

1  
2  
3  
4 **Phototrophic microbial community responses to late-Holocene environmental**  
5 **forcing of Southwest Greenland lakes**  
6  
7  
8  
9

10  
11  
12  
13  
14 NINA S. REUSS<sup>1\*</sup>, N. JOHN ANDERSON<sup>2</sup>, SHERILYN C. FRITZ<sup>3</sup>, GAVIN L. SIMPSON<sup>4</sup>  
15

16 <sup>1</sup>Freshwater Biological Laboratory, Department of Biology, University of Copenhagen,  
17 Helsingørsgade 51, DK-3400 Hillerød, Denmark  
18

19  
20 <sup>2</sup>Department of Geography, Loughborough University, Loughborough, LE11 3TU, U.K.  
21

22 <sup>3</sup>Department of Geosciences and School of Biological Sciences, University of Nebraska, 302  
23 Bessey Hall, Lincoln, NE 68588-0340, USA  
24

25 <sup>4</sup>Department of Geography, University College London, Pearson Building, Gower Street,  
26 London, WC1E 6BT, U.K.  
27  
28  
29  
30  
31  
32  
33  
34  
35

36 Keywords: Palaeolimnology, arctic, pigments, phototrophic bacteria, biogenic silica, carbon,  
37 energy and mass transfer, *Em* flux  
38  
39  
40  
41  
42

43 \* Corresponding author: Nina S. Reuss, e-mail: nreuss@bio.ku.dk  
44  
45  
46

47 Running title: Phototrophic community responses to environmental forcing in SW Greenland  
48  
49  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60

## Summary

1. The biological structure of arctic lakes is changing rapidly, apparently in response to global change processes, such as increasing air temperatures, although altered nutrient stoichiometry may also be an important driver. Equally important, however, are local factors (e.g. landscape setting, hydrological linkages and trophic interactions) that may mediate responses of individual lakes at the regional scale. Despite general acknowledgement of the importance of local factors, there has been little focus on among lake variability in the response to environmental change.
2. Sedimentary pigments, organic carbon and nitrogen and biogenic silica (BSi) in  $^{210}\text{Pb}$  and  $^{14}\text{C}$ -dated sediment cores from three contrasting lakes in the Kangerlussuaq area ( $\sim 67^\circ \text{N}$ ,  $51^\circ \text{W}$ ) of Southwest Greenland were used to reconstruct algal and phototrophic bacterial ecological change during the late Holocene. Water chemistry for the individual lakes varies in terms of conductivity (range:  $30\text{--}3000 \mu\text{S cm}^{-1}$ ) and stratification regimes (cold monomictic, dimictic and meromictic), linked to their position along the regional climate gradient from the coast and to the present ice sheet margin.
3. Despite essentially similar regional climate forcing over the last  $\sim 1000$  years, marked differences among lake types were observed in the phototrophic communities and their temporal variability. Considerable short-term variability occurred in an oligosaline, meromictic lake (SS1371), dominated by purple sulphur bacteria, likely due to a tight coupling between the position of the chemocline and the phototrophic community. Communities in a lake (SS86) located on a nunatak, just beyond the edge of the present ice sheet shifted in a non-linear pattern, approximately 1000 cal. yrs BP, possibly due to lake-level lowering and loss of outflow during the Medieval Climate Anomaly. This regime shift was marked by a substantial expansion of green sulphur bacteria.
4. A dilute, freshwater coastal lake (SS49) dominated by benthic algae was relatively stable until ca. 1900 AD when rates of community change began to increase. These changes in

1  
2  
3 benthic algal pigments are correlated with substantial declines (1.3 to 0.44‰) in  $\delta^{15}\text{N}$  that  
4  
5 are indicative of increased deposition of atmospheric inputs of industrially-derived  $\text{NO}_x$   
6  
7 into the atmosphere.  
8

- 9  
10 5. Climate control on lake ecosystem functioning has been assumed to be particularly  
11  
12 important in the Arctic. This study, however, illustrates a complex spatial response to  
13  
14 climate forcing at the regional scale and emphasizes differences in the relative  
15  
16 importance of changes in the mass ( $m$ , both precipitation and nutrients) and energy flux  
17  
18 (E) to lakes for the phototrophic community structure of low-Arctic Greenland lakes.  
19  
20  
21  
22  
23  
24  
25  
26  
27  
28  
29  
30  
31  
32  
33  
34  
35  
36  
37  
38  
39  
40  
41  
42  
43  
44  
45  
46  
47  
48  
49  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60

## Introduction

Arctic ecosystems are changing rapidly in response to global change processes (Overpeck *et al.*, 1997; Post *et al.*, 2009). Much of the evidence for altered biological structure in arctic lakes over the last 50–150 years are derived from the natural archives recorded in lake sediment (Smol *et al.*, 2005). Considerable emphasis has been placed on increased air temperature at high latitude and its impact on lake thermal budgets as a driver of these observed changes (Smol *et al.*, 2005) but other global change processes, such as increased regional N deposition are undoubtedly important (Bergstrom, Blomqvist & Jansson, 2005; Wolfe, Cooke & Hobbs, 2006). While these large scale, hemispheric, processes impact lake functioning, indirect effects, largely mediated through catchment processes such as soil development, vegetation succession, altered nutrient cycling and hydrology can have profound impacts on lakewater chemistry and biological structure (Kane *et al.*, 1992; Battarbee, 2000; Engstrom *et al.*, 2000). The effect of these direct and indirect fluxes of mass and energy on lake response to climate has been highlighted in a conceptual model developed by Leavitt *et al.* (2009). This model indicates the extent to which direct climate impacts on lakes have to be balanced by consideration of catchment-mediated fluxes of mass ( $m$  - water, solutes, and particles) and energy ( $E$  - e.g. irradiance [PAR, UVR], wind friction, or transfer of atmospheric heat). This balance may be very important in the Arctic where precipitation is often very low, nutrients are limiting, and there is a strong marked seasonality of energy inputs (Vincent, Hobbie & Laybourn-Parry, 2008).

The area around Kangerlussuaq, Southwest (SW) Greenland (Fig. 1) contains ~20 000 lakes and has a strong regional climate gradient, from a maritime coastal zone to the arid interior, which is in part caused by the strong orographic effect of the Sukkertoppen ice-cap (Fig. 1). Marked differences in precipitation ( $m$  flux) and effective solar radiation inputs ( $E$  flux) exist between the coast (where summer fog banks are common) and the inland areas. Groundwater flows are minimal in SW Greenland due to permafrost, and so hydrological ( $m$ ) fluxes to lakes reflect the influence of catchment-lake ratios, landscape position and relief. The regional

1  
2  
3 environmental (climate) gradient influences both contemporary water chemistry (Anderson *et*  
4 *al.*, 2001) and biological structure (Brodersen & Anderson, 2000; Pla & Anderson, 2005; Perren  
5 *et al.*, 2009).

6  
7  
8  
9  
10 Recent palaeolimnological work in the Kangerlussuaq area has focussed on both fresh  
11 and oligosaline lakes at the head of the fjord (Fig. 1) where changing effective moisture (i.e. the  
12 balance between precipitation and evaporation) has resulted in a number of lakes with  
13 evaporatively concentrated ion chemistry (Anderson *et al.*, 2001). McGowan *et al.* (2008) and  
14 Anderson *et al.* (2008) concluded that lake ontogeny was a significant factor controlling algal  
15 communities and biological structure in SW Greenland lakes. The interaction of climate and  
16 catchment processes also was highlighted by a terrestrial-aquatic macrofossil study of a small,  
17 dilute lake (Heggen *et al.*, 2010). In-lake processes such as the nature and strength of  
18 stratification as some of the oligosaline lakes are meromictic (McGowan, Ryves & Anderson,  
19 2003), also can affect lake response to environmental/climate forcing.

