Climatically controlled chemical and biological development in Arctic lakes

Neal Michelutti,^{1,2} Alexander P. Wolfe,¹ Jason P. Briner,³ and Gifford H. Miller⁴

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[1] We investigated the factors controlling lake evolution in Arctic ecosystems using a multiproxy paleolimnological approach on a small lake on Baffin Island, Arctic Canada. Lakewater pH was inferred from fossil diatom assemblages, whereas primary production was assessed from sediment concentrations of diatom valves and spectrally inferred chlorophyll *a*. Our reconstructed limnological variables registered synchronous changes and showed a close coupling to Holocene climatic fluctuations, as inferred by numerous independent paleoclimate proxies. Without exception, our highest pH and production values occurred during warm intervals, and vice-versa. A return towards paleolimnological conditions of the warm early Holocene has occurred since the midtwentieth century, corresponding to climate warming following the Little Ice Age. Maximum recent values of our reconstructed parameters are either directly comparable to, or in some cases exceed, values attained during the Holocene Thermal Maximum, 8000–10,000 years ago. Our data suggest that climate has a first-order influence on primary production and the regulation of in-lake DIC dynamics (and hence on lakewater pH) through its modulation of lake ice cover. We conclude that direct forcing by climate is more important than catchment processes in controlling the chemical and biological development of ice-dominated Arctic lake ecosystems, at the scale of the Holocene.

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1. Introduction

[2] Lake ontogeny, which is the limnological counterpart to successional development in terrestrial ecosystems, has long fascinated limnologists because it offers the basis of a general ecological theory and hence a predictive tool for long-term changes in lacustrine systems [Deevey, 1984]. Paleolimnology has been central to the development of lake ontogeny models, largely because it is the primary empirical means of obtaining information concerning processes operating on centennial to millennial timescales. The earliest models based on sedimentary archives proposed the progressive eutrophication of lakes with age [Deevey, 1942]. Subsequent models suggested that Holocene production and organic matter accumulation follow a sigmoidal ontogenetic trajectory, culminating in either paludification or eutrophication [Binford *et al.*, 1983]. More recently, the general view from boreal landscapes glaciated during the Late Pleistocene is that lakes progressively acidify and become

more oligotrophic, following a transient interval of relatively high production [Engstrom *et al.*, 2000].

[3] In all of these models of lake ontogeny, the role of catchment processes has been deemed critical. For example, base cation sequestration from unweathered glacial deposits is typically invoked as the source of early postglacial alkalinity [Mackereth, 1966; Whitehead *et al.*, 1989; Ford, 1990; Renberg, 1990]. As terrestrial vegetation and soils progressively develop, catchment-supplied dissolved organic carbon (DOC) and nutrients such as nitrogen (N) become increasingly important in driving lake ontogeny. This highlights the intimate linkage between lakes and the successional changes occurring in catchment soils, vegetation, and hydrology. These processes have been illustrated by lacustrine chronosequences spanning boreal landscapes of different age in southern Alaska [Engstrom *et al.*, 2000]. However, far less is known concerning Holocene lake ontogeny in the Arctic where, due to the much sparser tundra vegetation, minimal soil development, continuous permafrost, and persistent lake ice, we hypothesize that the catchment's role in lake development will be severely curtailed.

[4] Here, we use paleolimnology to reconstruct the chemical and biological development of a small lake on Baffin Island in the Canadian Arctic since its inception ~11,000 years ago. Lakewater pH is inferred using a diatom-based transfer function, and whole-lake production is assessed from sedimentary concentrations of diatom valves and spectrally inferred chlorophyll *a* (Chl *a*). These data are presented at sufficient temporal resolution (decadal

¹Department of Earth and Atmospheric Sciences, University of Alberta, Edmonton, Alberta, Canada.

²Now at Department of Biology, Queen's University, Kingston, Ontario, Canada.

³Department of Geology, State University of New York, Buffalo, New York, USA.

⁴Department of Geological Sciences and Institute of Arctic and Alpine Research, University of Colorado, Boulder, Colorado, USA.

Figure 1. Map showing the location of Lake CF3 on Baffin Island, Nunavut. Black circles are locations of the 60 lakes used to develop the diatom-based pH inference model [Joynt and Wolfe, 2001]. These training set sites extend at least 400 km in a north-south transect from Lake CF3, in a strategy aimed to capture, within modern climate space, the range of possible conditions encountered at Lake CF3 over the Holocene.

to centennial throughout) to clearly identify linkages between chemical and biological patterns, and to place recent limnological changes associated with the ‘Holocene-Anthropocene’ transition in the context of the prior 11 millennia of natural variability. In so doing, we propose a model of lake development for Arctic lakes that is driven, foremost, by climate.

