystems towards states for which no prior natural analogs exist.

Introduction

The Arctic is highly susceptible to climate change because of elevated sensitivity to feedbacks involving cryospheric processes (Kattenberg et al., 1996; Comiso, 2003). Although much of the Arctic is currently warming, in part due to anthropogenic greenhouse gas forcing (Overpeck et al., 1997), strong regional differences exist in both the amplitude and seasonal expression of this trend (Serreze et al., 2000; Overland et al., 2004). Arctic lake sediments chronicle 20th-century environmental change with a high degree of fidelity, as illustrated by paleolimnological analyses from an increasingly dense network of sites (Douglas et al., 1994; Gajewski et al., 1997; Wolfe, 2000, 2003; Sorvari et al., 2002; Michelutti et al., 2003; Rühland et al., 2003; Perren et al., 2003). In these examples, diatom assemblages examined at high stratigraphic resolution reveal floral diversifications that are consistent with increased algal production, lengthening of the growing season, and expanded availability of littoral and planktonic habitats. These trends have been corroborated by additional microfossil groups, including chrysophyte algae (Wolfe and Perren, 2001), as well as lacustrine invertebrates including chironomids (Quinlan et al., 2005) and cladocerans (Smol et al., 2005). On the other hand, as rapidly as climate may be warming and hence modulating the ecology of arctic lakes, it may be argued whether current summer temperatures exceed those of the early to middle Holocene, when summer insolation was significantly higher (Berger and Loutre, 1991). Thus, the unprecedented character of recent ecological changes preserved in arctic lake sediments might imply that some agent compounded to recent climate change may be involved.

It has been known for some time that significant atmospheric pollution reaches the Arctic (Barrie et al., 1985, 1988). However, anthropogenic nitrogen (N) deposition has been considered insufficient to induce high-latitude ecological changes (AMAP, 1998), despite experimental results that confer a pronounced sensitivity of arctic freshwater (Benstead et al., 2005) and terrestrial (Gordon et al., 2001) ecosystems to even modest N additions. At the same time, glaciochemical time-series consistently reveal late-20th century nitrate (NO_3^-) concentrations that rise markedly above natural background levels in Greenland (Mayewski et al., 1990; Fischer et al., 1998), Svalbard (Simões and Zagorodnov, 2001; Isaksson et al., 2003), northwestern Canada (Yalcin and Wake, 2001), and the eastern Canadian archipelago (Goto-Azuma and Koerner, 2001). The question remains whether the rates of long-distance atmospheric N deposition implied by these studies (0.2–0.5 kg NO_3^- ha^{-1} yr^{-1}) are ecologically relevant or not. In this paper, we apply high-resolution paleolimnological analyses to explore the potential contributions of anthropogenic N deposition to recent changes recorded in arctic lake sediments. Conceptually and methodologically, this work builds upon efforts to document the ecological and biogeochemical consequences of considerably higher rates of N deposition (2–5 kg N ha^{-1} yr^{-1}) to mid-continental alpine lakes (Baron et al., 2000; Wolfe et al., 2001, 2002, 2003; Saros et al., 2003, 2005). These studies have demonstrated the utility of paleolimnological proxies, including diatom assemblages and nitrogen stable isotopes, in evaluating the ecological consequences of elevated N deposition.

Study Sites

Baffin Island is the largest in the Canadian Arctic Archipelago, spanning 15° of latitude and three major climatic and vegetational zones (Fig. 1). The island's east coast is characterized by steep fiordlands that dissect Precambrian crystalline lithologies. The Clyde Foreland, to the north and west of the Inuit hamlet of Clyde River (population 300), comprises a thick apron of Quaternary glacial and marine sediments at the foot of mountainous terrain rising westward.

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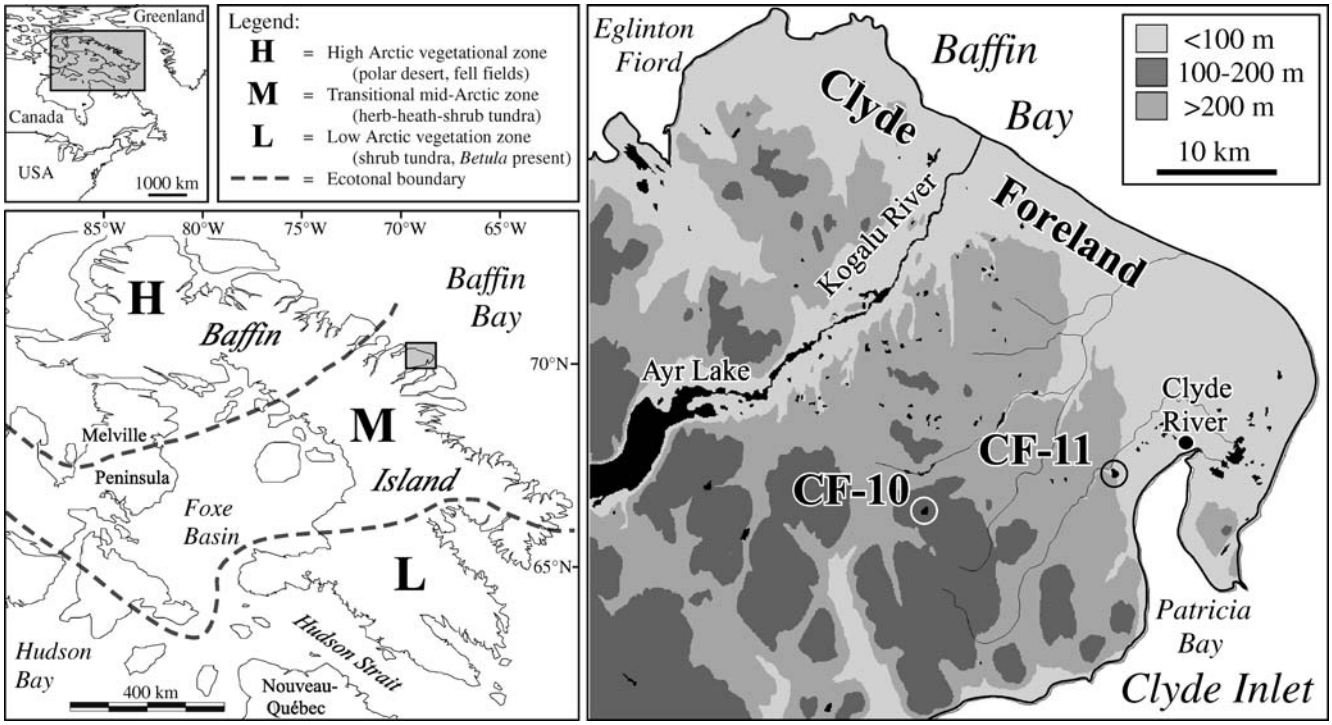


FIGURE 1. Location maps situating the study sites in the mid-arctic climate zone of eastern Baffin Island, Nunavut, Canada.

The study sites are two small unnamed lakes referred to as CF-10 (70°26.121′N, 69°07.223′W) and CF-11 (70°28.758′N, 68°40.190′W), 15 km from each other. CF-10 is an upland lake situated in an alpine col at 435 m. The lake has an area of 0.086 km^2, a maximum depth of 9.6 m, and a steeply sloping catchment of 2.66 km^2. A small glacier drains into this lake from the north. In contrast, CF-11 is considerably lower (96 m), somewhat larger (0.11 km^2) and deeper (11.0 m), and occupies a very small catchment (0.57 km^2) of low relief in unconsolidated Quaternary glacial deposits.

Both lakes are highly oligotrophic (total phosphorous, TP <4 μg L^{-1}), dilute (conductivities <50 μS cm^{-1}), and slightly acidic (pH range: 6.0–6.9). The buffering capacity of these waters is negligible. Relatively high dissolved inorganic nitrogen (DIN, $NO_3^- + NO_2 + NH_4^+$) concentrations (>100 μg L^{-1}) were measured in the epilimnion of CF-10 (Table 1), which was sampled on the first day of snowmelt (28 May 2002). Both snowmelt and bottom waters from this lake, as well as the epilimnion of CF-11, also had DIN concentrations >10 μg L^{-1}. However, $NO_3^- + NO_2$ concentrations in the hypolimnia of both lakes were very low (<1 μg L^{-1}), probably reflecting the cumulative effects of winter denitrification near the sediment-water interface. NH_4^+ concentrations in both lakes were also higher in the epilimnion than in the hypolimnion, implying that microbial remineralization of sediment organic matter is a secondary source of water column NH_4^+. Although the available water chemistry (Table 1) is limited in the sense of solely reflecting late-winter limnological conditions, it nonetheless supports the concept that snowmelt is the dominant contributor of DIN to the lakes, as in alpine catchments at lower latitudes (Baron and Campbell, 1997).