20  
21  
22  
23  
24  
25  
26  
27  
28  
29  
30  
31  
32 Much of the evidence for change within lake communities in the Arctic has focussed on  
33 diatoms, but there is also some evidence for altered food web dynamics (Quinlan, Douglas &  
34 Smol, 2005) and increased lake productivity derived from reflectance spectroscopy (Michelutti  
35 *et al.*, 2005). While diatoms are an important component of arctic lake ecosystems, non-siliceous  
36 algae also can be significant (Vadeboncoeur *et al.*, 2003; Bonilla, Villeneuve & Vincent, 2005).  
37 Pigments are produced by algae and other phototrophic organisms, some of which are  
38 taxonomically specific while others are ubiquitous in most algal groups (Jeffrey, Mantoura &  
39 Wright, 1997). Therefore, pigment analysis by High Performance Liquid Chromatography  
40 (HPLC) provides important information about a wide range of primary producers and  
41 phototrophic bacteria and hence whole-lake responses to environmental forcing (Leavitt &  
42 Hodgson, 2001). Analysis of sedimentary pigments is now an important component of  
43 palaeolimnological studies (Vinebrooke *et al.*, 1998; Leavitt *et al.*, 1999; Leavitt and Hodgson  
44 2001) but their application to arctic lakes has been relatively limited (Pienitz *et al.*, 2000;  
45  
46  
47  
48  
49  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60

1  
2  
3 Anderson *et al.*, 2008; Reuss *et al.*, 2010a; Reuss *et al.*, 2010b). In a pigment study of two  
4 oligosaline lakes in SW Greenland, McGowan *et al.* (2008) highlighted the individual and non-  
5 linear response of lakes to climate forcing at centennial timescales, perhaps due to differences in  
6 lake morphometry and benthic-pelagic productivity. Here we use pigment analysis to address  
7 recent ecological change in lake phototrophic communities (i.e. algae and phototrophic bacteria).  
8  
9

10  
11  
12  
13  
14 Focusing on three lakes positioned along the regional climate gradient in the  
15 Kangerlussuaq area (Fig. 1) and with very different hydrological budgets, conductivity and  
16 stratification regimes, we evaluate the degree of spatial variability in the extent to which algal  
17 and phototrophic bacterial responses to late-Holocene climate change in this area are mediated  
18 by changing fluxes in mass ( $m$ ) and energy (E) (sensu Leavitt *et al.*, 2009). Closed basin lakes  
19 with increased ion-concentration and strong stratification is expected to respond strongly to  
20 direct effects of mass and energy flux determining changes in effective moisture (Battarbee,  
21 2000; Leavitt *et al.*, 2009). Previous work on this lake type in the Kangerlussuaq area suggests  
22 that the over-riding effect on lake functioning is via direct mass transfer: i.e. the impact of  
23 hydrological fluxes on lake levels (McGowan *et al.*, 2003; Aebly & Fritz, 2009; Leavitt *et al.*,  
24 2009). In contrast, freshwater, dilute lakes in this area are more affected by indirect changes in  
25 mass flux mediated by the specific catchment properties (Anderson *et al.*, 2008; Heggen *et al.*,  
26 2010).  
27  
28  
29  
30  
31  
32  
33  
34  
35  
36  
37  
38  
39  
40  
41  
42  
43  
44

## 45 **Methods**

### 46 *Study sites*

47  
48 Kangerlussuaq (Søndre Strømfjord in Danish) is a 150-km long glacial fjord in SW Greenland  
49 (Fig. 1), and the area between the coast and the ice sheet is the widest ice-free land area in  
50 Greenland today (~180 km). There are thousands of lakes that are well suited for palaeoclimatic  
51 investigations and direct anthropogenic influence is minimal (Anderson *et al.*, 2001); however,  
52 as with the majority of arctic lakes, they are impacted by northern hemisphere atmospheric  
53  
54  
55  
56  
57  
58  
59  
60

1  
2  
3 pollution (Bindler *et al.*, 2001). The area encompasses a strong climatic gradient from the inland  
4  
5 continental climate to a more maritime environment at the coast. Annual precipitation at  
6  
7 Kangerlussuaq at the head of the fjord is  $<150 \text{ mm yr}^{-1}$ , while precipitation at the coast is  $>500$   
8  
9  $\text{mm yr}^{-1}$ . The annual temperature range is greatest at Kangerlussuaq town with average  
10  
11 summer/winter temperatures of  $10^{\circ}/-18^{\circ}\text{C}$  compared to  $6^{\circ}/-12^{\circ}\text{C}$  at the coast. Estimated  
12  
13 evaporation at the head of the fjord is  $\sim 300 \text{ mm}$  and together with the low precipitation results in  
14  
15 negative effective precipitation in the area extending to approximately half way (80 km) to the  
16  
17 coast from the ice sheet margin (Hasholt & Sogaard, 1978).  
18  
19

20  
21 An extensive study of 86 lakes from the Kangerlussuaq area along a transect from the ice  
22  
23 sheet to the coast identified the main characteristics of lake chemistry (Anderson *et al.*, 2001).  
24  
25 The majority of the lakes are dilute ( $<300 \mu\text{S cm}^{-1}$ ), with an mean conductivity of  $160 \mu\text{S cm}^{-1}$   
26  
27 while the remaining (ca. 10% ) of the lakes are oligosaline, have conductivity  $>800 \mu\text{S cm}^{-1}$  and  
28  
29 are characterised by Na Mg- $\text{CO}_3$  chemistry (Anderson *et al.*, 2001). These oligosaline lakes  
30  
31 (Williams, 1991) are situated well above the marine limit and cannot therefore have been formed  
32  
33 by trapping of marine water due to isostatic uplift, which is a common method of oligosaline  
34  
35 lake formation in the Arctic and Antarctic (Burton, 1981; Ouellet *et al.*, 1987). The primary  
36  
37 cause of elevated salinity in these saline closed-system lakes is most likely evaporation enhanced  
38  
39 by inputs of salts from nearby terrestrial sources, brought to the lakes by aeolian activity  
40  
41 (Anderson *et al.*, 2001; Willemsse *et al.*, 2004).  
42  
43  
44

45  
46 Details of the physical and chemical characteristics of the three study lakes are given in  
47  
48 Table 1. The study lakes are representative of the range of water chemistry (fresh-oligosaline),  
49  
50 DOC concentration and thermal stratification (cold monomictic, dimictic and meromictic) found  
51  
52 in the Kangerlussuaq area (see Table 1) (Anderson *et al.*, 2001). The lakes are also spatially  
53  
54 separated along the geographic gradient from the coast to the inland ice (Fig. 1). Lake SS49 is  
55  
56 located  $\sim 120 \text{ km}$  to the west of the ice sheet at 340 m a.s.l. and is typical of the dilute  
57  
58 oligotrophic lakes ( $< 50 \mu\text{S cm}^{-1}$ ; DOC  $12 \text{ mg L}^{-1}$ ) found along the coastal margin (Anderson *et*  
59  
60

1  
2  
3 *al.*, 2001; Anderson & Stedmon, 2007). SS1371 located ~45 km west of the ice sheet is an  
4  
5 oligosaline closed-basin lake, surrounded by extensive fossil shorelines indicative of  
6  
7 substantially higher lake levels in the past. SS86 is located on a nunatak, an isolated mountain  
8  
9 top that protrudes over the surface of the ice sheet, approximately 5 km inside the ice sheet and  
10  
11 has significantly higher conductivity and alkalinity compared to both a neighbouring lake (SS32)  
12  
13 (Table 1) and to other freshwater lakes in the Kangerlussuaq area (Anderson *et al.*, 2001). SS86  
14  
15 and SS1371 are fishless but sticklebacks (*Gateus* sp.) were observed in SS49.  
16  
17

### 20 21 *Sediment coring*

22  
23 Sediment cores (22–34 cm in length) were collected in April 2000 by freeze-coring which results  
24  
25 in cores with an undisturbed surface layer. The sediments from SS1371 and SS86 were  
26  
27 laminated, whereas the core from SS49 had no visible laminations. The cores were shipped to the  
28  
29 laboratory frozen and wrapped in black plastic to limit light and oxygen access. Cores were  
30  
31 stored undisturbed at –20°C in the dark until sectioning. Sectioning was conducted in a cold  
32  
33 room at less than –10°C by removing the outer layers of the core with a wood planer and cutting  
34  
35 into 0.5 cm sections using a band saw. The samples were then cleaned with a glass plate and  
36  
37 stored at –80°C until freeze-drying and extraction of pigments.  
38  
39  
40  
41  
42

### 43 44 *Sediment chronology*

45  
46 Cores were dated by means of  $^{210}\text{Pb}$  (Fig. 2), determined using gamma assay (Appleby *et al.*,  
47  
48 1986) and bulk AMS  $^{14}\text{C}$  were determined for selected depths on Russian cores taken at the same  
49  
50 time as the freeze cores and within 2 m of the freeze core location. The Russian and freeze cores  
51  
52 were readily cross-correlated using distinctive carbon profiles (total carbon or organic carbon  
53  
54 wt%, see Fig. 3).  
55  
56  
57  
58

### 59 60 *Pigment analyses*



1  
2  
3 0.1–0.5 g of homogenized freeze-dried sediment was extracted in 5 ml cold 100% acetone and  
4  
5 spiked with an appropriate amount of internal standard  $\beta$ -apo-8-carotenal (25–100 $\mu$ l). The  
6  
7 mixture was sonicated, extracted overnight at  $-20^{\circ}\text{C}$ , filtered and diluted to a final concentration  
8  
9 of 80% acetone to increase peak resolution before run by High Performance Liquid  
10  
11 Chromatography (HPLC) (Reuss & Conley, 2005). Quantitative analyses of all pigments were  
12  
13 conducted on a Shimadzu HPLC equipped with an on-line photodiode array detector (SPD-  
14  
15 M10Avp) for quantification and fluorescence detector (RF-10Axl) for identification purposes  
16  
17 only. The run method was a modification of Wright *et al.* (1991) as described by Reuss &  
18  
19 Conley (2005).  
20  
21  
22