2. Methods

2.1. Site Description

[5] Lake CF3 (70° 31.951’N, 68° 22.047’W) is a small (0.2 km²), shallow ($Z_{\max}$ = 7.2 m) tundra lake located near the hamlet of Clyde River on east-central Baffin Island, Nunavut Territory, Arctic Canada (Figure 1). The lake occupies a coastal lowland (27 m a.s.l.) within a small catchment of 0.6 km² that has no permanent inflow or outflow. Lakewater chemistry, from collections obtained in May 2003 and 2005, indicates that the lake is oligotrophic, dilute, and slightly acidic (Table 1). As these water samples were recovered from beneath ~2 m of ice, they reflect limnological conditions of Lake CF3 during late winter, as opposed to summer conditions upon which our paleolimnological inferences are based.

[6] Climate at Clyde River, based on 1971–2000 normals, is categorized as mid-Arctic, with mean annual and July temperatures of –12.83°C and +4.43°C, respectively, and mean annual precipitation of 233 mm yr^{–1}, of which over 80% falls as snow. Accordingly, extended periods of snow and ice cover characterize lakes in this region, with the open water season lasting less than three months (July–September) in typical years. Present-day vegetation is classified within the

prostrate shrub zone of Edlund and Garneau [2000], including *Salix* spp. and numerous heath taxa (*Cassiope tetragona*, *Empetrum nigrum*, *Vaccinium uliginosum*) amid abundant bryophytes.

[7] Bedrock is composed of Precambrian crystalline lithologies, primarily granite and gneiss. The Clyde forelands surrounding Lake CF3 were covered by the Laurentide Ice Sheet at the Last Glacial Maximum and were deglaciated by ~12 000 yr before present (BP). Cosmogenic exposure dating results and regional terrain mapping indicate that this landscape was glaciated by non-erosive cold-based ice [Briner et al., 2005]. Tills are therefore conspicuously absent and pre-glacial landforms are preserved. The cryptic signature of this glacial style led previous investigations to conclude that the region was not glaciated during the Last Glacial Maximum [Miller et al., 1977].

[8] Lacustrine, marine, and ice-core proxies from the Baffin Bay region reveal a spatially and temporally coherent pattern of Holocene climate change that appears broadly regulated by summer insolation (Figures 2a–2e). Maximum postglacial warmth occurred between ~10 000 and 8000 yr BP, followed by progressive cooling culminating in Neoglaciation and, ultimately the Little Ice Age (1450–1850 A.D.). Early Holocene summer warmth is captured by the amount of melt on the summit of ice caps [Fisher et al., 1995], attendant decreases of sea ice that allowed range expansions of large cetaceans [Dyke et al., 1996], and the isotopic signature of chitin from chironomid larvae in lake sediments [Wooler et al., 2004]. Chironomid faunal assemblages, obtained from the same sediment cores used in this present study, reveal a pronounced summer Holocene Thermal Maximum as much as 5°C warmer than present [Briner et al., 2006] (Figure 2e), in close synchrony with the records illustrated in Figures 2a–2d. Marked warming during the last ~150 years is driven at least partially by anthropogenic greenhouse gas forcing [Overpeck et al., 1997] (Figure 2f). This most recent warming has been sufficient to induce ecological regime shifts within many Arctic lakes, including those of Baffin Island [Smol et al., 2005; Michelutti et al., 2005].

2.2. Sediment Processing and Data Analysis

[9] Our sedimentary analyses were performed on sub-samples from the same cores described by Briner et al. [2006]. The sediment stratigraphy is a composite of two

Table 1. Available Water Chemistry Data for Lake CF3

Variable	30 May 2003	19 May 2005
pH	–	5.89
Conductivity, $\mu\text{S cm}^{-1}$	–	68.90
Chlorophyll <i>a</i> , $\mu\text{g L}^{-1}$	0.98	–
TP, $\mu\text{g L}^{-1}$	6.10	4.60
TDP, $\mu\text{g L}^{-1}$	2.40	1.20
TN, $\mu\text{g L}^{-1}$	349.34	281.03
TDN, $\mu\text{g L}^{-1}$	261.57	236.32
DOC, mg L ^{–1}	3.00	1.73
DIC, mg L ^{–1}	0.96	1.45
Cl [–] , mg L ^{–1}	15.28	15.15
SO ₄ ^{2–} , mg L ^{–1}	2.27	2.21
Na, mg L ^{–1}	7.63	7.42
K, mg L ^{–1}	0.84	0.84
Ca, mg L ^{–1}	1.62	1.39
Mg, mg L ^{–1}	1.45	1.64
Si, mg L ^{–1}	0.26	0.14

Figure 2. Paleoclimate records from the Baffin Bay region including (a) summer insolation anomalies at 65°N [Berger and Loutre, 1991]; (b) percent summer melt record from the Agassiz Ice Cap on Ellesmere Island [Fisher et al., 1995]; (c) frequency distribution of ^{14}C dated bowhead whales in the inter-island channels [Dyke et al., 1996]; (d) chironomid based $\delta^{18}\text{O}$ mean annual temperature record from southwestern Greenland [Wooller et al., 2004]; (e) chironomid-inferred July air temperatures from the Lake CF3 sediment core [Briner et al., 2006]; and (f) inferred circum-arctic summer temperature anomalies for the last four centuries [Overpeck et al., 1997].