Mean annual temperature at Clyde River is −12.3°C, with July temperatures averaging 4.5°C. Annual precipitation is approximately 300 mm, with roughly 80% falling as snow. Thick lake ice cover (to 190 cm) is a salient feature of these lakes. In typical years, ice cover extends from September to July, with an annual ice-free period of less than 2 mo. Vegetation and soil development in the catchments are minimal, especially in the CF-10 basin which is primarily talus and bedrock. A sparse heath tundra with abundant bryophytes surrounds CF-11.

TABLE 1

Chemistry of surface and bottom waters in the study lakes, and snowmelt on the surface of CF-10. Samples were collected on 28 May (CF-10) and 31 May (CF-11), 2002. Bottom waters were collected within 20 cm of the mud-water interface without any disturbance of sediments (DL = detection limit).

Sample	NH_4^+ (μg L^{-1})	$NO_2 + NO_3^-$ (μg L^{-1})	TP (μg L^{-1})	Conductivity (μS cm^{-1})	pH
CF-10 epilimnion	54.36	79.62	1.4	23.9	6.46
CF-10 hypolimnion	18.76	0.36	3.5	19.5	6.36
CF-10 meltwater on ice surface	7.50	18.70	2.3	14.1	6.04
CF-11 epilimnion	11.40	3.33	2.9	42.7	6.91
CF-11 hypolimnion	6.26	<0.1 (*DL*)	2.1	30.7	6.90

Methods

CORING AND SEDIMENT CHRONOLOGY

The deepest points in the central basins of both lakes were cored using a modified Kajak-Brinkhurst gravity corer that preserves the mud-water interface intact (Glew, 1989). Both cores were extruded continuously in the field (Glew, 1988), in 0.25-cm increments from the surface to 5.0 cm, and in 0.5-cm increments below 5.0 cm. All subsampled core material was weighed and freeze-dried in the laboratory prior to subsequent analyses. The cores were dated using ^{210}Pb activities ($t_{1/2} = 22.3$ yr) measured by α-spectroscopy, and applying the constant rate of supply (CRS) accumulation model to unsupported ^{210}Pb inventories (Appleby and Oldfield, 1978). Dating recent arctic lake sediments is not straightforward for two reasons. First, the short seasonal ice-free window limits the effective delivery of unsupported ^{210}Pb, the product of atmospheric ^{222}Rn decay, to the lake system. Second, low primary production results in both extremely low recent

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sediment accumulation rates (e.g., in the order of 20 to 30 yr per cm), as well as inefficient ^{210}Pb scavenging by sedimenting organic matter. Despite these caveats, clear demarcations between zones of unsupported and supported ^{210}Pb are apparent in the cores from both lakes (Fig. 2). Supported ^{210}Pb, which originates from ^{226}Ra associated with U-bearing minerals in the inorganic sediment matrix, was thus approximated from the activities of the lowermost samples, and subtracted from total activities of the upper samples to derive unsupported ^{210}Pb activities for these intervals.

DIATOM ANALYSIS

For diatom analysis, 100 mg of freeze-dried sediment were oxidized with 15% H_2O_2 for 24 h, then 30% H_2O_2 for an additional 24 h, and finally heated (90°C) for 2 h. Digested slurries were centrifuged and rinsed with deionized water four times, and then diluted to 10 mL. In order to estimate diatom absolute abundances, a spike of *Eucalyptus* pollen (146,000 ± 2400 grains) was added to 200 µL aliquots from each sample (Wolfe, 1997). These secondary slurries were diluted again (to 5 mL), thoroughly mixed, and further 200 µL aliquots were strewn onto coverslips, dried at air temperature, and permanently mounted to slides in Naphrax® medium. Diatoms and *Eucalyptus* markers were counted at 1000× under oil immersion with differential interference contrast optics, to sums of at least 400 valves per sample. Counts are expressed as both relative frequencies of taxa in relation to the sum, and concentrations of valves per gram dry sediment based on the recovery of introduced markers (Battarbee and Kneen, 1982). Diatom taxonomy is based on a large number of bibliographic sources that address low-alkalinity lake floras; a complete listing is provided by Joynt and Wolfe (2001).

BIOGENIC SILICA

Biogenic silica ($BSiO_2$) reflects the total contribution of amorphous silica to sediments from siliceous algae (diatoms and chrysophytes), and thus is a proxy for total paleoproduction by these groups, which can be independent of absolute cell numbers when their size or degree of silicification is variable (Conley and Schelske, 2001). $BSiO_2$ was analyzed by wet digestion of 500 mg sediment samples following DeMaster (1991). Organic matter was first removed with 30% H_2O_2 at room temperature for 4 d. $BSiO_2$ was then dissolved with 1% Na_2CO_3 (20 mL) at 80°C in a heated agitating water bath. Aliquots were examined microscopically every half-hour until all diatom valves were dissolved (5 h). Upon complete digestion of $BSiO_2$, slurries were centrifuged and 1.0 mL aliquots of supernatant were neutralized with 9.0 mL 0.02 N HCl prior to analysis of total dissolved silica by molybdate blue reduction (APHA, 1985). Measured $BSiO_2$ was then converted to a percentage of original sample dry mass. Duplicate analyses indicate a reproducibility of $BSiO_2$ analyses of ±1.0%. Analyses of incremental hourly digests (1–7 h) confirmed stabilized $BSiO_2$ concentrations after 5 h, suggesting that this approaches the optimal digestion period for arctic lake sediments (Kaplan et al., 2002). Longer digestion is likely to overestimate $BSiO_2$ through chemical attack of aluminosilicates.

SEDIMENT ORGANIC MATTER AND ISOTOPIC CHARACTERIZATION

Total sediment organic matter was estimated by loss on ignition (LOI) at 550°C for 1.5 h, and is expressed as a percentage of total preburn dry mass (Dean, 1974). Following ignition, samples were cooled in a desiccating chamber prior to weighing, in order to eliminate condensation. Measurements of sediment N and δ^{15}N are used to fingerprint potential changes in nitrogen biogeochemistry (Robinson,

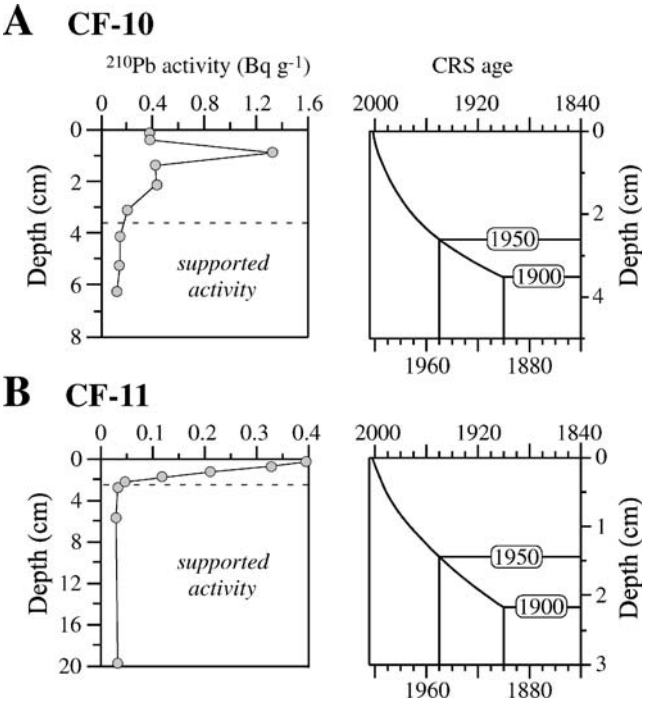


FIGURE 2. Down-core sediment ^{210}Pb activities and corresponding CRS age-depth models for the upper few cm of the CF-10 (A) and CF-11 (B) gravity cores. Dashed horizontal lines indicate the estimated depth of exhaustion of unsupported ^{210}Pb.