23 Tentative identification of individual pigments was based on a combination of retention  
24  
25 time and absorption spectra. The quality of the absorption spectra diminished down core for  
26  
27 some pigments, and identification were then primarily based on the retention time. Reference  
28  
29 absorbance spectra for identification and quantification of algal pigments were obtained from a  
30  
31 single run of standards from DHI Water and Environment, Denmark and (Jeffrey *et al.*, 1997).  
32  
33 Absorbance spectra of bacterial pigments were obtained from a standard of okenone provided by  
34  
35 A. Lami (CNR.ISE, Italy) and a culture of a green sulphur bacterium *Chlorobium*  
36  
37 *phaeobacteroides* containing bacteriochlorophyll-e provided by R. Cox (University of Southern  
38  
39 Denmark, Denmark). Concentrations of bchlorophyll-e and okenone standards were determined  
40  
41 at absorbance maxima using specific molar extinction coefficients (467 nm,  $10 \times 10^4 \text{ l mmol}^{-1}$   
42  
43  $\text{cm}^{-1}$  (Frigaard, Larsen & Cox, 1996), and 487 nm,  $13.4 \times 10^4 \text{ l mmol}^{-1} \text{ cm}^{-1}$  (Züllig, 1985),  
44  
45  
46 respectively). Quantification of chlorophyll-a (chl-a) and pheopigment-a's (ppn-a) was  
47  
48 conducted at 666 nm while carotenoids, chlorophyll-b, pheopigment-b's and  
49  
50 bacteriochlorophylls were quantified at 449 nm. Normalisation of concentrations of all samples  
51  
52 were conducted by scaling to the highest value of the internal standard,  $\beta$ -apo-8-carotenal, except  
53  
54 for the top half of the core from SS86 where the bacteriochlorophylls interfered with the internal  
55  
56 standard peak. The ratio of chl-a/ppn-a was calculated to provide a simple preservation index as  
57  
58  
59  
60

1  
2  
3 chlorophyll-a is readily degraded while its degradation products are much more stable. Three  
4  
5 samples from lake SS49 were excluded from the dataset due to a very irregular injection peak  
6  
7 and chromatogram.  
8  
9

#### 10 11 *Carbon (C), nitrogen (N), biogenic silica (BSi) and stable isotope (<sup>15</sup>N) analyses*

12  
13 Carbon and nitrogen content were determined using a CHNS elemental analyzer (CE instruments  
14  
15 EA1110). The total fraction of C and N was measured on freeze-dried samples, while the  
16  
17 inorganic C-fraction was measured on combusted samples (2h at 500°C) and corrected for loss of  
18  
19 organic mass. Organic C was calculated as the difference between the total and inorganic  
20  
21 fraction. Biogenic silica (BSi) analyses were carried out on every or every other level in SS86,  
22  
23 SS49, and SS1371. BSi was measured using the DeMaster (1981)-method as modified by Conley  
24  
25 & Schelske (2001). Quality control of BSi-analyses was carried out by using reference samples  
26  
27 used in an international inter-laboratory comparison (Conley, 1998) and 10% duplicate samples.  
28  
29 Samples for <sup>15</sup>N/<sup>14</sup>N at SS49 were analysed at the UC Davis Stable Isotope Facility, California,  
30  
31 USA on Hydra 20-20 or Anca-GSL isotope ratio mass spectrometers. Freeze-dried sub-samples  
32  
33 of sediment were milled to a fine powder using a Retsch mixer mill. Approximately 0.001g of  
34  
35 milled sediment was transferred to pre-weighed tin capsules, which were then sealed. The  
36  
37 isotopic ratio of <sup>15</sup>N/<sup>14</sup>N is expressed using the delta (δ) notation in parts per thousand (or per  
38  
39 mille, ‰), where  $\delta^{15}\text{N} (\text{‰}) = [(R_{\text{sample}} / R_{\text{standard}}) - 1] \times 1000$ , where R is the <sup>15</sup>N/<sup>14</sup>N ratio in the  
40  
41 measured sample or the appropriate standard. The standard for nitrogen is the <sup>15</sup>N of  
42  
43 atmospheric nitrogen (commonly referred to as AIR).  
44  
45  
46  
47  
48  
49

#### 50 51 *Statistical analyses*

52  
53 In order to summarize changes occurring in the sediment pigment composition in the three lakes  
54  
55 over time, Principal Component Analysis (PCA) was undertaken using CANOCO for Windows  
56  
57 version 4.5. The analyses were based on log<sub>10</sub>(x+1) transformed pigment concentrations, which  
58  
59  
60

1  
2  
3 were centred and standardised prior to analysis. Bacterial chlorophyll-e homologs and their  
4 degradation products were grouped into total Bchl-e and Bppn-e before analysis of lake SS86.  
5  
6 Because of possible uncertainties in the sediment chronologies, only visual comparisons between  
7 the PCA-axis 1 scores and the independent proxies for global environmental change (ice-core  
8 temperature inferences, effective precipitation) were undertaken.  
9  
10  
11  
12

## 13 14 15 16 **Results**

### 17 *Sediment chronology*

18  
19 The cores used in this study cover a period of approximately 600–1600 years. Unsupported  $^{210}\text{Pb}$   
20 activities decline more or less exponentially with depth in SS86 and SS49 (Fig. 2). At SS86,  
21 total  $^{210}\text{Pb}$  activity reaches equilibrium with the supporting  $^{226}\text{Ra}$  at a depth of around 5 cm and  
22 at SS49 at 14 cm (Fig. 2).  $^{137}\text{Cs}$  activity had a relatively well resolved peak between 2.5–2.75  
23 cm at SS86 and at SS49 at 4.5–5.5 cm. The presence of a clear  $^{241}\text{Am}$  peak around 2.5–2.75 cm  
24 at SS86 (Fig. 2) provides further support for allocating a bomb-testing date (1963) to these  
25 levels. Application of the CRS dating model (Appleby & Oldfield, 1978; Appleby, 2001), to the  
26 unsupported  $^{210}\text{Pb}$  data, constrained by the  $^{137}\text{Cs}$  profiles, suggests a sediment accumulation of  
27  $\sim 0.11 \text{ cm y}^{-1}$  over the last  $\sim 100$  years at lake SS49. At SS86 the CRS model again indicates a  
28 relatively uniform sediment accumulation rate with 5.25 cm depth dating to  $\sim 1880$ . At SS86 the  
29 final chronology was determined using  $^{210}\text{Pb}$  activity and extrapolation of simple linear  
30 regression of the  $^{14}\text{C}$  chronology (Fig.3). For SS49 only the environmental changes that occur  
31 within the period of  $^{210}\text{Pb}$  activity are discussed in detail, so only the CRS chronology is referred  
32 to in the results and discussion.  
33  
34  
35  
36  
37  
38  
39  
40  
41  
42  
43  
44  
45  
46  
47  
48  
49  
50

51  $^{210}\text{Pb}$  activity at SS1371 was very low and irregular (P.G. Appleby, pers. comm.),  
52 possibly due to the extreme chemical environment of this permanently anoxic basin. As a result,  
53 the chronology for the freeze core is based on extrapolation of the  $^{14}\text{C}$  chronology using a simple  
54  
55  
56  
57  
58  
59  
60

1  
2  
3 linear regression fitted to the  $^{14}\text{C}$  dates and sediment surface ( $\sim 2000$  AD) (see Fig. 3 and  
4  
5 D'Andrea *et al.*, 2011 for details).  
6  
7

#### 9 10 *Carbon, nitrogen and biogenic silica (BSi)*

11 All three lakes showed changes in organic carbon (OC) content of about 8% and in nitrogen  
12 (TN) content of about 1% over time resulting in conspicuous changes in the molar OC/TN ratio  
13  
14 with a decline towards the present in SS49 and SS1371 while a very irregular profile was  
15  
16 observed in SS86 (Fig. 4). The low-conductivity coastal site (SS49) had a relatively high OC  
17  
18 content, 12–19 wt% with a decrease from the base of the core to  $\sim 13$  cm and a subsequent  
19  
20 increase to the top. An increase in TN content towards the top was reflected in a decrease in the  
21  
22 molar OC/TN ratio particularly in the top 10 cm. The oligosaline lake, SS1371, and the nunatak  
23  
24 lake, SS86, had lower organic carbon content, 4–12 and 3–11 wt%, respectively (Fig. 4). The TN  
25  
26 profile at SS86 increased from  $\sim 7$  cm to the top, while marked changes in the molar OC/TN ratio  
27  
28 is observed between 13 and 5 cm.  
29  
30  
31  
32  
33