cores; a 180 cm core obtained in May 2002 using a sled mounted percussion coring system [Nesje, 1992], and a 21 cm surface core with an intact mud-water interface obtained in May 2003 using a modified Kajak-Brinkhurst gravity device [Glew et al., 2001]. Surface sediment samples were extruded on site into 0.5 cm intervals to 5 cm depth, then 1 cm thereafter. The chronology of the percussion core is based on six accelerator mass spectrometry (AMS) ^{14}C ages on aquatic bryophytes (Table 2), which are demonstrably equilibrated with atmospheric CO_2 in Arctic lakes within granitic basins [Wolfe et al., 2004]. The surface core was dated with a constant-rate-of-supply (CRS) model using excess ^{210}Pb activities (Figures 3a and 3b and Table 3), and one additional macrophyte AMS ^{14}C age (Table 2). All ^{14}C ages were calibrated to years before present (cal yr BP) using CALIB 5.0.1 [Stuiver and Reimer, 1993]. The two cores were correlated on the basis of their respective radionuclide chronologies, and a composite stratigraphy was created with the upper 13.5 cm being represented by the surface core. The 2.5 cm depth in the percussion core corresponded to the interpolated age at 13.5 cm depth in the surface core, implying the former failed to capture intact the upper 11 cm (or ~1000 years) of the record. The master chronology was developed by splicing ^{210}Pb and ^{14}C dates from the surface

core with the youngest ^{14}C date from the percussion core, and by applying linear fits between (1) the ^{14}C ages at 13.5 cm and 71.5 cm depths, and (2) the ^{14}C ages at 71.5 and 174.5 cm depths (Figure 3). All dates were subsequently adjusted to a zero age for 2003 AD, when the surface core was collected. All sediment ages mentioned subsequently are expressed in relation to calibrated years before 2003 AD (hereafter yr BP), according to the master chronology (Figure 3c). This chronology was further used to calculate fluxes of certain sediment parameters (diatom valves, spectrally inferred Chl *a*).

2.3. Diatoms

[10] Sediments from 19 intervals in the surface core (0.5 cm continuous increments from 0 to 5 cm depth, 1.0 cm increments from 5 cm to 14 cm) and 61 intervals in the percussion core (sampled either every 2 or 3 cm) were prepared for diatom analysis. For each of these 80 samples, freeze-dried splits weighing 200 mg were oxidized with 30% hydrogen peroxide to remove organic matter, followed by repeated rinsing with distilled water until neutral pH was achieved. Oxidized sediment slurries were spiked with a known quantity of *Eucalyptus* pollen in order to estimate sediment diatom concentrations [Wolfe, 1997]. Diluted aliquots from the slurries were dried onto coverslips at

Table 2. Radiocarbon Dates From Lake CF3 Cores

Midpoint Depth, ^a cm	Composite Depth, cm	$\delta^{13}\text{C}$ (‰ PDB)	Accession Number	Radiocarbon Age (^{14}C yr BP)	Calibrated Age (cal. yr BP $\pm\sigma$)
S-14.5	n/a	-22.7	CURL-7117	1310 $\pm$ 25	1240 $\pm$ 50
P-2.5	13.5	-20.9	CURL-6956	1240 $\pm$ 30	1180 $\pm$ 80
P-27.5	38.5	-23.6	CURL-7049	3460 $\pm$ 25	3740 $\pm$ 90
P-60.5	71.5	-25.0	CURL-7048	6450 $\pm$ 35	7370 $\pm$ 48
P-98.5	109.5	-25.0	CURL-7047	8150 $\pm$ 40	9070 $\pm$ 50
P-121.5	132.5	-27.5	CURL-6955	8520 $\pm$ 45	9510 $\pm$ 21
P-163.5	174.5	-26.0	CURL-6775	9770 $\pm$ 40	11,210 $\pm$ 21

^aS, surface core; P, percussion core.

Figure 3. (a) Chronology for the surface core established using a CRS age model applied to the excess ^{210}Pb inventory shown in Figure 3b, and anchored to one bryophyte AMS ^{14}C age at 14.5 cm. (c) Master chronology of the composite stratigraphy created by splicing the surface and percussion cores on the basis of their respective radionuclide chronologies.

room temperature and mounted to slides using Naphrax[®] medium. A minimum of 300 valves was identified and enumerated at each interval and converted to relative abundances, as well as total diatom concentrations estimated from counting introduced *Eucalyptus* markers. Taxonomy and nomenclature follow numerous sources cited in our ongoing study of the Baffin Island diatom flora [Joynt and Wolfe, 2001; Briner et al., 2006].