2001). The concentration of N is reported as a percentage of dry mass, while the isotopic ratio is expressed in δ notation, where $\delta^{15}N\ (‰) = [(R_{sample}/R_{standard}) - 1] \cdot 1000$, R is the $^{15}N/^{14}N$ ratio, and the standard is air. These measurements were made using an elemental analyzer (Carlo Erba 2500) online with a continuous flow isotope ratio mass spectrometer (CF-IRMS, Finnigan Delta Plus). Analytical reproducibility is ±0.1% for N, and ±0.2‰ for δ^{15}N. Duplicate samples of different mass (0.25–1.00 µg N) verified that pyrolysis of organic matter was complete prior to isotopic measurement.

In order to refine the interpretation of δ^{15}N trends, a series of C isotopic measurements were also conducted. These measurements are only available for CF-10 due to insufficient remaining material from the CF-11 core, and represent completely independent analyses with respect to mass spectrometry. Freeze-dried sediments were isolated under vacuum and combusted in sealed Pyrex ampoules with CuO and Ag wire at 550°C for 9 h. The resulting CO_2 was evacuated under vacuum into a dual-inlet Finnigan MAT 252 IRMS. As with N, the C isotopic ratio is expressed in δ notation, δ^{13}C, relative to the Peedee Belemnite (PDB) standard. Duplicate analyses indicate an analytical reproducibility for δ^{13}C of ±0.1‰.

MULTIVARIATE DATA ANALYSIS

The major directions of variability in the diatom and biogeochemical data were summarized by a series of indirect ordinations using Principal Components Analysis (PCA). Preliminary ordinations of the diatom data using Detrended Correspondence Analysis (DCA) revealed first axis gradient lengths of 1.097 (CF-10) and 1.091 (CF-11) standard deviation units. Such short gradients mandate the use of linear techniques, such as PCA (ter Braak and Prentice, 1988). For consistency, and because there is no *a priori* basis to suspect unimodal (Gaussian) responses within the geochemical data, these too were treated with PCA. The first two PCA analyses involve the relative frequencies of all diatom taxa >1% in any one interval. This resulted in

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the inclusion of 36 taxa from 20 samples in the CF-10 core, and 47 taxa from 20 samples in CF-11 (Appendix 1). These PCA analyses were based on centered covariance matrices. The third analysis involved a combined data set representing all depths in both cores (35 samples) for which all five of the following variables were measured: diatom concentrations, $BSiO_2$, LOI, %N, and $\delta^{15}N$. Due to differences in units, these data were first standardized to normalize variances, and PCA was conducted on a centered correlation matrix. Subsequently, this matrix was subdivided into samples predating ($n = 21$) and postdating ($n = 14$) the year 1950 defined by ^{210}Pb chronologies, and each subset was reanalyzed separately by PCA in the same way. The year 1950 was chosen as a threshold for these analyses because it broadly corresponds to a global pulse of anthropogenic N loading to the atmosphere from combined industrial, fossil fuel, and agricultural sources (Galloway, 1989). All ordinations were implemented with CANOCO version 4.5a (ter Braak and Smilauer, 2002).

Results

CORE CHRONOLOGY

The sediments of both lakes are comprised of massive olive (CF-10) to reddish-gray (CF-11) silty gyttja with no apparent horizontal stratification. Both cores appear well-oxidized at their surfaces, with no evidence of an abrupt redox front below the mud-water interface. The complete inventories of unsupported ^{210}Pb are contained within the upper 4 cm (CF-10) and 3 cm (CF-11) of the cores (Fig. 2). Despite such low accumulation rates, the profile from CF-11 reveals an uninterrupted down-core decrease in ^{210}Pb activity that implies a relatively constant sedimentation rate. In CF-10, the situation is complicated by one interval (0.75–1.0 cm) that is anomalously enriched in ^{210}Pb, coupled to the apparent dilution of excess ^{210}Pb in the two overlying samples. These results imply that the history of ^{210}Pb scavenging and dilution has not been constant over recent decades, which is not surprising given the lake's unvegetated and steeply sloping catchment. However, stratigraphic reversals in ^{210}Pb activity are permissible in the CRS model, so that the down-core activities may nonetheless be converted to a corresponding age-depth profile for the upper ~3 cm (Fig. 2). Due to the challenges and uncertainties presented by ^{210}Pb dating in this environment, we use the dating results conservatively to approximate two time-lines, 1900 and 1950, which are used hereafter as chronostratigraphic references for all proxy data.

DIATOMS AND BIOGENIC SILICA

The diatom floras preserved in both cores are typical of low-alkalinity Baffin Island lakes (Joynt and Wolfe, 2001), although considerable differences exist between the taxa present at either site (Appendix 1). Both lakes record pronounced recent stratigraphic variability in diatom assemblages (Fig. 3). In the upper portion of the CF-10 core, *Aulacoseira distans* replaces *A. perglabra* as the dominant centric form, and proportions of both *Brachysira vitrea* and small colonial *Fragilaria* spp. (*F. virescens* var. *exigua* and *F. constricta*) increase. The latter taxonomic shift is initiated well before 1900, whereas the others occur later, between 1900 and 1950 (Fig. 3A). Given our present understanding of the autecology of these algae, their limnological significance remains somewhat limited. Possibly, expanded littoral habitats such as ice-free moats (Smol, 1988) have become increasingly available for colonization by shallow-water periphytic diatoms such as *B. vitrea*. It is noted that *B. vitrea* consistently produces high summer temperature optima in training set from Swedish Lapland (Bigler and Hall, 2002). In CF-11, marked increases of the planktonic genus *Cyclotella* (*C. rossii* and *C. stelligera*) are recorded in sediments deposited in the last ~150 yr (Fig. 3B). Because *Cyclotella* is associated with water column stratification and open-water conditions, it has been directly associated with climate warming in arctic (Smol et al., 2005) and subarctic (Rühland et al., 2003) lakes. Due to its lower elevation and the absence of shading by adjacent valley walls, CF-11 likely experiences locally ameliorated summer conditions relative to CF-10. Nonetheless, diatom communities in both lakes have clearly changed substantially (Fig. 3), and these changes were initiated well before 1900, as observed elsewhere in the Canadian Arctic (Douglas et al., 1994).

Stratigraphic trends in diatom valve concentrations do not reflect these taxonomic changes in any obvious way (Fig. 4). Although there is an erratic increase in diatom concentrations since approximately 1900 in CF-10, this is within the range of earlier variability. In CF-11, peak diatom concentrations occur in the 0.75- to 1.0-cm sample, but this single point is surrounded by low values well-within natural variability. Sediment $BSiO_2$ values broadly parallel down-core trends of diatom concentrations in both lakes prior to 1950, which is a predictable result since total diatom production is the most important contributor to both parameters. However, in the upper 2 cm of both cores, $BSiO_2$ and diatom concentrations become increasingly decoupled, most likely as a result of species-specific differences in silicification associated with taxonomic shifts (Fig. 3). Thus, neither diatom concentrations nor $BSiO_2$ reveal any directional changes in the most recent sediments of either lake. Although diatom assemblages have clearly changed, in response to changes of either habitat or water chemistry, the biogeochemistry of silica has remained comparatively complacent.

SEDIMENT ORGANIC CONSTITUENTS

In samples from both lakes, LOI and %N are highly correlated ($r^2 = 0.89$; $n = 40$; $P < 0.01$), indicating that sediment N is organic in origin. In CF-10, where C was measured separately, LOI and %C are even more strongly correlated ($r^2 = 0.94$; $n = 11$; $P < 0.01$), confirming the complete absence of carbonate minerals. In both lakes, LOI and %N increase sharply in sediments postdating 1950, reaching peak values at the surface (Fig. 4). LOI and %N values are two to three times greater in CF-10 than in CF-11. This indicates that organic matter is largely responsible for the higher sediment accumulation rates observed in CF-10 (Fig. 2), and that the small glacier in its catchment does not contribute appreciable quantities of inorganic sediment. In CF-11, the stratigraphies of LOI and N parallel those of diatom valve concentrations and $BSiO_2$ in pre-1900 sediments, suggesting a close association between diatom production and organic matter sedimentation at this time. In CF-10, these proxies track each other to a somewhat lesser extent, but in both lakes there is evidence for a decoupling of $BSiO_2$ and organic matter in late-20th century sediments (Fig. 4). This likely implies a progressive increase of organic matter attributable to nondiatom sources such as soft-bodied algae. Although the possibility exists that the recent increases in organic content are transient artifacts due to incomplete organic matter decomposition, many lakes on Baffin Island preserve highly organic sediments of early Holocene age, with minimal indication of post-depositional organic matter degradation (Miller et al., 2005). For this reason, we retain that recent increases of sediment organic matter (LOI, %N, and %C) reflect real limnological changes, namely that organic matter production and sequestration have increased sharply in recent decades.