34 Biogenic silica (BSi) content exhibited differences of an order of magnitude between the  
35  
36 three lakes (Fig.4). At SS49 the BSi content is high throughout the core (12–22 wt%) indicating  
37  
38 high diatom abundance but is low (1–2.4 wt%) at SS1371 possibly due to silica dissolution. At  
39  
40 SS86 BSi is more variable, with highest values (3–5 wt%) in the bottom part of the core.  
41  
42  
43  
44

#### 45 *Sediment pigment record*

46 Different pigments dominate in the three study lakes and their concentrations differ with an order  
47  
48 of magnitude between the relatively dilute lakes (SS49 and SS86) and the oligosaline lake  
49  
50 (SS1371) (Fig. 4). Although chlorophyll-a, chlorophyll-b and fucoxanthin are labile compounds,  
51  
52 the remainder of the pigments are relatively stable once incorporated into the sediment record.  
53  
54  
55 Pigments at the coastal lake SS49 (Fig. 4a) indicate dominance of chromophyte algae  
56  
57 (fucoxanthin, diatoxanthin, diadinoxanthin-like), as well as the presence of green algae and  
58  
59  
60

1  
2  
3 higher plants (lutein, pheophytin-b) and cyanobacteria (canthaxanthin, echinenone). The labile  
4 pigments, chlorophyll-a and fucoxanthin, increase towards the top of the core. All other  
5 pigments have relatively uniform profiles throughout the core. The preservation indicator,  
6 chlorophyll-a/pheophytin-a (chl-a/ppn-a) ratio, indicates that no major changes in preservation  
7 conditions have occurred throughout the period covered by the core, except for significantly  
8 increased preservation (or more likely incomplete degradation) of the labile chlorophyll-a in the  
9 top few cm. These changes were reflected in the ordination of the pigments that showed  
10 clustering of all samples except for the top samples that diverged from the rest.  
11  
12  
13  
14  
15  
16  
17  
18  
19

20  
21 The dominant pigment at the oligosaline lake SS1371 is okenone, which increases  
22 towards the core top (Fig. 4b) and is specific to phototrophic purple sulphur bacteria, which  
23 require photic zone anoxia to thrive (Pfennig 1989). Generally high but fluctuating concentration  
24 of all pigments and high values of the preservation indicator (chl-a/ppn-a) indicates that  
25 preservation at this site was good, in accordance with the inferred anoxic conditions due to  
26 presence of the anoxygenic phototrophic bacteria. The algal pigments indicate dominance of  
27 green algae and higher plants (lutein, chlorophyll-b and pheophytin-b) and chromophyte algae  
28 (diatoxanthin). Indicators of total algal biomass ( $\beta$ -carotene, chlorophyll-a, pheophytins-a) show  
29 no marked trend throughout the core. Profiles of green algae and higher plant pigments (lutein,  
30 chlorophyll-b, pheophytins-b) show peak concentration at 14–16 cm and a decreasing trend  
31 towards the surface. Ordination of the pigments showed no discernible trends in the samples.  
32  
33  
34  
35  
36  
37  
38  
39  
40  
41  
42  
43  
44

45 At SS86, located on the nunatak, marked changes in the pigment record occurred around  
46 16–18 cm, with a shift from algal pigments to near complete dominance by bacterial  
47 chlorophyll-e (bchl-e) homologs specific to green sulphur bacteria (Fig. 4c). Like the purple  
48 sulphur bacteria, green sulphur bacteria require photic zone anoxia to grow (Pfennig 1989). This  
49 profile follows that of OC% but with a small lag. Algal pigments exhibited relatively large  
50 variation down core with two peaks near the base of the core, a peak at 5–9 cm, and a surface  
51 peak. Chromophyte-algal indicators (fucoxanthin, diatoxanthin, diadinoxanthin-like) dominate  
52  
53  
54  
55  
56  
57  
58  
59  
60

1  
2  
3 the record, but indicators of green algae (lutein, pheophytin-b) are also an important component.  
4  
5 Several common algal and higher plant indicators (diatoxanthin, alloxanthin, canthaxanthin and  
6  
7 lutein) disappear or were masked by large amounts of bacterial pigments (bchl-e homologs) in  
8  
9 the top half of the core. The preservation indicator show periods of increased preservation  
10  
11 coinciding with periods of maximum algal pigment concentration, e.g. around 5–9 cm and at the  
12  
13 core surface. Ordination of the pigments showed two distinct clusters representing the top and  
14  
15 bottom half of the core.  
16  
17

### 20 21 *Stable $\delta^{15}\text{N}$ isotopes at SS49*

22  
23 Stable isotope measurements from SS49 (Fig. 5a) showed that prior to 1920, sediment  $\delta^{15}\text{N}$   
24  
25 measurements, though sparse, were stable around 1.3‰. Subsequently,  $\delta^{15}\text{N}$  values decline by  
26  
27 ~1‰ from ~1920, whilst organic C and total N content of sediment samples almost double (Fig.  
28  
29 4a). OC/TN molar ratio exhibits a distinct decrease over the same period. The pigment PCA-1  
30  
31 scores follow the observed isotopic trend. The timing of the decline in the lake sediment proxies  
32  
33 are well in line with changes in  $\text{NO}_3$  and  $\delta^{15}\text{N}$  from the Greenland ice cores (Hastings, Jarvis,  
34  
35 and Steig 2009, Fig. 5b) while no trend is observed in the temperature trends from Nuuk (Fig.  
36  
37 5c).  
38  
39  
40  
41  
42

## 43 **Discussion**

44  
45  
46 Changes in the phototrophic communities of three contrasting lakes in the Kangerlussuaq area,  
47  
48 SW Greenland revealed very different responses to environmental change over the last 600–1600  
49  
50 years which are interpreted in relation to Leavitt *et al.*'s (2009) E-m flux model (Fig. 6). At  
51  
52 SS1371 and SS86 the main driver is variability of E flux via effects on stratification and light  
53  
54 availability for photosynthetic bacteria. At SS86 m flux was also important historically, resulting  
55  
56 in lake level lowering, creation of a closed-basin system and the development of chemical  
57  
58 stratification. At SS49 m and E fluxes were more constant as medium-term (decadal to  
59  
60

1  
2  
3 centennial) climate variability reduced, although atmospheric nutrient loading may have  
4  
5 increased (Fig. 5). Our interpretations of the observed in-lake changes not only emphasize the  
6  
7 varying importance of the mechanisms involved in transferring and filtering environmental  
8  
9 forcing into ecological response by phototrophic communities but also that these relationships  
10  
11 can themselves change over time.  
12  
13

#### 14 15 16 *Mass flux: precipitation*

17  
18 The most pronounced impact of the reduced precipitation from around 800 cal. yr BP on the  
19  
20 study lakes was observed at the nunatak lake (SS86), where the pigment, carbon and BSi profiles  
21  
22 indicate a switch from a freshwater, autotrophic lake to an increasingly chemolithotrophic  
23  
24 system within a few decades around 1000 cal. yr BP (Fig. 4). This is concomitant with the latter  
25  
26 part of the Medieval Climate Anomaly (Fig. 6) and a lake-level lowering, ultimately falling  
27  
28 below the outlet level. This interpretation is consistent with studies of palaeoshorelines in the  
29  
30 area indicating that precipitation (and lake levels) decreased briefly prior to 1000 cal. yr BP and  
31  
32 then fell sharply to modern levels after ~700 cal. yr BP (Aebly & Fritz, 2009). SS86 has a very  
33  
34 small catchment, too small to supply the lake with sufficient runoff to sustain an outflow during  
35  
36 a period of negative precipitation balance. The role of lake:catchment ratios is exemplified by the  
37  
38 neighbouring lake, SS32, an open basin with a larger catchment and an outflow resulting in  
39  
40 different water chemistry characteristics (Anderson *et al.*, 2001; Table 1), that have not  
41  
42 undergone the dramatic switches seen at SS86 (Perren *et al.*, 2009).  
43  
44  
45  
46

47  
48 Closed-basin lakes are thought to be especially sensitive to climate change due to the  
49  
50 tight coupling between water level and chemistry, forced by changes in temperature and  
51  
52 effective precipitation (Fritz, 1996; Battarbee, 2000). However, the time period covered by the  
53  
54 freeze core at SS1371 was too short to include the effect of the lake level lowering inferred for  
55  
56 the area from ~1000 cal. yr BP. However, the presence of fossil shorelines around the lake  
57  
58 indicate that this site has indeed been subject to lake-level lowering and solute concentration as  
59  
60



1  
2  
3 well as expansion of purple sulphur bacteria around 1000 cal. yr BP as observed in other lakes  
4  
5 (e.g. SS4) in the region (McGowan *et al.*, 2008).  
6