[11] Diatom-inferred pH (DIPH) reconstructions were derived from the transfer function of Joynt and Wolfe [2001] developed from 60 Baffin Island lakes with similar catchment geology and limnological characteristics to Lake CF3. This pH inference model, based on weighted-averaging calibration and regression, produces a highly significant relationship between measured and diatom-inferred pH values ($r^2_{\text{boot}} = 0.44$, $p < 0.01$, $\text{RMSE}_{\text{boot}} = 0.34$), which furthermore lacks any trend within residuals ($r^2 = 0.00$).

[12] Ordination by detrended correspondence analysis (DCA) was undertaken on down-core diatom relative frequencies, and regressed against DIPH with the objective of assessing the importance of pH in structuring diatom communities. DCA is an indirect ordination technique that models species as unimodal responses along synthetic variables (axes) that possess no *a priori* ecological information, in this way revealing the main directions of variation in fossil assemblages. Thereafter, detrended canonical correspondence analysis (DCCA), the constrained form of DCA, was used to obtain quantitative estimates of species turnover, or β -diversity, following Smol et al. [2005]. For this calculation, given in standard deviation (SD) units, sample ages assigned from the master chronology (Figure 3c) comprise the sole environmental variable to which diatom assemblages are constrained. The first axis of DCCA obtained this way enables direct comparisons of diatom

compositional changes during the Anthropocene (the last ~ 150 cal yr BP) to those expressed over the Holocene. All ordinations were undertaken with CANOCO version 4.5a [ter Braak and Smilauer, 2002].

2.4. Spectrally Inferred Sediment Chl *a*

[13] Sedimentary Chl *a* reconstructions were inferred using a newly developed application of visible reflectance spectroscopy which relates sediment spectral properties to wet-chemical measurements of pigment concentrations [Michelutti et al., 2005; Wolfe et al., 2006]. Briefly, sediment reflectance spectra between 350 and 2500 nm were obtained on freeze-dried and sieved core samples ($<125 \mu\text{m}$) using a FieldSpec[®] Pro spectroradiometer. Each sample was the average of 10 scans and expressed as % reflectance relative to a spectralon reference panel. The algorithm developed for Baffin Island lakes by Michelutti et al. [2005] was used to infer total Chl *a* from sediment spectral data, which includes primary Chl *a*, Chl *a* isomers, and the associated degradation products pheophytin *a* and pheophorbide *a*. This

Table 3. Lead 210 Dating From the Lake CF3 Gravity Core^a

Core Depth, cm	Unsupported ^{210}Pb Activity, Bq g^{-1}	Sediment Accum. Rate, $\text{g m}^{-2} \text{yr}^{-1}$	CRS Age, year AD
0.0–0.5	0.850	161	2003
0.5–1.0	0.704	157	1996
1.0–1.5	0.474	160	1989
1.5–2.0	0.853	152	1983
2.0–2.5	0.605	135	1968
2.5–3.0	0.535	138	1951
3.0–3.5	0.159	184	1920
3.5–4.0	0.125	108	1898
4.0–4.5	0.038	69	1851

^aNo unsupported ^{210}Pb is present beneath 5 cm. CRS, constant rate of supply.

CF3, Baffin Island, Nunavut: dominant diatom taxa
(70°31.95'N, 68°22.05'W)

Figure 4. The 10,900 year diatom record from Lake CF3, showing relative frequencies of the 17 most abundant taxa ordered by increasing DCA axis 1 species scores.

spectral technique exploits the depth of the red absorption band preserved in lake sediments (650–700 nm), which is sensitive to changes in total Chl *a* concentrations on the order of 0.01 mg g⁻¹ (dry sediment mass). The effects of variable sedimentation rates were adjusted for by calculating fluxes (units m⁻² yr⁻¹) for both diatom and chlorophyll concentrations.

3. Results

3.1. Sediment Lithology and Chronology

[14] The lithology of the composite sediment stratigraphy (surface and percussion cores) from Lake CF3 comprises 174 cm of olive-brown gyttja with scattered mosses, underlain by a 5 cm layer of moss-rich organic sediment dated to 11,210 cal yr BP. Beneath this, 15 cm of inorganic fine sand were recovered. Our paleolimnological record begins at the base of the 174 cm gyttja sequence (ca. 10,900 yr BP), as this corresponds to the earliest record of postglacial diatoms and the start of appreciable concentrations of autochthonous organic matter [Briner *et al.*, 2006]. Sediment accumulation from the base of this gyttja to 70 cm (ca. 7300 yr BP) was relatively rapid (0.31 mm yr⁻¹), but decreased more than three-fold thereafter (0.093 mm yr⁻¹) (Figure 3c). The excess ²¹⁰Pb inventory indicates the last ~150 years of sediment accumulation was confined to the upper 4.25 cm (Table 3). The plot of ²¹⁰Pb activity versus sediment core depth shows a down-core exponential decay (Figure 3a) characteristic of undisturbed surface sediments.