Sediment $\delta^{15}N$ values range between 1.9 and 3.7‰ before 1950, once again tracking down-core changes in diatom concentrations and $BSiO_2$ in both lakes, as well as LOI and N in CF-11 (Fig. 4). In sharp contrast to the early portions of the records, marked depletions of $\delta^{15}N$ characterize sediments deposited since 1950 in both lakes, with mean values for this time interval of 1.37 ± 0.26‰ in CF-10 and 1.22 ± 0.31‰ in CF-11. The relationship between sediment $\delta^{15}N$ and %N is represented graphically (Fig. 5). Low $\delta^{15}N$ values (<1.7‰) associated

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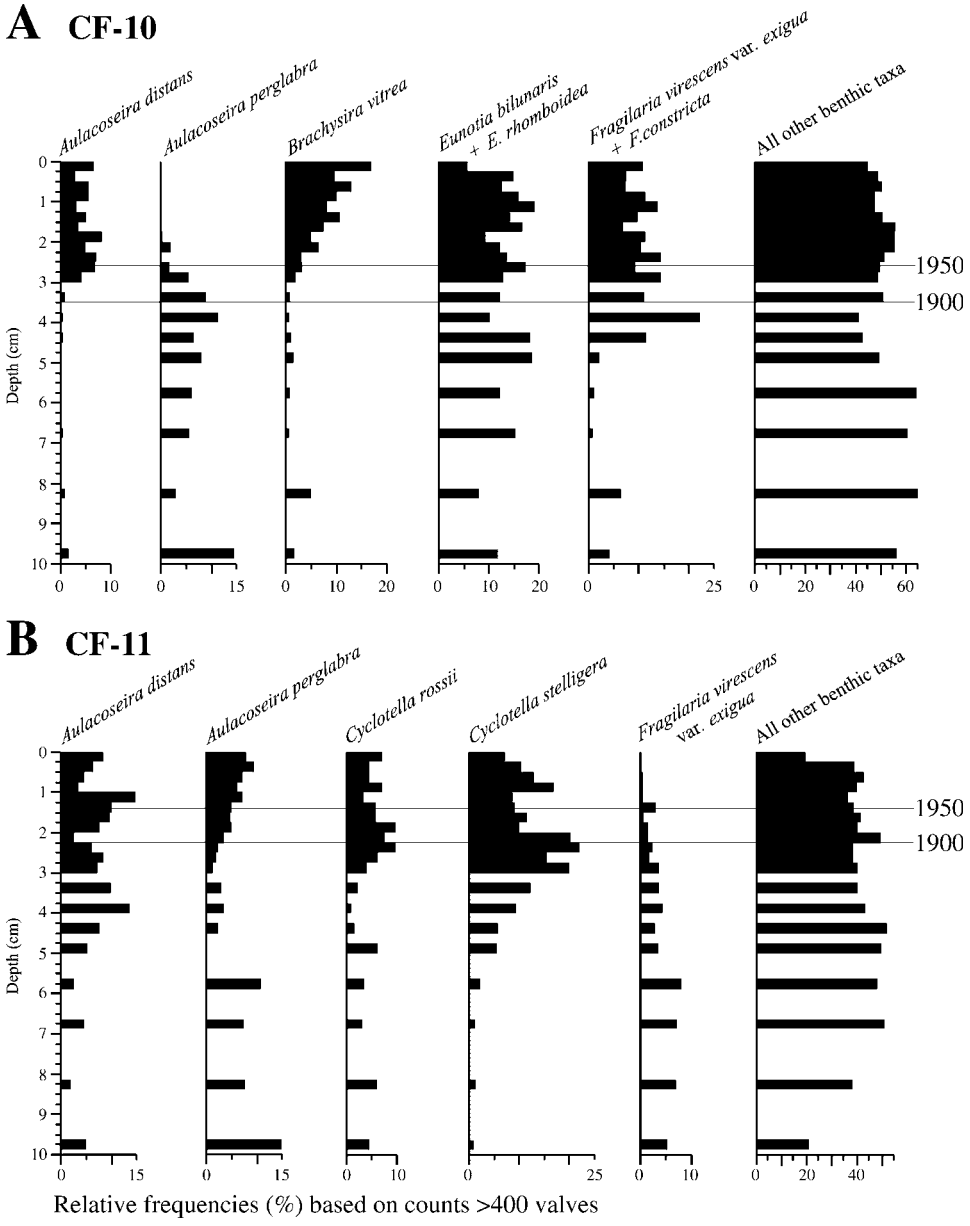


FIGURE 3. Relative frequencies of selected diatom taxa from the CF-10 (A) and CF-11 (B) cores.

with the most recent sediments from both lakes have N concentrations that range broadly from 0.2 to 1.2%, forming a discrete population of samples that is distinct from older sediments spanning a larger range of $\delta^{15}N$ (1.7–3.6‰) at the low end of N concentrations (<0.3%). Consequently, linear correlations between $\delta^{15}N$ and N are much stronger within these these two populations ($r^2 = 0.71$ and 0.89 for pre- and post-1950 samples, respectively) than for the complete data set ($r^2 = 0.36$).

In the CF-10 core, $\delta^{13}C$ values range from –25 to –23‰, and reveal stratigraphic changes that broadly parallel those of diatom concentrations, $BSiO_2$, and $\delta^{15}N$ prior to 1950 (Fig. 6). This supports the first-order interpretation that, over this interval, these four proxies are primarily modulated by lake primary production, of which diatoms represent a major component. In contrast, after 1950 the C and N isotope stratigraphies diverge, with $\delta^{13}C$ becoming enriched by nearly 2‰ in synchrony to the depletion of $\delta^{15}N$ outlined above. It is noted that comparably enriched $\delta^{13}C$ values (~–23‰) are also recorded prior to 1900, implying that the 20th century C-isotopic increase is not as exceptional in the lake's history as the negative excursion of $\delta^{15}N$ (Fig. 6). However, only the most recent high $\delta^{13}C$ values are accompanied by declining sediment C:N molar ratios, which indicate enhanced autochthonous contributions to sediment organic matter (Wolfe et al., 2002, 2003).

PRINCIPAL COMPONENTS ANALYSIS

The above results have been summarized by extracting the first axes of PCA and arranging the resulting sample scores stratigraphically (Fig. 7). This provides objective overviews of the major biological and biogeochemical trajectories of both lakes. Diatom PCA results indicate that 42.6 and 37.3% of variance is explained by the first axis in CF-10 and CF-11, respectively. It is noted that a far greater number of taxa contribute to these results than those illustrated (Fig. 3, Appendix 1). The greater floristic richness of the CF-11 core (47 taxa) accounts for a slightly weaker PCA axis 1 eigenvalue. Diatom PCA axis 1 scores from both lakes show sustained inflections that are initiated somewhat before 1900, and are especially well expressed in CF-11. In the uppermost sediments of both cores, this trend is partially reversed, suggesting a second phase of community change (Fig. 7A). In contrast, PCA axis 1 sample scores based on the combination of biogeochemical