7  
8 An unquantified but possibly important component of changing *m*-flux in the  
9  
10 Kangerlussuaq area is the increased regional deposition of loess that accompanied increased  
11  
12 aeolian activity since the neoglacial cooling (Willemse *et al.*, 2003). The area between SS1371  
13  
14 and the present ice sheet margin (Fig. 1) is subject to considerable aeolian input, sufficient to  
15  
16 dilute the organic content of lake sediments particularly during the Little Ice Age (Anderson *et*  
17  
18 *al.* 2012). The ecological effects of dust loading on remote lakes are well known due to their  
19  
20 nutrient content (Field *et al.*, 2010). The input of loess in the Kangerlussuaq area has been shown  
21  
22 to have had considerable impact on the biological communities of these lakes (Perren *et al.*,  
23  
24 2012; Bullard *et al.*, submitted).  
25  
26  
27  
28

### 29 30 *Mass flux: nutrient influx*

31  
32 The coastal and dilute lake (SS49) shows few changes in algal community composition prior to  
33  
34 ~1900 AD (50 cal. yr BP) when pigment concentration increased (Fig. 4). Primary production in  
35  
36 oligotrophic arctic lakes is dominated by relatively stable benthic communities, which can  
37  
38 contribute substantially (80–98%) to the primary production (Vadeboncoeur *et al.*, 2003). The  
39  
40 inference of a substantial benthic community at SS49 is supported by the stratigraphic record  
41  
42 with high concentrations of both chromophyte algal pigments and biogenic silica (the latter an  
43  
44 order of magnitude higher than the other lakes, Fig. 4), indicating the importance of diatoms.  
45  
46 Pigment indicators of green algae, higher plants, and cyanobacteria also likely originate from a  
47  
48 benthic community, based on contemporary regional lake surveys. Michelutti *et al.* (2005)  
49  
50 reported production increases in a number of arctic lakes, which they attributed primarily to  
51  
52 recent warming although increased nutrient availability is implicit. Oligotrophic lakes are  
53  
54 sensitive to even small increases in nutrient input due to the very low initial content (Leavitt *et*  
55  
56 *al.*, 2009) and there is now increasing evidence of the ecological effects of N-deposition at  
57  
58  
59  
60



1  
2  
3 remote arctic and alpine lakes (Galloway *et al.*, 2008; Holtgrieve *et al.*, 2011). Deposition rate of  
4  
5 NO<sub>x</sub> are not well prescribed for western Greenland, but are low (<0.5 kg N ha yr<sup>-1</sup>) although  
6  
7 loadings are presumably greater in coastal regions where precipitation is higher (Hasholt &  
8  
9 Sogaard, 1978).

10  
11 The  $\delta^{15}\text{N}$  data at SS49 represent a significant change in the N biogeochemistry of the lake  
12  
13 initiated at the beginning of the 20<sup>th</sup> Century onwards (Fig. 5). The trend towards more-depleted  
14  
15  $\delta^{15}\text{N}$  values is comparable with those observed in other Arctic and northern hemisphere  
16  
17 mountain lakes (e.g. Wolfe, Edwards & Aravena, 1999; Wolfe *et al.*, 2003; Holtgrieve *et al.*,  
18  
19 2011) and is consistent with increased inputs of anthropogenic N arising from fossil fuel  
20  
21 combustion. Such anthropogenic sources are generally isotopically depleted in <sup>15</sup>N, and ice core  
22  
23 records from Greenland document a progressive change from preindustrial  $\delta^{15}\text{N}$  values of ~11‰  
24  
25 to ca. -1‰ in NO<sub>3</sub><sup>-</sup> concomitant with a doubling in nitrate concentration in deposition during the  
26  
27 20<sup>th</sup> Century (Hastings *et al.*, 2009; Fig. 5). The agreement between  $\delta^{15}\text{N}$ , pigment PCA-1 and  
28  
29 the increasing organic C content since ~1880 is strongly supportive of a linear change in  
30  
31 productivity in response to increased nutrient input.  
32  
33  
34  
35

36 Both the interpretation of  $\delta^{15}\text{N}$  and PCA-1 of the pigments can be affected by diagenesis  
37  
38 (Leavitt, 1993; Talbot, 2001; Galman, Rydberg & Bigler, 2009) however, at SS49 the sediment  
39  
40 data are inconsistent with a diagenetic effect as the observed changes occur over longer time  
41  
42 frames (several decades) than expected with diagenesis. In general, pigments in the top 1-3 cm of  
43  
44 lake sediments are interpreted to be affected by post-depositional degradation (Leavitt, 1993).  
45  
46 Moreover, the anticipated isotopic effect of such processes would lead to an enrichment trend in  
47  
48  $\delta^{15}\text{N}$  (Galman *et al.*, 2009), the converse of that observed. Climate change in recent decades in  
49  
50 combination with its effects on changes in the catchment could be posited as a cause for the  
51  
52 observed  $\delta^{15}\text{N}$  record, however, following statistical analysis of the meteorological data  
53  
54 (Simpson, unpublished; Fig.5), no trend in 20<sup>th</sup> Century monthly mean temperature (at Nuuk)  
55  
56 was observed, a local pattern that is consistent with temperature trends throughout south west  
57  
58  
59  
60

1  
2  
3 Greenland (Box, 2002). Furthermore, the timing of recent Arctic warming is inconsistent with  
4  
5 the observed change in  $\delta^{15}\text{N}$  at this site.  
6

7 Nitrogen accrual is an important process in Arctic ecosystems largely associated with  
8  
9 terrestrial vegetation succession and the role of N-fixation by *Dryas* spp. and *Alnus* sp.  
10  
11 (Engstrom *et al.*, 2000). However, changes of the in-lake N pool can also result from N-fixation  
12  
13 by cyanobacteria which form extensive littoral mats in oligotrophic Arctic lakes. The soil  
14  
15 microbial and hydrological processes that can result in N transfer from land to water are complex  
16  
17 and highly seasonal due to the relationships between N-mineralization, snow melt and plant  
18  
19 uptake (Hobbie, Nadelhoffer & Hogberg, 2002). Moreover, these processes are hypothesized to  
20  
21 change with warming of the Arctic but presumably this is not important in the Kangerlussuaq  
22  
23 area given the relatively steady temperatures recorded for much of the 20th Century (Fig. 5). The  
24  
25 SS49 catchment is sparsely vegetated with thin soils, in contrast to inland catchments with more  
26  
27 extensive shrub tundra and thicker soils. The greater precipitation at the coast, coupled with thin  
28  
29 soils suggests that the sediment  $\delta^{15}\text{N}$  profile of SS49 is reflecting direct deposition of  $\text{NO}_x$  on to  
30  
31 the lake. Interestingly,  $\delta^{15}\text{N}$  profiles from sediment cores from inland lakes do not show the  
32  
33 characteristic depletion observed in many Arctic lakes (Simpson, unpublished).  
34  
35  
36  
37  
38  
39

#### 40 *Energy flux*

41  
42 SS1371 showed considerable short-term variability but little directional changes in phototrophic  
43  
44 community composition over time (Fig. 4). The pigment record at indicates input from higher  
45  
46 plants and green algae, probably including input from the extensive *Chara* beds in the littoral  
47  
48 zone, as well as chromophytes and the dominant phototrophic sulphur bacterial communities.  
49  
50 The continual dominance of phototrophic purple sulphur bacteria suggests that the lake was  
51  
52 meromictic or had prolonged periods of anoxic bottom water throughout the approximately 650-  
53  
54 year period covered by the core. Indicators of phototrophic sulphur bacteria often are observed in  
55  
56 both saline and freshwater lakes where light penetrates to anoxic waters and have been used  
57  
58  
59  
60

1  
2  
3 previously to infer major shifts in lake status (Leavitt, Carpenter & Kitchell, 1989; Vinebrooke *et*  
4 *al.*, 1998; Pienitz *et al.*, 2000; Squier, Hodgson & Keely, 2002; McGowan *et al.*, 2008). The  
5 inference of meromictic conditions in the modern lake is supported by temperature thermistors  
6 deployed over 5 years at SS1371 (Anderson, unpublished). Strong stratification and good light  
7 transparency could promote the high-frequency variability in community structure, due to tight  
8 coupling between stratification and the phototrophic community (e.g. Pfennig, 1989; Vila *et al.*,  
9 1998). While the continued negative precipitation balance and resulting decreasing lake level  
10 inferred from fossil shorelines (Aebly & Fritz, 2009) over the last ~1000 years is the primary  
11 cause of the establishment of closed basin systems in the area it cannot explain the observed  
12 variability of the phototrophic community. On the other hand, considerable changes in  
13 temperature has been inferred from the ice cores during this period (Dahl-Jensen *et al.*, 1998)  
14 and direct E-flux is therefore inferred as the primary controlling mechanism of the lake response  
15 (Fig. 6).  
16  
17  
18  
19  
20  
21  
22  
23  
24  
25  
26  
27  
28  
29  
30  
31