3.2. Diatom Stratigraphy

[15] The state of diatom preservation was excellent throughout the sediment core as there was no evidence of dissolution in even the most lightly silicified taxa. The diatom record from Lake CF3 shows four distinct assemblage zones, assessed visually from the relative frequency data (Figure 4). While the first and last diatom zone

boundaries are relatively well-defined biostratigraphically (zones 1–2 and 3–4), the transition between zones 2 to 3 is much more gradual (Figure 4). In zone 1, beginning at ca. 10,900 yr BP, there is a short-lived assemblage composed primarily of acidophilous *Aulacoseira* taxa, including *A. distans* and *A. perglabra*, small acidophilous *Eunotia* (cf. *exigua*), and low frequencies of the alkaliphilous fragilarioid genera *Staurosira* and *Pseudostaurosira*. Although represented by only one sample, this assemblage is crucial because it indicates that the original state of the lake following deglaciation was not highly alkaline. Although slides from samples immediately below this level contained too few diatoms to count, they also contain valves predominantly of acidophilous *Aulacoseira* and *Eunotia* taxa. It is noteworthy that a very similar assemblage has been described in the earliest organic sediments from lakes elsewhere on Baffin Island [Wolfe, 1996].

[16] In zone 2, this pioneer assemblage is rapidly replaced by colonial alkaliphilous taxa including *Staurosirella pinnata*, *Staurosira construens* var. *venter*, and *Pseudostaurosira pseudoconstruens*. These diatoms dominate assemblages for several millennia, until approximately 7800 cal. yr BP in the case of the latter taxon, while that former two remain dominant until 5000 cal. yr BP (Figure 4). These small, benthic *Fragilaria sensu lato* taxa are common to Arctic lakes, where they are typically associated with high lake-water alkalinity and elevated summer water temperatures [Pienitz *et al.*, 1995, 1999; Weckström *et al.*, 1997; Schmidt *et al.*, 2004]. Additional taxa present during this period in lower frequencies include *Fragilariforma exiguiformis*, *Nitzschia fonticola*, and *Encyonema* species. Two planktonic taxa, *Cyclotella tripartita* and to a lesser extent *C. stelligera*, also appear during this zone, typically attaining combined relative abundances of around 10%. Autecological data on *C. tripartita* indicates it is common in slightly acidic water with low dissolved inorganic carbon (DIC) content [Joynt and Wolfe, 2001; Rühland *et al.*, 2003].

Figure 5. Relationships between (a) diatom-inferred and measured lakewater pH for the 60-lake modern training set; (b) DCA axis 1 sample scores from the 80 fossil diatom assemblages enumerated in the Lake CF3 sediment core and their corresponding DIpH estimates; and (c) Holocene DIpH and percent sediment inorganic content calculated from loss-on-ignition (LOI) measurements [Briner *et al.*, 2006]. Four of the 80 down-core diatom assemblages lacked LOI from corresponding sediment intervals.

[17] In zone 3, from approximately 7000 cal. yr BP onwards, a gradual transition occurs in diatom assemblages, so that by 4300 yr BP the dominance of small fragilarioid diatoms ceded to a more diverse, and predominantly acidophilous assemblage consisting of *Navicula seminulum*, *N. schmassmannii*, *Aulacoseira distans*, *Pinnularia biceps*, *Cavinula pseudoscutiformis*, and *Achnanthes levanderi*. In the fourth and final zone representing the uppermost sediments, there is a rebound of the small fragilarioid diatoms that characterized the early to mid-Holocene, in particular *Fragilariaforma exiguiformis*. Although zone 4 still contains mainly acidophilous taxa, the increase of small fragilarioid diatoms is clearly discernible (Figure 4).

3.3. Reconstructed Lakewater pH and Primary Production

[18] The Baffin Island training set indicates that pH can be inferred from regional diatom assemblages with a precision of ± 0.34 pH units. This bootstrapped error margin originates from weighted-averaging calibration estimates of modern DIpH in the 60-lake training set (Figure 5a). Just over 60% of the calibration set taxa occur in the sediment core assemblage, and this number rises to 70% when only the dominant taxa (at least 10% relative abundance) are considered. Indeed, the pH transfer function of Joynt and Wolfe [2001] is designed to perform at sites such as Lake CF3, which is located roughly midway in latitude amongst the training set lakes (Figure 1). When this transfer function is applied to the 80 down-core assemblages from Lake CF3, the lowest diatom-rich sample returns a slightly acidic DIpH value (6.6), after which DIpH jumps rapidly to values between 7.8–8.0 for the ensuing two millennia (Figure 6a). The bottom-most diatom interval is dominated by *Aulacoseira distans*, *A. peglabra*, and acidophilous *Eunotia* species, all of which are well represented in the training set. After peaking in the early Holocene, DIpH progressively declines and the lake becomes acidic at 4300 cal. yr BP. This gradual acidification persists until approximately 150 yr BP, after which a slight but steady increase is detected, nearly to the point of neutrality within the last decade (Figure 6a). Although the surface sediment DIpH estimate from Lake CF3 (6.9) is considerably higher than lakewater pH measured from beneath the ice in May 2005 (5.89, Table 1), we emphasize that DIpH addresses open-water summer conditions, when the training set lakes were sampled, and when primary production approaches its annual high. The pH of Baffin Island lakes clearly declines during cold conditions, whether these are expressed seasonally or over much longer time scales [Wolfe, 2002]. This phenomenon, which will be reconsidered below, underscores the discrepancy between spring under-ice pH measured at Lake CF3 and the contemporary sediment DIpH value. The extremely high correlation observed between down-core DIpH and DCA axis 1 sample scores ($r^2 = 0.99$, $p < 0.001$, Figure 5b) confirms that pH is the dominant ecological gradient that regulates Holocene diatom assemblage composition in Lake CF3.