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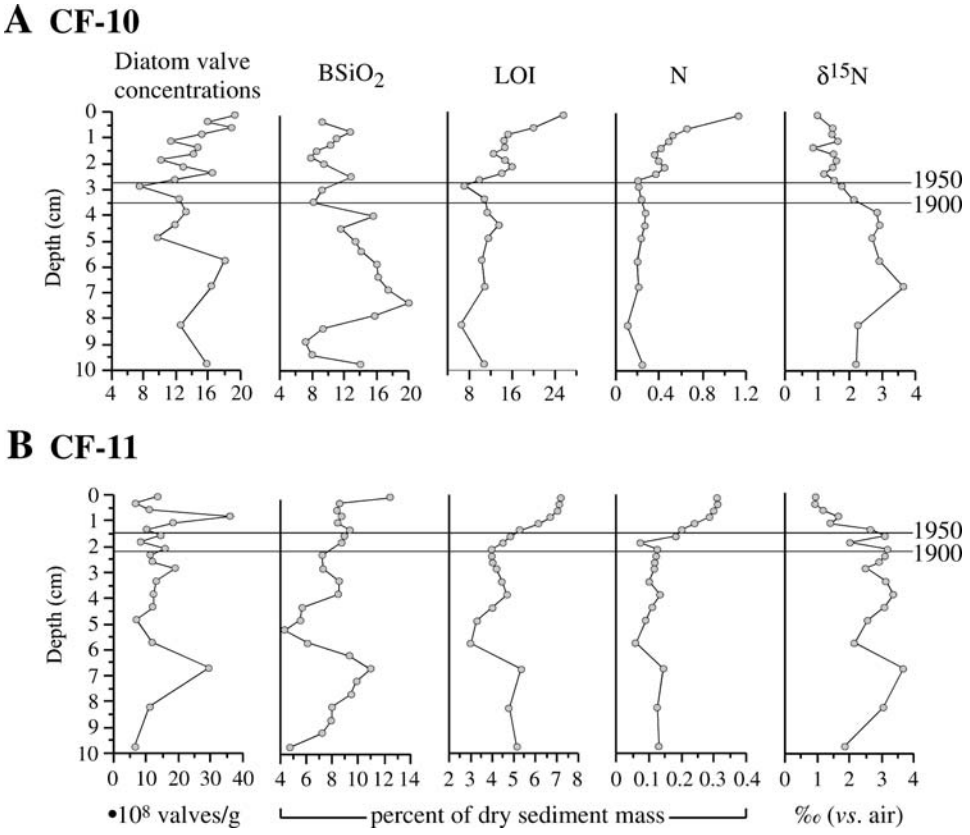


FIGURE 4. Stratigraphies of diatom concentrations and biogeochemical proxies from the CF-10 (A) and CF-11 (B) cores.

proxies and diatom concentrations from both lakes (53.0% variance explained) rise above background variability only after 1950, and do not show any reversal of trend in the uppermost sediments (Fig. 7B).

Discussion

These PCA analyses, and the raw data upon which they are derived, reveal similar recent paleolimnological histories for both lakes, despite their contrasting catchments and somewhat different diatom floras. This degree of coherence implies that regional inferences concerning environmental change may be advanced. These elements of similarity include relatively subtle features that are only revealed by the high stratigraphic resolution of the various analyses presented here. Foremost among these is the observation that diatom assemblage composition began to change directionally before any likely anthropogenic effects to the region (i.e., pre-1900), but that pronounced changes in lake biogeochemistry have occurred in subsequent decades, primarily since 1950. Our preferred interpretation of these results is that they represent two suites of limnological responses, the first towards climate change, and the second in relation to anthropogenic nitrogen deposition. We discuss the diatom and biogeochemical results sequentially below, prior to synthesizing how this interpretation is achieved.

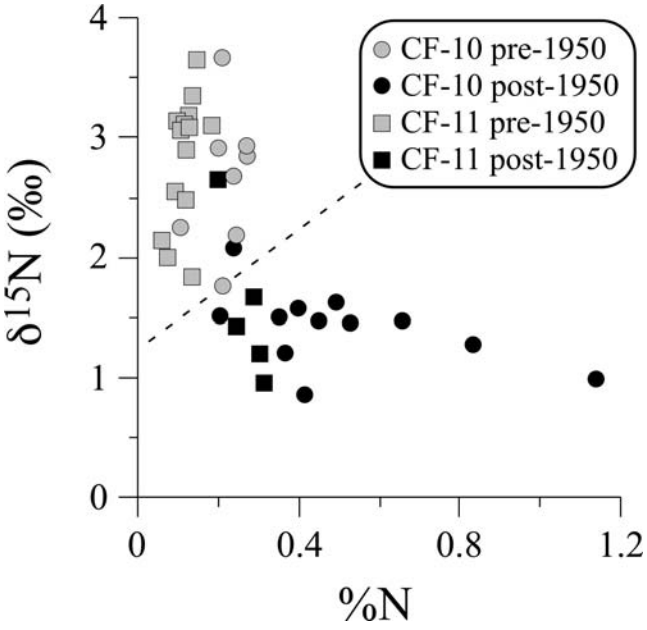


FIGURE 5. Relationship between sediment $\delta^{15}N$ and %N in the CF-10 and CF-11 cores, arranged as samples deposited before (gray symbols) and after (black) 1950. The dashed line differentiates two apparently distinct sample populations.

DIATOM COMMUNITIES AND RECENT CLIMATE CHANGE

Diatom communities in dilute Baffin Island lakes have been shown to respond sensitively to climate gradients in both space (Joynt and Wolfe, 2001) and time (Wolfe, 2003). However, these relationships are unlikely to reflect a direct ecophysiological control of temperature over diatoms, but rather indirect responses to limnological parameters mediated by climate, such as lake ice (Smol, 1988) and dissolved inorganic carbon dynamics (Wolfe, 2002). Diatom PCA axis 1 scores, which reflect the overall stability of community structure, correspond well to the composite arctic summer temperature reconstruction of Overpeck et al. (1997; Fig. 7C), supporting the general sensitivity of diatom assemblages to climate variability on decadal to centennial timescales. Significant warming of the Arctic began around 1850, at the close of the Little Ice Age, which itself was likely the coldest interval of the mid- to late Holocene in many regions (Bradley, 2000; Kaplan et al., 2002). Although the 20th century is regarded as

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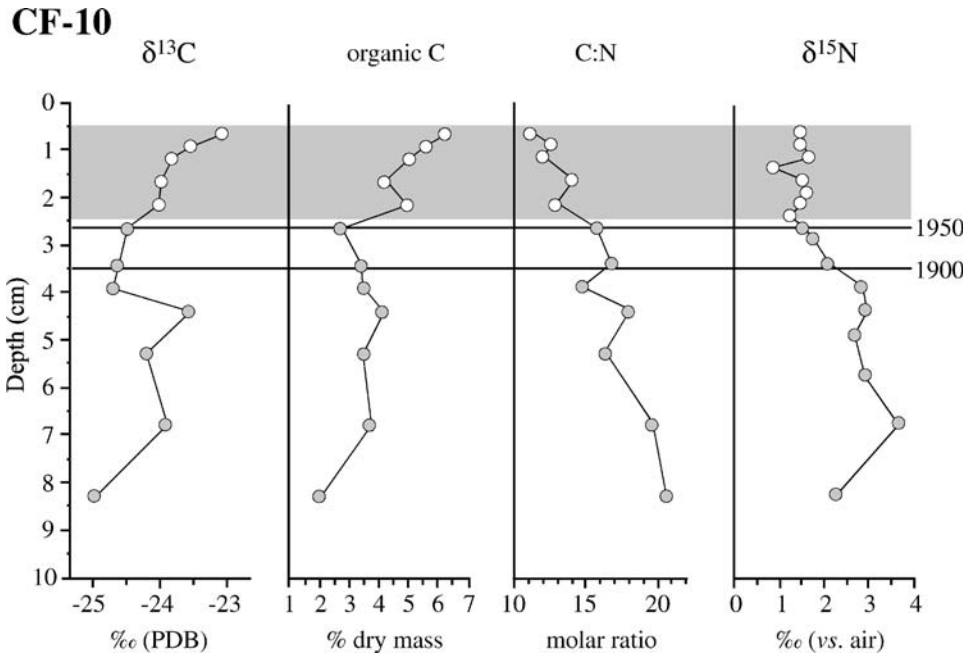


FIGURE 6. Carbon isotopic ratios ($\delta^{13}C$), organic C content, and C:N molar ratios from the CF-10 core, shown alongside $\delta^{15}N$ for comparison. The shaded region indicates the zone of divergence between $\delta^{13}C$ and $\delta^{15}N$ trends.

the warmest of at least the last 400 yr (Overpeck et al., 1997), a distinct cooling of the Labrador Sea region since approximately 1960 is superimposed on this overall trend. This regional cooling, in the order of 0.4°C (sea surface) to 1.3°C (west Greenland coast) between 1958 and 2001, is attributed to the positive phase of the North Atlantic Oscillation (Hanna and Cappelen, 2003). This second-order trend is apparent in the available instrumental temperature record from Baffin Island (Serreze et al., 2000), and can also be discerned in the composite record of summer temperature proxies (Fig. 7C). Moreover, PCA scores from the diatom assemblages in the uppermost 1 to 2 cm of both cores considered here show inflections in the opposite direction to earlier community shifts that we attribute to warming (Fig. 7A). On the other hand, PCA scores based on the sediment geochemical and diatom concentration data (Fig. 7B) preserve neither pre-1900 changes associated with post-Little Ice Age warming, nor any relaxation in recent decades. This supports the notion that the observed late-20th century biogeochemical changes are at least partially attributable to nonclimatic processes.