32 At the nunatak lake (SS86), dominance of green sulphur bacterial pigments in the upper  
33 half of the sediment core (Fig. 4) also indicate chemical stratification and light penetration into  
34 an anoxic hypolimnion (Pfennig 1989). The observed changes in the phototrophic community  
35 and geochemical markers suggest a transition from a dilute, oligotrophic lake to one with  
36 stronger seasonal chemical stratification, hypolimnic anoxia, and substantial phototrophic  
37 bacterial production over the last ~1000 years. A change in the main controlling mechanism of  
38 this lake from direct mass (precipitation) to energy (Fig. 6) is inferred in accordance with  
39 interpretations of trends at the other lakes in this study.  
40  
41  
42  
43  
44  
45  
46  
47  
48  
49  
50

### 51 *Synthesis*

52 Changes in the phototrophic community in the Southwest Greenland lakes are driven by a  
53 combination of factors, including direct and indirect E and *m* forcing (e.g. radiative forcing,  
54 temperature, effective precipitation, ice-free period), mediated by location, catchment:lake ratios,  
55  
56  
57  
58  
59  
60

1  
2  
3 and in-lake processes (e.g. stratification). Although precipitation levels are low in much of the  
4  
5 Arctic, with associated reduced hydrological fluxes from land to lake, catchment processes are  
6  
7 still important and will mediate  $m$  flux to the lake. For example, Anderson *et al.* (2008) found  
8  
9 significant effects on biological structure associated with the arrival of *Betula nana* in SW  
10  
11 Greenland through sequestration of nutrients. Any assessment of “drivers” of limnological  
12  
13 change in the Arctic should include a terrestrial/vegetation component (Wookey *et al.*, 2009) but  
14  
15 conceptual models that have been developed to account for recent biological change at high  
16  
17 latitudes have tended to downplay alternative hypotheses such as catchment processes (Smol &  
18  
19 Douglas 2007). Moreover, as well as changing E and  $m$ -fluxes, in-lake processes (altered trophic  
20  
21 interactions, benthic-pelagic coupling) and other ontogenetic processes (long-term accumulation  
22  
23 of salts, dissolved organic carbon) may have considerable influence on biological structure  
24  
25 independent of climate (Anderson *et al.*, 2004), therefore making it difficult to infer  
26  
27 unambiguously climate from biological remains in lake sediments (Lotter & Birks, 2003). The  
28  
29 contrasting responses of the three lakes in this study to global environmental change processes  
30  
31 over the last ~1600 years highlights the need to consider greater regional variability as we  
32  
33 attempt to disentangle the ecological response of arctic lakes to multiple stressors. This is true  
34  
35 even in a relatively small area (<150 km) such as the Kangerlussuaq lake district, which is a  
36  
37 reasonably homogenous in terms of geology and vegetation. As highlighted by Kaufman (2012),  
38  
39 recognizing the spatial complexity of lakes to regional climate forcing and the associated non-  
40  
41 climatic filters is critical if lake sediment records are to be used to reconstruct past-climate  
42  
43 variability in the Arctic. There is clearly a need for greater replication at the regional scale to  
44  
45 help identify signals of environmental change.  
46  
47  
48  
49  
50

## 51 52 53 **Acknowledgements**

54  
55 We are grateful to I. Renberg during coring and sub-sampling of the cores, B. Perren for help  
56  
57 during sub-sampling, and B. Møller for assistance in the lab. We also thank P. Appleby for  
58  
59  
60

1  
2  
3 conducting the  $^{210}\text{Pb}$  dating. A. Lami and R. Cox kindly provided standards of purple and green  
4  
5 sulphur bacteria, respectively. L. Schlüter provided assistance for separation of bacterial  
6  
7 pigments from culture. Funding for field collection of cores and dating was provided by a U.S.  
8  
9 NSF grant (ATM-0081226) to S. Fritz. Additional fieldwork and analysis was funded by grants  
10  
11 from the Danish National Science Foundation (SNF 21-02-0390 and SNF 51-00-0288) to N.J.  
12  
13 Anderson and a STENO-grant from The Danish Council for Independent Research - Natural  
14  
15 Sciences (FNU) to N. S. Reuss (09-064952).  
16  
17  
18  
19  
20  
21  
22  
23  
24  
25  
26  
27  
28  
29  
30  
31  
32  
33  
34  
35  
36  
37  
38  
39  
40  
41  
42  
43  
44  
45  
46  
47  
48  
49  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60

Copy for Review

## References

Aebly F.A. & Fritz S.C. (2009) Palaeohydrology of Kangerlussuaq (Søndre Strømfjord), West Greenland during the last similar to 8000 years. *Holocene*, **19**, 91-104.

Anderson N.J., Brodersen K.P., Ryves D.B., McGowan S., Johansson L.S., Jeppesen E. & Leng M.J. (2008) Climate versus in-lake processes as controls on the development of community structure in a low-arctic lake (South-West Greenland). *Ecosystems*, **11**, 307-324.

Anderson N.J., Harriman R., Ryves D.B. & Patrick S.T. (2001) Dominant factors controlling variability in the ionic composition of West Greenland Lakes. *Arctic Antarctic and Alpine Research*, **33**, 418-425.

Anderson N.J., Liverside A.C., McGowan S. & Jones M.D. (2012) Lake and catchment response to Holocene environmental change: spatial variability along a climate gradient in southwest Greenland. *Journal of Paleolimnology*, **48**, 209-222.

Anderson N.J., Ryves D.B., Grauert M. & McGowan S. (2004) Holocene paleolimnology of Greenland and the north Atlantic islands (north of 60°N). In: *Long-term environmental change in Arctic and Antarctic lakes*, 319-347, Springer, Netherlands.

Anderson N.J. & Stedmon C.A. (2007) The effect of evapoconcentration on dissolved organic carbon concentration and quality in lakes of SW Greenland. *Freshwater Biology*, **52**, 280-289.

Appleby P.G. (2001) Chronostratigraphic techniques in recent sediments. In: *Tracking Environmental Change Using Lake Sediments, Basin Analysis, Coring, and Chronological Techniques, vol. 1*, 171-203, Kluwer Academic Publishers, Dordrecht.

Appleby P.G., Nolan P.J., Gifford D.W., Godfrey M.J., Oldfield F., Anderson N.J. & Battarbee R.W. (1986) Pb-210 dating by low background gamma-counting. *Hydrobiologia*, **143**, 21-27.

Appleby P.G. & Oldfield F. (1978) The calculation of <sup>210</sup>Pb dates assuming a constant rate of supply of unsupported <sup>210</sup>Pb to the sediment. *Catena*, **5**, 1-8.

Battarbee R.W. (2000) Palaeolimnological approaches to climate change, with special regard to the biological record. *Quaternary Science Reviews*, **19**, 107-124.

Bergstrom A.K., Blomqvist P. & Jansson M. (2005) Effects of atmospheric nitrogen deposition on nutrient limitation and phytoplankton biomass in unproductive Swedish lakes. *Limnology And Oceanography*, **50**, 987-994.

Bindler R., Renberg I., Anderson N.J., Appleby P.G., Emteryd O. & Boyle J. (2001) Pb isotope ratios of lake sediments in West Greenland: inferences on pollution sources. *Atmospheric Environment*, **35**, 4675-4685.

Bonilla S., Villeneuve V. & Vincent W.F. (2005) Benthic and planktonic algal communities in a High Arctic Lake: Pigment structure and contrasting responses to nutrient enrichment. *Journal of Phycology*, **41**, 1120-1130.

Box J.E. (2002) Survey of Greenland instrumental temperature records: 1873-2001. *International Journal of Climatology*, **22**, 1829-1847.

1  
2  
3 Brodersen K.P. & Anderson N.J. (2000) Subfossil insect remains (Chironomidae) and lake-water  
4 temperature inference in the Sisimiut Søndre Strømfjord region, southern West Greenland.  
5 *Geology of Greenland Survey Bulletin* 186: 78-82.

6  
7 Burton H.R. (1981) Chemistry, physics and evolution of antarctic saline lakes - a review.  
8 *Hydrobiologia*, **81**, 339-362.

9  
10 Conley D.J. (1998) An interlaboratory comparison for the measurement of biogenic silica in  
11 sediments. *Marine Chemistry*, **63**, 39-48.

12  
13 Conley D.J. & Schelske C.L. (2001) Biogenic silica. In: *Tracking environmental change using*  
14 *lake sediments. Volume 3: Terrestrial, algal, and siliceous indicators.*, 281-293, Kluwer  
15 Academic Publishers, Dordrecht.

16  
17  
18 D'Andrea W.J., Huang Y., Fritz S.C. & Anderson N.J. (2011) Abrupt Holocene climate change  
19 as an important factor for human migration in West Greenland. *Proc.Nat.Acad.Sci.*, **108**, 9765-  
20 9769.

21  
22 DeMaster D.J. (1981) The supply and accumulation of silica in the marine environment.  
23 *Geochimica et Cosmochimica Acta*, **45**, 1715-1732.