[19] Sediment concentrations of spectrally inferred Chl *a* and diatom valves show remarkably synchronous changes throughout the 10,900-year record, given that they represent independently generated proxies of past aquatic primary production. Both measures, as well as their respective

Figure 6. Profiles of (a) DIpH; (b) spectrally inferred total sediment chlorophyll *a*; (c) sediment chlorophyll *a* flux; (d) diatom valve concentrations relative to dry mass; (e) diatom valve flux; and (f) diatom diversity expressed by Hill's N2 [ter Braak and Smilauer, 2002], for the composite core from Lake CF3 according to its master chronology.

fluxes, record rapid increases that lag closely the establishment of the lake's initial diatom communities (Figure 6). All of these variables remain high throughout the early Holocene. Rapid declines in Chl *a* and diatom valve concentrations occur subsequently, around 8000 and 7000 cal. yr BP, respectively. This interval also marks the deceleration of sediment accumulation rates (Figure 3), which indicates that sedimentation in this lake is intimately linked to primary production and autochthonous organic matter supply. Because flux calculations are equally sensitive to changes in concentrations and sedimentation rates, they depict this early to mid-Holocene transition as an abrupt event. In general, all proxies of lake primary production remain low throughout the middle and late Holocene, with values well-below those attained prior to 8000 cal. yr BP. However, each proxy begins to rise again in the uppermost sediments of the surface core (Figure 6). Once more, it is the fluxes that capture these recent changes most effectively. We note that these fluxes are based on dry mass accumulation rates, so that the effect of elevated water content in near-surface sediments has been eliminated.

4. Discussion

4.1. A Model of Climate-Driven Lake Ontogeny

[20] Our results challenge whether lake ontogeny in Arctic ecosystems is driven primarily by edaphic processes, as is generally concluded from glaciated boreal regions.

[21] Here we use Lake CF3 as a 'template' for Arctic lake ontogeny, primarily because of the array of proxy data available and tight dating control; however, we note that there are numerous examples of lakes on Baffin Island that show similar ontogenetic processes to those recorded in this study [e.g., Wolfe and Smith, 2004; Wolfe et al., 2000; Miller et al., 1999; Wolfe and Härtling, 1996; Wolfe, 1994].

[22] In glaciated boreal regions, the long-term evolution of lakewater pH has involved initial contributions of alkalizing base cations from easily weathered glacial deposits [Mackereth, 1966; Whitehead et al., 1989]. However, in the absence of significant till cover of last-glacial age, and in the presence of highly dilute lake waters, the regulation of

acid-base equilibria is more closely dictated by climate. In fact, climate has been demonstrated to exert a first-order control on pH in several poorly buffered lakes in alpine and Arctic regions [Psenner and Schmidt, 1992; Wolfe, 2002; Larsen et al., 2006; Michelutti et al., 2006]. In these studies, cool intervals have been linked to low lakewater pH, and vice-versa, primarily through regulation of in-lake DIC dynamics. Extended ice cover during cold intervals traps respired CO₂ within the water column and inhibits light penetration, which further reduces photosynthetic draw-down of limnetic CO₂. The resulting increase of water column H₂CO₃ lowers lakewater pH. Conversely, reduced ice cover during warm intervals allows respired CO₂ to more freely escape to the atmosphere, while promoting algal production and carbon sequestration, thereby increasing pH.

[23] Ontogenic trends in lake primary production have also been linked to vegetation successional dynamics, specifically changes in available nitrogen generated by enhanced soil fixation under *Alnus* (alder) thickets [Engstrom et al., 2000]. However, in sparsely vegetated Arctic tundra with few N₂-fixers, climate supercedes the influence of edaphic factors on aquatic primary production. This occurs largely through climatic modulation of lake ice cover, which exerts a profound influence on primary producers, because even slight warming will create longer open-water periods and hence growing seasons [e.g., Smol et al., 2005, and references therein].