DEVIATIONS OF THE LACUSTRINE NITROGEN CYCLE

The stratigraphic trends of $BSiO_2$, LOI, and $\delta^{15}N$ are broadly parallel in pre-1950 sediments from both study lakes (Fig. 4). The coupling of these same parameters has been noted elsewhere in the

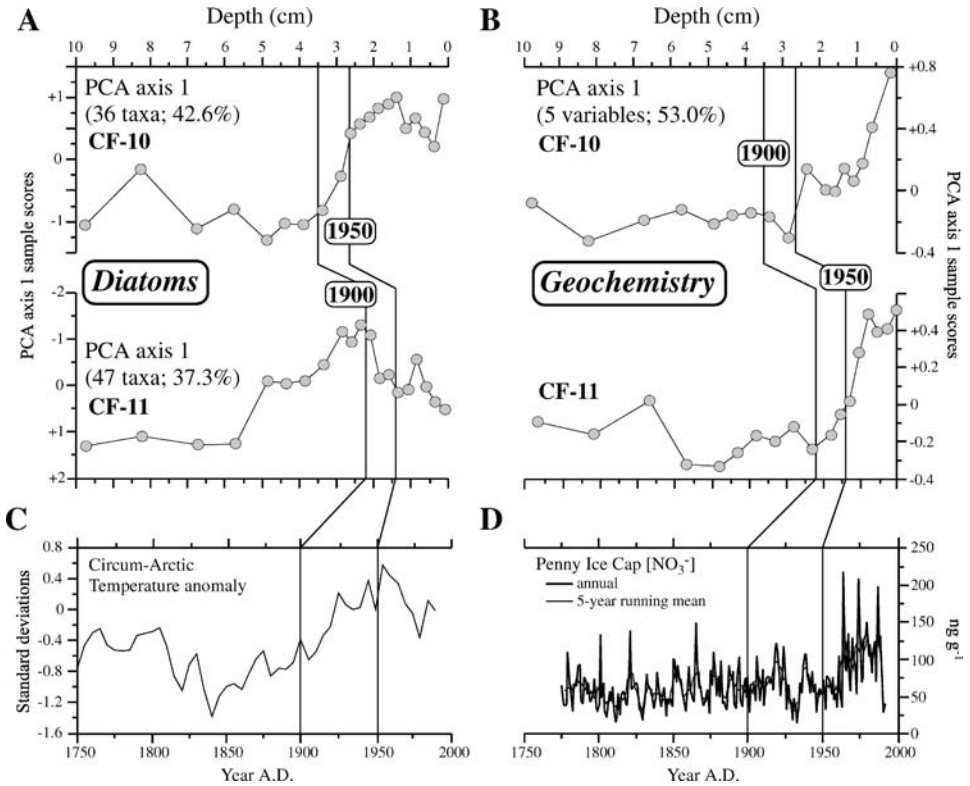


FIGURE 7. Stratigraphic evolution of PCA axis 1 sample scores summarizing major changes of both diatom assemblages (A) and sediment geochemistry (B) in the CF-10 and CF-11 cores. For comparison, these results are shown in relation to (C) the multi-proxy composite of pan-arctic summer temperature departures from Overpeck et al. (1997), and (D) the nitrate concentration record from the Penny Ice Cap, approximately 400 km to the southeast of Clyde River (Goto-Azuma et al., 2002).

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Arctic on longer (Holocene) timescales (Hu et al., 2001), suggesting that N isotopes are potentially sensitive to pre-industrial changes in lake productivity. This is because N-isotopic enrichment accompanies elevated production as a consequence of either increased rates of aquatic N cycling (Gu et al., 1996), or weakened physiological fractionation against ^{15}N during DIN uptake when N limitation becomes exacerbated by elevated primary production (Goericke et al., 1994). On these grounds, it may therefore be predicted that recent increases of organic matter sedimentation (Fig. 4) should be accompanied by enriched $\delta^{15}N$ values, which is opposite to the observed trends of post-1950 N-isotopic depletion.

However, before these recent $\delta^{15}N$ excursions can be interpreted in terms of biogeochemical change, the potential effects of post-depositional processes must be carefully evaluated. The most conspicuous early-diagenetic process is sediment denitrification, which strongly favors ^{14}N and is thus predicted to leave the remaining substratum enriched in ^{15}N (Hodell and Schelske, 1998; Talbot, 2001). Once again, this is opposite to the trends of progressive $\delta^{15}N$ depletion reported here. Furthermore, if denitrification were progressively fractionating significant proportions of the organic N inventory from these sediments and thus regulating their $\delta^{15}N$, a near-linear relationship should evolve between $\delta^{15}N$ and N concentrations, which is clearly not the pattern observed (Fig. 5). Controlled-environment experiments under both oxidizing and reducing conditions indicate that most diagenetic overprinting of sediment $\delta^{15}N$ occurs within weeks to months (Lehmann et al., 2002). The trends reported here are expressed on timescales of several decades, which further supports their interpretation as a primary environmental signature. This assessment is therefore consistent with the interpretation of lake sediment $\delta^{15}N$ as capturing, foremost, changing sources of N rather than diagenetic processes, a conclusion reached elsewhere in a variety of paleolimnological investigations (Brenner et al., 1999; Finney et al., 2000; Teranes and Bernasconi, 2000).

Carbon isotopes from CF-10 lend further support to the notion of recent (post-1950) biogeochemical changes involving the N cycle. In a general sense, increased primary production results in the enrichment of sediment $\delta^{13}C$, directly reflecting greater proportions of ^{13}C in the inorganic carbon pool exploited by photosynthetic autotrophs, as the consequence of sustained exploitation of $^{12}CO_2$ under productive regimes. Thus, increased lake production frequently entails parallel enrichments of sediment $\delta^{13}C$ and $\delta^{15}N$ on a variety of timescales (Hodell and Schelske, 1998; Hu et al., 2001). The decoupling of C and N isotope observed in recent CF-10 sediments (Fig. 6) implies a different suite of controlling processes, including the possibility of atmospheric subsidies of isotopically-depleted N species.

ANTHROPOGENIC NITROGEN DEPOSITION IN THE CANADIAN ARCTIC

We consider the recent $\delta^{15}N$ excursions observed in both lakes (Fig. 4), coupled to the decoupling of $\delta^{15}N$ from $\delta^{13}C$ trends in CF-10 (Fig. 6), to be strong indications that inputs of anthropogenic N are detectable in the sediments of the two investigated lakes. This is because anthropogenic sources, with the exclusion of waste-water effluent and animal manure, are generally isotopically-depleted (Macko and Ostrom, 1994; Talbot, 2001). For example, NH_3 from coal combustion has an $\delta^{15}N$ range from 0 to $-10‰$ (Heaton, 1986), notwithstanding kinetic fractionation effects during atmospheric transport and oxidation to NO_3^-, in addition to photochemical processes that may further deplete the $\delta^{15}N$ of far-field NO_3^- in arctic precipitation (Hastings et al., 2004; Heaton et al., 2004). The chronology of the N-isotopic excursions reported here, coupled to the unlikelihood that they represent diagenetic artifacts (Fig. 5), augment the interpretation of an anthropogenic origin. Indeed, in their timing, amplitude, and direction, these isotopic shifts directly comparable to those reported from lakes in Colorado (Wolfe et al., 2003) and upstate New York (Owen et al., 1999), two regions that are demonstrably impacted by atmospheric N pollution.