24  
25 Dahl-Jensen D., Mosegaard K., Gundestrup N., Clow G.D., Johnsen S.J., Hansen A.W. &  
26 Balling N. (1998) Past temperatures directly from the Greenland Ice Sheet. *Science*, **282**, 268-  
27 271.

28  
29 Engstrom D.R., Fritz S.C., Almendinger J.E. & Juggins S. (2000) Chemical and biological trends  
30 during lake evolution in recently deglaciated terrain. *Nature*, **408**, 161-166.

31  
32 Field J.P., Belnap J., Breshears D.D., Neff J.C., Okin G.S., Whicker J.J., et al. (2010) The  
33 ecology of dust. *Frontiers in Ecology and the Environment*, **8**, 423-430.

34  
35 Frigaard N.U., Larsen K.L. & Cox R.P. (1996) Spectrochromatography of photosynthetic  
36 pigments as a fingerprinting technique for microbial phototrophs. *Fems Microbiology Ecology*,  
37 **20**, 69-77.

38  
39 Fritz S.C. (1996) Paleolimnological records of climatic change in North America. *Limnology*  
40 *And Oceanography*, **41**, 882-889.

41  
42 Galloway J.N., Townsend A.R., Erisman J.W., Bekunda M., Cai Z., Freney J.R., Martinelli L.A.,  
43 Seitzinger S.P. & Sutton M.A. (2008) Transformation of the nitrogen cycle: Recent trends,  
44 questions, and potential solutions. *Science*, **320**, 889-892.

45  
46 Galman V., Rydberg J. & Bigler C. (2009) Decadal diagenetic effects on delta(13)C and  
47 delta(15)N studied in varved lake sediment. *Limnology And Oceanography*, **54**, 917-924.

48  
49 Hasholt B. & Søgaard H. (1978) Et forsøg på en klimatisk-hydrologisk regionsinddeling af  
50 Holsteinsborg kommune (Sisimiut). *Geografisk Tidsskrift*, **77**, 72-92.

51  
52 Hastings M., Jarvis J. & Steig E. (2009) Anthropogenic Impacts on Nitrogen Isotopes of Ice-  
53 Core Nitrate. *Science*, **324**, 1288.

54  
55 Heggen M.P., Birks H.H. & Anderson N. (2010) Long-term ecosystem dynamics of a small lake  
56 and its catchment in west Greenland. *Holocene*, **20**, 1207-1222.



- 1  
2  
3 Hobbie S.E., Nadelhoffer K.J. & Hogberg P. (2002) A synthesis: The role of nutrients as  
4 constraints on carbon balances in boreal and arctic regions. *Plant and Soil*, **242**, 163-170.  
5  
6 Holtgrieve G.W., Schindler D.E., Hobbs W.O., Leavitt P.R., Ward E.J., Bunting L., *et al.* (2011)  
7 A Coherent Signature of Anthropogenic Nitrogen Deposition to Remote Watersheds of the  
8 Northern Hemisphere. *Science*, **334**, 1545-1548.  
9  
10 Jeffrey S.W., Mantoura R.F.C. & Wright S.W. (1997) *Phytoplankton pigments in oceanography*,  
11 UNESCO Publishing, Paris.  
12  
13 Kane D.L., Hinzman L.D., Woo M. & Everett K.R. (1992) Arctic hydrology and climate change.  
14 In: *Arctic ecosystems in a changing climate*, 35-57, Academic Press, Inc., San Diego, California.  
15  
16 Kaufman D.S. (2012) Introduction to the JoPL special issue, "Holocene paleoenvironmental  
17 records from Arctic lake sediment". *Journal of Paleolimnology*, **48**, 1-7.  
18  
19 Leavitt P.R. (1993) A review of factors that regulate carotenoid and chlorophyll deposition and  
20 fossil pigment abundance. *Journal of Paleolimnology*, **9**, 109-127.  
21  
22 Leavitt P.R., Carpenter S.R. & Kitchell J.F. (1989) Whole-lake experiments: the annual record of  
23 fossil pigments and zooplankton. *Limnol. Oceanogr.*, **34**, 700-717.  
24  
25 Leavitt P.R., Findlay D.L., Hall R.I. & Smol J.P. (1999) Algal responses to dissolved organic  
26 carbon loss and pH decline during whole-lake acidification: Evidence from paleolimnology.  
27 *Limnology And Oceanography*, **44**, 757-773.  
28  
29 Leavitt P.R., Fritz S.C., Anderson N.J., Baker P.A., Blenckner T., Bunting L., *et al.* (2009)  
30 Paleolimnological evidence of the effects on lakes of energy and mass transfer from climate and  
31 humans. *Limnology And Oceanography*, **54**, 2330-2348.  
32  
33 Leavitt P.R. & Hodgson D.A. (2001) Sedimentary pigments. In: *Tracking environmental change*  
34 *using lake sediments. Volume 3: Terrestrial, algal, and siliceous indicators.*, 295-325, Kluwer  
35 Academic Publishers, Dordrecht, The Netherlands.  
36  
37 Lotter A.F. & Birks H.J.B. (2003) The Holocene palaeolimnology of Sagistalsee and its  
38 environmental history - a synthesis. *Journal of Paleolimnology*, **30**, 333-342.  
39  
40 McGowan S., Juhler R.K. & Anderson N.J. (2008) Autotrophic response to lake age,  
41 conductivity and temperature in two West Greenland lakes. *Journal of Paleolimnology*, **39**, 301-  
42 317.  
43  
44 McGowan S., Ryves D.B. & Anderson N.J. (2003) Holocene records of effective precipitation in  
45 West Greenland. *The Holocene*, **13**, 239-249.  
46  
47 Michelutti N., Wolfe A.P., Vinebrooke R.D., Rivard B. & Briner J.P. (2005) Recent primary  
48 production increases in arctic lakes. *Geophysical Research Letters*, **32**, L19715-  
49 Doi:10.1029/2005GL023693.  
50  
51 Ouellet M., Bisson M., Page P. & Dickman M. (1987) Physicochemical limnology of meromictic  
52 saline Lake Sophia, Canadian arctic archipelago. *Arctic and Alpine Research*, **19**, 305-312.  
53  
54 Overpeck J., Hughen K., Hardy D., Bradley R., Case R., Douglas M., *et al.* (1997) Arctic  
55 environmental change of the last four centuries. *Science*, **278**, 1251-1256.  
56  
57  
58  
59  
60



- 1  
2  
3 Perren B.B., Anderson N.J., Douglas M.S.V. & Fritz S. (2012) The influence of temperature,  
4 moisture, and eolian activity on Holocene lake development in West Greenland. *Journal of*  
5 *Paleolimnology*, **48**, 223-239.  
6
- 7 Perren B.B., Douglas M.S.V. & Anderson N.J. (2009) Diatoms reveal complex spatial and  
8 temporal patterns of recent limnological change in West Greenland. *Journal of Paleolimnology*,  
9 **42**, 233-247.  
10
- 11 Pfennig N. (1989) Ecology of phototrophic purple and green sulfur bacteria. In: *Autotrophic*  
12 *Bacteria*, 97-116, Science Tech Publ., Madison, WI.  
13
- 14 Pienitz R., Smol J.P., Last W.M., Leavitt P.R. & Cumming B.F. (2000) Multi-proxy Holocene  
15 palaeoclimatic record from a saline lake in the Canadian Subarctic. *Holocene*, **10**, 673-686.  
16
- 17 Pla S. & Anderson N.J. (2005) Environmental factors correlated with chrysophyte cyst  
18 assemblages in low arctic lakes of southwest Greenland. *Journal of Phycology*, **41**, 957-974.  
19
- 20 Post E., Forchhammer M.C., Bret-Harte M.S., Callaghan T.V., Christensen T.R., Elberling B. *et*  
21 *al.*, (2009) Ecological dynamics across the Arctic associated with recent climate change. *Science*,  
22 **325**, 1355-1358.  
23
- 24 Quinlan R., Douglas M.S.V. & Smol J.P. (2005) Food web changes in arctic ecosystems related  
25 to climate warming. *Global Change Biology*, **11**, 1381-1386.  
26
- 27 Reuss N. & Conley D.J. (2005) Effects of sediment storage conditions on pigment analyses.  
28 *Limnology and Oceanography-Methods*, **3**, 477-487.  
29
- 30 Reuss N., Hammarlund D., Rundgren M., Segerström U., Eriksson L. & Rosén P. (2010a) Lake  
31 Ecosystem Responses to Holocene Climate Change at the Subarctic Tree-Line in Northern  
32 Sweden. *Ecosystems*, **13**, 393-409.  
33
- 34 Reuss N., Leavitt P., Hall R., Bigler C. & Hammarlund D. (2010b) Development and application  
35 of sedimentary pigments for assessing effects of climatic and environmental changes on  
36 subarctic lakes in northern Sweden. *Journal of Paleolimnology*, **43**, 149-169.  
37
- 38 Smol J.P. & Douglas M.S.V. (2007) From controversy to consensus: making the case for recent  
39 climate change in the Arctic using lake sediments. *Front Ecol Environ*, **5**, 466-474.  
40
- 41 Smol J.P., Wolfe A.P., Birks H.J.B., Douglas M.S.V., Jones V.J., Korhola A., *et al.* (2005)  
42 Climate-driven regime shifts in the biological communities of arctic lakes. *Proceedings of the*  
43 *national academy of sciences of the United States of America PNAS*, **102**, 4397-4402.  
44
- 45 Squier A.H., Hodgson D.A. & Keely B.J. (2002) Sedimentary pigments as markers for  
46 environmental change in an Antarctic lake. *Organic Geochemistry*, **33**, 1655-1665.  
47
- 48 Talbot M.R. (2001) Nitrogen isotopes in palaeolimnology. In: *Tracking Environmental Change*  
49 *Using Lake Sediments. Volume 2: Physical and Geochemical Methods*, 401-439, Kluwer  
50 Academic Publishers, Dordrecht, The Netherlands.  
51
- 52 Vadeboncoeur Y., Jeppesen E., Vander Zanden M.J., Schierup H.H., Christoffersen K. & Lodge  
53 D.M. (2003) From Greenland to green lakes: cultural eutrophication and the loss of benthic  
54 pathways in lakes. *Limnology And Oceanography*, **48**, 1408-1418.  
55  
56  
57  
58  
59  
60