4.2. Holocene History of Lake CF3

[24] The diatom assemblages from the earliest postglacial sediments in Lake CF3 reveal that the lake's initial environment was slightly acidic. This starting point must reflect limited inputs of base cations sourced in the catchment, due to the crystalline nature of the local bedrock and the scarcity of fresh tills due to non-erosive cold-based glaciological regimes. Indeed, if detrital sources of catchment alkalinity were important, the most alkaline period of the lake's history would immediately follow deglaciation. Instead, following a short-lived acidic transient, pH rose markedly, and remained high (i.e., >7) for thousands of years during the early to middle Holocene. Furthermore, sediment inorganic

Figure 7. (a–f) Stratigraphies of the same parameters illustrated in Figure 6, with the addition of summed percentages of alkaliphilous colonial fragilarioid diatoms (Figure 7a), restricted to the last 150 years, and expressed in relation to the CRS ^{210}Pb chronology. Arrows and bold numbers indicate maximum values attained over the entire 10,900-year record.

content varies inversely with DIpH (Figure 5c), which is contrary to the predicted relationship if mineral erosion were at all involved in alkalinity generation. Indeed, stoichiometry predicts that the biological sequestration of CO_2 (aq.) necessary to raise pH through in-lake processes is proportional to the amount of organic matter buried in sediments, especially in the case of small lakes lacking significant outflow for carbon export, such as Lake CF3. With this in mind, we examine the possibility that delayed soil weathering following deglaciation, and hence of clay mineral production, is the proximate cause for the dramatic pH changes in the earliest portion of the record (Figure 6a). This is because clays are highly relevant in terms of enhancing organic matter preservation in a variety of depositional environments [Hedges and Keil, 1995]. In Lake CF3 sediments, clays increase from 25% to 50% of the mineral fraction around 10,500 cal. yr BP [Briner et al., 2006], concomitantly with increases of DIpH and sediment photoactive organic matter (Figure 6). However, by 10,000 cal. yr BP clay content returns to values $\sim 40\%$, which remain remarkably stable throughout the Holocene. This suggests that clays exerted little influence on the preservation of C sequestered from the water column, and hence on acid-base equilibrium, during the progressive acidification and production declines that characterize the zone 2–3 transition.

[25] The interval of maximum regional Holocene summer warmth, as reflected by numerous independent paleoclimate proxies (Figure 2), occurred between 10,000 and 8000 cal. yr BP. At this time, elevated lake production accompanied DIpH > 7.5 , suggesting a tight coupling between climate, biology, and lakewater chemistry. The diatom assemblage changes expressed across the zone 2–3 transition (Figure 4), and attendant DIpH declines (Figure 6a), closely precede regional glacier advances associated with cooling during the Neoglacial [Miller et al., 2005]. The DIpH and primary production proxies all faithfully track this gradual cooling. Palynological evidence from sediment cores throughout Baffin Island reveal that the modern sedge/heath tundra

became fully established ~ 8000 cal. yr BP, with only minor variations thereafter [Miller et al., 2005; Kerwin et al., 2004]. Thus, it is unlikely that the gradual acidification in Lake CF3 during this time was caused by changes in terrestrial vegetation. Following this cool interval, inferred Chl *a* and diatom concentrations remain low and show relatively little variability throughout the period of neoglaciation, with the exception of a rise in Chl *a* around 2500 cal. yr BP (Figures 6b and 6c). Neoglaciation culminated in the Little Ice Age (XV–XIXth centuries AD), after which climate warmed.

[26] The occurrence of *C. tripartita*, and to a lesser extent *C. stelligera*, during the onset of cooler conditions around 8000 cal. yr BP (Figure 4) appears contradictory as planktonic taxa are frequently associated with warm conditions [e.g., Smol et al., 2005]. However, autecological data on *C. tripartita* from Baffin Island indicates a slightly acidic pH optimum of 6.84 [Joynt and Wolfe, 2001], and preference for low DIC (0.8 mg/L) concentrations [Rühland et al., 2003], consistent with cool conditions and associated pH declines during this period. The importance of pH as a driver of diatom species composition in Lake CF3 is shown by the highly significant correlation between DCA axis 1 scores and DI-pH (Figure 5b). Still, autecological data on Arctic diatom taxa is far from complete, and whether the *Cyclotella* increase is driven by pH changes or other factors such as decreased erosion and hence increased light penetration, cannot be definitively determined.

[27] Diatom diversity, as estimated by Hill's N2 index, increases during intervals we associate with cold climates, especially the transition from early Holocene warmth to the Neoglacial (Figure 6f). In fact, N2 values closely mirror those of DIpH throughout the Lake CF3 record. Very similar results have been obtained from Fog Lake on northern Cumberland Peninsula, 500 km to the southeast of Lake CF3 [Wolfe, 2003]. One hypothesis is that fragmentation of benthic habitats, where small pockets of primarily epipelagic diatoms form discrete patches of growth, occurs during cold intervals. This allows a greater number

Table 4. Summary Results of Detrended Canonical Correspondence Analysis (DCCA) of Anthropocene and Holocene Diatom Assemblages Constrained to Time of Deposition (From the Age Model) as the Sole Environmental Variable^a

Interval	DCCA Axis 1 Gradient Length (β -Diversity in SD Units)	λ_1	λ_1/λ_2	<i>P</i>
Anthropocene (0–150 cal yr Bp)	1.00	0.09	1.17	0.003
Holocene (0–10,900 cal yr Bp)	2.809	0.519	3.604	0.001

^aSignificance of the ordination results (*P*) is obtained by Monte Carlo permutation tests (499 random permutations).

of taxa to flourish, albeit at lower levels of success in terms of total production relative to warm and species-poor intervals dominated by a few highly competitive taxa. Spatial fragmentation of benthic habitats has been observed in *Eunotia* and *Pinnularia* species in lakes elsewhere on Baffin Island [Wolfe, 1996].