An alterative possibility for the N-isotopic results, borne partially out of the relatively high DIN concentrations measured in the epilimnia of the study lakes (Table 1), is that nitrogen limitation has become alleviated in recent decades, allowing lake autotrophs to fractionate more strongly against ^{15}N, and resulting in the progressive depletion of $\delta^{15}N$ in sedimenting autochthonous organic matter. This possibility should be tested more explicitly, for example by ascertaining whether or not excess DIN (i.e., N saturation) arises in streams flowing from the lakes in mid-summer, when algal production reaches its annual maximum. However, physiological fractionation effects are rarely well-expressed in nature (Goericke et al., 1994), and so we consider this a secondary interpretation of the $\delta^{15}N$ excursions. Despite the recent $\delta^{13}C$ enrichment observed in CF-10 (Fig. 6), evidence for coeval increases in diatom production in either lake is equivocal, based on trends in $BsiO_2$, valve concentrations, and assemblage composition. This contrasts alpine lakes in which N deposition has induced distinctive shifts towards mesotrophic diatom taxa (Wolfe et al., 2001, 2003; Saros et al., 2003, 2005).

As an exploratory attempt to separate these potential influences on sediment $\delta^{15}N$, the PCA of biogeochemical data and diatom concentrations was alternately restricted to samples pre- and postdating 1950. For these analyses, we illustrate loadings of the five equally weighted variables as vectors on the first two PCA axes, for both time frames (Fig. 8). The overall structure of the PCA remains generally similar over both intervals, with the striking exception of $\delta^{15}N$, for which the gradient direction changes dramatically, particularly in its relation to $BSiO_2$. This pronounced deviation supports the contention that a source change is the primary control over sediment $\delta^{15}N$. Furthermore, in the post-1950 analysis, $\delta^{15}N$ becomes nearly anti-correlated with three proxies associated with lake primary production, $BSiO_2$, LOI, and %N (Fig. 8B). This is not the predicted association if $\delta^{15}N$ was modulated by DIN uptake dynamics and Rayleigh fractionation (Hodell and Schelske, 1998), or an intensification of internal N cycling (Hu et al., 2001). It remains that, whether recent depletions of sediment $\delta^{15}N$ are interpreted as a signature of isotopically depleted N sources, or alternatively as evidence for incomplete DIN assimilation (N saturation), the primary conclusion of increased N availability from anthropogenic sources remains equally valid.

Glaciochemical time-series are highly relevant for comparative purposes, and we focus on results from the Penny Ice Cap on Cumberland Peninsula, which is the closest record of long-term atmospheric N deposition to our study lakes. In this ice core, NO_3^- concentrations begin to increase around 1960 and remain elevated thereafter (Fig. 7D). Most of this NO_3^- appears to originate from emissions in eastern North America (Goto-Azuma et al., 2002). Parallel increases of N availability in lakes regionally therefore seem a likely, if not predictable, consequence of the long-distance atmospheric deposition history evidenced by the ice core record. Lake sediment isotopic shifts that potentially fingerprint anthropogenic sources are also compatible with this interpretation, as are recent $\delta^{15}N$ depletions of more than $5‰$ in NO_3^- from the Greenland Ice Sheet (Freyer et al., 1996).

The 1945–1982 mean NO_3^- deposition rate to the Penny Ice Cap is estimated to be 0.28 kg ha^{-1} yr^{-1} (Goto-Azuma and Koerner, 2001). However, this must be viewed as a conservative low-end value, because revolatilization of NO_3^- from firn, coupled to potential losses from both meltwater percolation and wind scouring, are widespread phenomena (Goto-Azuma et al., 2002). We therefore estimate that current N deposition rates on eastern Baffin Island lie somewhere

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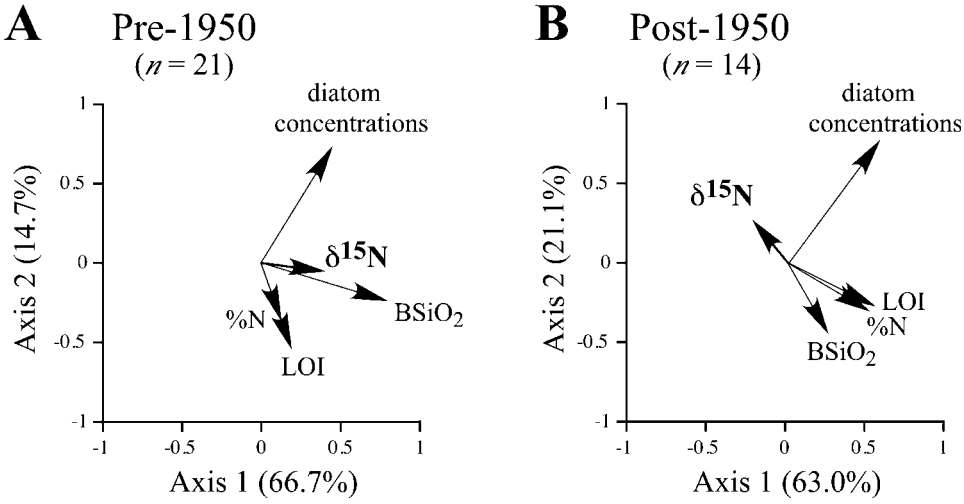


FIGURE 8. Vectors of PCA loadings on the first two axes for the four biogeochemical proxies and diatom concentrations, in two separate analyses restricted to samples from both lakes predating (A) and postdating (B) the year 1950 as defined by ^{210}Pb chronology.

between several-fold to one order of magnitude above natural background fluxes, based on comparisons with other remote regions (Galloway et al., 1982). In catchments that are steep and unvegetated (CF-10), or small relative to lake area (CF-11), the potential for terrestrial retention of atmospherically derived N appears minimal. Furthermore, arctic soils remain frozen when snowmelt is initiated, which further limits the interception of N derived from what is presumed to represent the peak annual flux of anthropogenic N to surface waters. Unfortunately, at present we are unable to fully address the importance of terrestrial processes in N cycling within lake catchments on Baffin Island, while acknowledging their potential significance with respect to both organic and inorganic N pools, even in talus-dominated basins (Williams et al., 1997). However, $\delta^{15}N$ values of microbially cycled NO_3^- in talus waters retain a dominantly atmospheric signature (Campbell et al., 2002), implying that our interpretation of N-isotopic shifts are unlikely to be greatly modified by catchment cycling processes.

It has been demonstrated that bryophyte-dominated arctic tundra ecosystems encounter a threshold for change in ecosystem structure, or critical load, at N deposition rates between 5 and 10 kg N ha^{-1} yr^{-1} (Gordon et al., 2001). Our paleolimnological results, viewed in light of the Penny Ice Cap NO_3^- time-series (Fig. 7), suggest that lakes on Baffin Island may have critical loads for N deposition that are considerably lower. This is consistent with the general assertion that arctic lakes are bellwethers for environmental change (Douglas et al., 1994). Moreover, the combined effects of climate change and atmospheric deposition is likely to induce synergistic limnological responses with consequences that are only beginning to be explored (Schindler, 2001).

Conclusions

Human activities emit more fixed nitrogen globally (140–160 Mt yr^{-1}) than the sum total of natural fixation processes (Galloway et al., 2003). This rate of emission continues to rise with population growth and increased industrialization. In this context, it is perhaps not surprising that lake sediments from remote arctic regions appear to record biogeochemical shifts that are consistent with N pollution. Increased organic matter sequestration is the first apparent consequence observed, superimposed on dynamic species changes that relate more closely to underlying climate variability (Smol et al., 2005). Collectively, these impacts have produced new ecological states lacking any prior natural analog. Although limnological changes in the Arctic are almost certain to be sustained in the future, the exact nature of future ecological trajectories remains uncertain. The acidification potential arising from enhanced deposition of N oxides cannot be ignored, especially since trends of anthropogenic SO_4^{2-} either match, or occasionally predate, those of NO_3^- across the Arctic (Goto-Azuma and Koerner, 2001). Despite the focus of this paper, N should not be considered the sole component of atmospheric deposition that is potentially relevant to arctic limnological regimes, nor should atmospheric deposition and climate change be viewed as independent of each other. For example, the progressive melting of perennial snowpacks and glaciers as climate continues to warm will release their cumulative inventories of postindustrial anthropogenic pollutants into surface waters, irrespectively of future atmospheric deposition rates.

Disentangling the effects of multiple and potentially synergistic stressors is not an easy task in remote arctic regions, due to difficult access and challenging field conditions. Paleolimnology offers one strategy to approach these questions, although very low sediment accumulation rates, coupled to limited background limnological data, are persistent problems. Clearly, additional biogeochemical studies of arctic catchments are warranted. The results presented here nonetheless confirm the ecological and biogeochemical sensitivity of these ecosystems, and verify that they should not be considered pristine. It is our view that anthropogenic nitrogen deposition is presently contributing to the complex suite of changes observed in many arctic lakes. Given current trends in global biogeochemical cycles (Falkowski et al., 2000), the magnitude of these changes is likely to become amplified in the future.

Acknowledgments

This research was supported by the Natural Sciences and Engineering Research Council of Canada (Discovery Grant to APW), the National Science Foundation of the USA (awards to Gifford Miller), and the University Centre in Svalbard (Norway). A large number of colleagues have contributed to the data and ideas presented here, including Jason Briner, Roy Coulthard, and Gifford Miller (fieldwork); Derek Muir and Fan Wang (sediment dating); David Harris and Karlis Muehlenbachs (stable isotopes); Dan Conley and Mingshen Ma (biogenic silica); Lauren Blackburn, Kris Hadley, and Neal Michelutti (diatoms); David Fisher, Kumiko Goto-Azuma, and Christian Zdanowicz (ice-core nitrate records); and lastly two journal reviewers for comments that substantially improved the manuscript. This work would not have been possible without the generosity of these individuals and organizations. This paper is dedicated to the memory of J. Platt Bradbury, whose influence on the field of paleolimnology is truly immeasurable.

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APPENDIX 1

Names, authorities and summary statistics for all diatom taxa in the CF-10 and CF-11 cores that were included in the PCA analyses. Dashes indicate absence.

Taxon	CF-10			CF-11		
	Minimum frequency (%)	Maximum frequency (%)	Number of occurrences	Minimum frequency (%)	Maximum frequency (%)	Number of occurrences
Achnanthes altaica (Poretzky) Cleve-Euler 1953	—	—	—	0.00	7.92	12
Achnanthes carissima Lange-Bertalot *in* Lange-Bertalot & Krammer 1989	—	—	—	0.00	8.46	17
Achnanthes conspicua A. Mayer 1919	0.00	1.50	2	—	—	—
Achnanthes helvetica var. *minor* Flower & Jones 1989	6.03	18.80	20	—	—	—
Achnanthes levanderi Hust. 1933	6.75	18.55	20	—	—	—
Achnanthes saccula Carter *in* Carter & Watts 1981	—	—	—	2.48	16.34	20
Achnanthes sp. 1	0.00	2.24	14	0.00	1.75	10
Achnanthes sp. 2	—	—	—	0.50	3.66	20
Aulacoseira distans (Ehr.) Simonsen 1979	0.00	8.25	18	1.99	14.93	20
Aulacoseira perglabra (Oestrup) Haworth 1988	0.00	14.57	16	0.25	10.92	20
Aulacoseira sp.	0.00	1.00	3	0.00	2.49	8
Brachysira arctoborealis Wolfe & Kling 2001	0.00	3.55	17	—	—	—
Brachysira brebissonii Ross *in* Hartley 1986	0.50	7.82	20	—	—	—
Brachysira vitrea (Grun.) Ross *in* Hartley 1986	0.50	17.00	20	—	—	—
Brachysira spp.	0.00	1.71	17	—	—	—
Cavinula cocconeiformis (Greg. *ex* Greville) Mann & Stickle 1990	—	—	—	3.50	12.28	20
Cavinula pseudoscutiformis (Hust.) Mann & Stickle 1990	—	—	—	1.00	5.26	20
Cyclotella bodanica var. *lemanica* (O. Müller *ex.* Schroter) Bachmann 1903	—	—	—	0.00	3.98	16
Cyclotella rossii Håkansson 1990	—	—	—	1.00	9.75	20
Cyclotella stelligera (Cleve & Grun. in Cleve) Van Heurck 1882	—	—	—	0.74	21.75	20
Cymbella gaeumannii Meister 1934	0.00	2.25	18	—	—	—
Cymbella helvetica Kütz. 1844	0.00	1.00	2	0.00	2.75	17
Cymbella herbridica (Grunow *in* Cleve) Cleve 1894	0.00	5.76	7	0.00	1.50	15
Cymbella sp.	0.00	1.00	7	0.00	1.25	11
Eunotia bilunaris (Kütz.) Lagerst. 1884	1.71	11.25	20	—	—	—
Eunotia exigua (Bréb. *ex* Kütz.) Rabenh. 1864	—	—	—	0.00	1.25	3
Eunotia pectinalis var. *minor* (Kütz.) Rabenh. 1864	0.00	1.00	8	0.00	2.49	9
Eunotia praerupta Ehr. 1843	0.00	1.99	16	0.00	4.21	11
Eunotia rhomboidea Hust. 1950	2.50	15.04	20	—	—	—
Eunotia triodon Ehr. 1837	0.00	3.76	15	—	—	—
Eunotia sp.	0.00	1.49	10	0.00	1.74	6
Fragilaria constricta Ehr. 1843	0.50	10.25	20	—	—	—
Fragilaria vaucheriae var. *capitellata* (Grun. *in* Van Heurck) Ross 1947	—	—	—	0.00	2.43	6
Fragilariaforma exiguiformis (Grunow *in* Van Heurck) Jones & Flower 1996	0.50	14.25	20	0.00	8.01	18
Fragilariaforma virescens (Ralfs) Williams & Round 1988	—	—	—	0.00	1.74	5
Frustulia rhomboides var. *saxonica* (Rabenh.) De Toni 1891	2.26	12.72	20	0.00	2.50	12
Frustulia sp.	—	—	—	0.00	2.23	13
Navicula peregrina (Ehr.) Kütz. 1844	—	—	—	0.49	4.70	20
Navicula radiosa var. *tenella* Lange-Bertalot 1985	0.00	1.00	12	—	—	—
Navicula schmassmannii Hust. 1937	0.00	1.00	8	3.22	17.24	20
Navicula variostriata Krasske 1923	—	—	—	0.00	4.70	19
Navicula sp.	0.00	1.00	2	0.00	3.00	4
Neidium iridis (Ehr.) Cleve 1894	0.00	3.76	18	0.00	3.05	13
Nitzschia gracilliformis Lange-Bertalot & Simonsen 1978	—	—	—	0.50	6.73	20
Nitzschia palea (Kütz.) W. Sm. 1856	—	—	—	0.00	4.50	18
Nitzschia sp.	—	—	—	0.00	1.50	10
Nupela sp.	0.00	1.24	4	0.50	5.97	20
Pinnularia biceps Greg. 1856	1.25	8.54	20	0.00	2.50	16
Pinnularia brebissonii (Kütz.) Rabenh. 1864	—	—	—	0.00	2.26	15
Pinnularia microstauron (Ehr.) Cleve 1891	0.00	1.00	1	0.00	2.76	13
Pinnularia mesolepta (Ehr.) W. Smith. 1853	—	—	—	0.00	2.48	8
Pinnularia sp.	0.00	1.75	7	0.00	4.00	16
Pseudostaurosira brevistriata Williams & Round 1987	0.00	1.76	1	—	—	—
Stauroneis anceps Ehr. 1843	—	—	—	0.50	4.26	20
Stauroneis anceps fo. *gracilis* Rabenh. 1864	—	—	—	0.00	1.75	12
Stauroneis kriegeri Patrick 1945	0.00	3.51	18	0.00	1.25	19
Staurosirella pinnata (Ehr.) Williams & Round 1987	—	—	—	0.00	3.71	14
Surirella ovalis Hust. 1965	0.00	1.00	4	0.00	4.48	14
Surirella sp.	—	—	—	0.00	1.74	6
Synedra ulna (Nitzsch) Ehr. 1836	—	—	—	0.00	3.49	11
Tabellaria flocculosa (Roth) Kütz. 1844	8.25	26.00	20	0.00	1.75	19

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