[28] The model of climate-driven lake ontogeny presented here appears wholly validated by trends of the most recent decades, in which all proxies return to their ‘warm’ states, with respect to pH and primary production. The rapidity of recent changes, coupled to the absence of corresponding changes in sediment lithology, strongly imply they are not regulated by catchment processes. The ²¹⁰Pb chronology of these most recent changes and their relatively high stratigraphic resolution (Figure 7) indicate considerable synchrony with the history of recent Arctic warming (Figure 2f). Furthermore, the trend towards more productive limnological states in Baffin Island lakes is apparently widespread. A survey of 6 additional lakes near Clyde River on Baffin Island all reveal sharp recent increases in spectrally inferred Chl *a* [Michelutti *et al.*, 2005]. With current climate warming and prolongation of the ice-free growing season, continued alkalization of lakes is predicted as primary production increases apace. For example, the Lake CF3 results are directly compatible with detailed records from Finnish Lapland [Weckström *et al.*, 2006], the alpine zone of central Norway [Larsen *et al.*, 2006], and Boothia Peninsula in central Nunavut [LeBlanc *et al.*, 2004], where recent increases of alkaliphilous fragilioid taxa are reminiscent of early Holocene assemblages. But the likelihood seems equally great that anthropogenic atmospheric deposition may interfere, via either positive or negative feedbacks, with the above processes. Anthropogenic fluxes of strong acid anions to eastern Baffin Island are non-negligible, exceeding 50 kg km⁻² yr⁻¹ for both nitrate and sulfate [Goto-Azuma and Koerner, 2001]. While inorganic nitrogen deposition is almost certain to induce some level fertilization both on land and in lakes (which may or may not be transient), sulfate deposition necessarily implies acidification potential. A remaining question is the fate of pollutants already archived in regional glaciers that are undergoing rapid melt [e.g., Blais *et al.*, 2001].

4.3. Anthropocene and Arctic Lake Ontogeny

[29] The results of DCCA ordination applied to the Lake CF3 diatom assemblages constrained to time help assess the extent of anthropogenic changes in the context of the Holocene. Because time is the only predictor variable used in these ordinations, the fit of axis 1 with respect to diatom assemblages is highly significant ($p < 0.005$). The analysis for the last 150 years produced an axis 1 gradient length, or β -diversity, of 1.002 SD units (Table 4), which is directly comparable with many sites from the circumpolar Arctic,

and slightly higher than the benchmark for undisturbed boreal lakes (0.98 SD units [Smol *et al.*, 2005]). However, the gradient length for the complete 10,900-year diatom record is 2.809 SD units, which implies that recent rates of diatom compositional turnover are about a third of that accomplished since deglaciation. We believe this finding is probably typical for a large fraction of Arctic lakes, likely the majority, based on our recent multiple-lake studies [Michelutti *et al.*, 2005; Smol *et al.*, 2005]. However, the results from Lake CF3 are the first to place these recent changes explicitly in the context of the entire postglacial record of lake ontogeny. Indeed, we have developed the most highly resolved paleolimnological record to date for the Arctic, exceeded by only select northern hemisphere sites [Renberg, 1990].

[30] Two of six paleolimnological variables that demonstrably track lake development are currently at levels beyond any encountered in the last 10,900 years (Figure 7). Thus, there remains little doubt that marked deviations from natural trajectories of lake ontogeny have already occurred in the Arctic during the Anthropocene. We view the feedbacks operating between climate, primary production, and water chemistry as the central drivers of lake ontogeny on Baffin Island, largely outweighing the importance of catchment processes in terms of long-term biogeochemical regulation. Although the key limnological processes involved have clearly been sensitive to Holocene climate variability, and although we now understand the directions of biological and chemical shifts in relation to climate change, we remain incapable of predicting how the future will unfurl for lakes such as Lake CF3. This is because the current pace of climate change in the Arctic, amid uncertain futures with respect to anthropogenic atmospheric emissions, will conspire to produce an environment with no historical precedent.

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J. P. Briner, Department of Geology, State University of New York, Buffalo, NY 14260, USA.

N. Michelutti, Department of Biology, Queen's University, Kingston, ON, Canada M5S 3B1. (michelut@biology.queensu.ca)

G. H. Miller, Department of Geological Sciences and Institute of Arctic and Alpine Research, University of Colorado, Boulder, CO 80309-0450, USA.

A. P. Wolfe, Department of Earth and Atmospheric Sciences, University of Alberta, Edmonton, AB, Canada T6G 2E3.