1  
2  
3 Vila X., Abella C.A., Figueras J.B., Hurley J.P. (1998) Vertical models of phototrophic bacterial  
4 distribution in the metalimnetic microbial communities of several freshwater North-American  
5 kettle lakes. *Fems Microbiology Ecology*, **25**, 287-299.

6  
7 Vincent W.F., Hobbie J.E. & Laybourn-Parry J. (2008) Introduction to the limnology of high-  
8 latitude lake and river ecosystems. In: Polar Lakes and Rivers (Eds W.F. Vincent & J. Laybourn-  
9 Parry), pp. 1-24. Oxford University Press, New York.

10  
11 Vinebrooke R.D., Hall R.I., Leavitt P.R. & Cumming B.F. (1998) Fossil pigments as indicators  
12 of phototrophic response to salinity and climatic change in lakes of western Canada. *Canadian*  
13 *Journal of Fisheries and Aquatic Sciences*, **55**, 668-681.

14  
15 Willemse N.W., van Dam O., van Helvoort P.J., Dankers R., Brommer M., Schokker J., Valstar  
16 T.E. & de Wolf H. (2004) Physical and chemical limnology of a subsaline athalassic lake in  
17 West Greenland. *Hydrobiologia*, **524**, 167-192.

18  
19 Willemse N.W., Koster E.A., Hoogakker B. & van Tatenhove F.G.M. (2003) A continuous  
20 record of Holocene eolian activity in West Greenland. *Quaternary Research*, **59**, 322-334.

21  
22 Williams W.D. (1991) Comments on the so-called salt lakes of Greenland. *Hydrobiologia*, **210**,  
23 67-74.

24  
25 Wolfe A.P., Cooke C.A. & Hobbs W.O. (2006) Are current rates of atmospheric nitrogen  
26 deposition influencing lakes in the Eastern Canadian Arctic? *Arctic Antarctic and Alpine*  
27 *Research*, **38**, 465-476.

28  
29 Wolfe B.B., Edwards T.W.D. & Aravena R. (1999) Changes in carbon and nitrogen cycling  
30 during tree-line retreat recorded in the isotopic content of lacustrine organic matter, western  
31 Taimyr Peninsula, Russia. *Holocene*, **9**, 215-222.

32  
33 Wolfe B.B., Edwards T.W.D., Jiang H., MacDonald G.M., Gervais B.R. & Snyder J.A. (2003)  
34 Effect of varying oceanicity on early- to mid-Holocene palaeohydrology, Kola Peninsula,  
35 Russia: isotopic evidence from treeline lakes. *The Holocene*, **13**, 153-160.

36  
37 Wright S.W., Jeffrey S.W., Mantoura R.F.C., Llewellyn C.A., Bjørnland T., Repeta D. &  
38 Welschmeyer N. (1991) Improved HPLC method for the analysis of chlorophylls and  
39 carotenoids from marine phytoplankton. *Marine Ecology Progress Series*, **77**, 183-196.

40  
41 Wookey P.A., Aerts R., Bardgett R.D., Baptist F., Brathen K.A., Cornelissen J.H.C., *et al.*  
42 (2009) Ecosystem feedbacks and cascade processes: understanding their role in the responses of  
43 Arctic and alpine ecosystems to environmental change. *Global Change Biology*, **15**, 1153-1172.

44  
45 Züllig H. (1985) Pigmente phototropher Bakterien in Seesedimenten und ihre Bedeutung für die  
46 Seenforschung. *Schweizerische Zeitschrift für Hydrologie*, **47**, 87-126.

Table 1. Mean morphological and water chemistry characteristics of the lakes shown in Fig. 1 and other sites referred to in the text (SS32, SS4). For comparative purposes the mean water chemistry of three groups of lakes located along the fjord are given (Fig. 1) (see Anderson *et al.*, 2001; Pla & Anderson, 2003 for details). The formal name for SS4 is Braya Sø.

		Mean chemistry – lake groups			Study lakes				
					Coastal	Oligosaline	Nunatak		
	Units	Coastal	Central: head of fjord	Ice margin	SS49	SS1371	SS86	SS32	SS4
Latitude	°N	66.77	66.97	67.05	66.86	67.06	66.96	66.97	66.99
Longitude	°W	52.67	51.26	50.33	52.66	51.13	49.81	49.8	51.04
Altitude	m	240.6	257.6	394.1	320	150	470	470	170
Z max	m	11	14.9	15.8	10.5	22	13	21	23
Area	ha	13.9	28.9	18.7	20.7	21.7	5	16.3	73
pH		7	7.9	7.6	6.63	8.8	8.26	7.3	9
Cond.	µS cm <sup>-1</sup>	50	298.3	119.2	26.7	3080	323	48	2540
TN	µg L <sup>-1</sup>	208	717.8	455.6	143.3	2150	587	308	803
TP	µg L <sup>-1</sup>	4.3	6.8	8	3.7	14	3.6	5	9
Ca	µeq L <sup>-1</sup>	225.2	1257.4	416.8	123	3422	1256	215	1194
Mg <sup>2+</sup>	µeq L <sup>-1</sup>	113.7	1012.1	490.3	48	14227	1956	233	13544
Na <sup>+</sup>	µeq L <sup>-1</sup>	153.1	738.1	157.9	100	15308	619	81	14814
K <sup>+</sup>	µeq L <sup>-1</sup>	24.2	232.6	75.3	8.7	4363	277	33.7	3091
Alk	µeq L <sup>-1</sup>	241.6	1953.6	811.7	86.7	8860	3060	367	13102
SO <sub>4</sub> <sup>2-</sup>	µeq L <sup>-1</sup>	65.8	194.1	20.6	30.7	8395	3.5	32	1679
Cl	µeq L <sup>-1</sup>	138.7	702.3	131.2	102	26150	498	55.7	12515
DOC	mg L <sup>-1</sup>	4.8	53.8	9.2	4.8	83	30.7	7.6	90

## Figure legends

Figure 1. Location of the study sites (SS49, SS1371, SS86) along the Kangerlussuaq fjord (Southwest Greenland) on a transect from the sea to the ice sheet margin, together with the location of two additional lakes referred to in the text (SS4, SS32; see Table 1). The rectangles indicate the approximate location of the three lake groups referred to in Table 1 (coastal, central, ice margin) for comparative purposes.

Figure 2.  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  profiles for SS49 (left) and SS86 (right). Uppermost panels are total and supported  $^{210}\text{Pb}$  activity; the central panels are unsupported  $^{210}\text{Pb}$  while the lowermost plots show the  $^{137}\text{Cs}$  and  $^{241}\text{Am}$  at SS86, the latter indicating the well resolved radionuclide profiles despite the low sedimentation rates.

Figure 3. Cross correlation of freeze and Russian cores. A: SS86 (Nunatak lake), B: SS1371 (oligosaline lake), C: SS49 (coastal lake).  $^{14}\text{C}$  dates used for constructing an age-depth curve are included in A, while they fall outside the range of the plot in B (for further details see D'Andrea *et al.*, 2011). In C the bottom  $^{210}\text{Pb}$  date and one of two  $^{14}\text{C}$  dates are included in the plot while all three points have been used to construct the age-depth curve for samples below 11cm. Note that SS1371 is correlated by %OC (organic carbon) not %TC (total carbon) since there were significant amounts of inorganic carbon in the sediment from this lake.

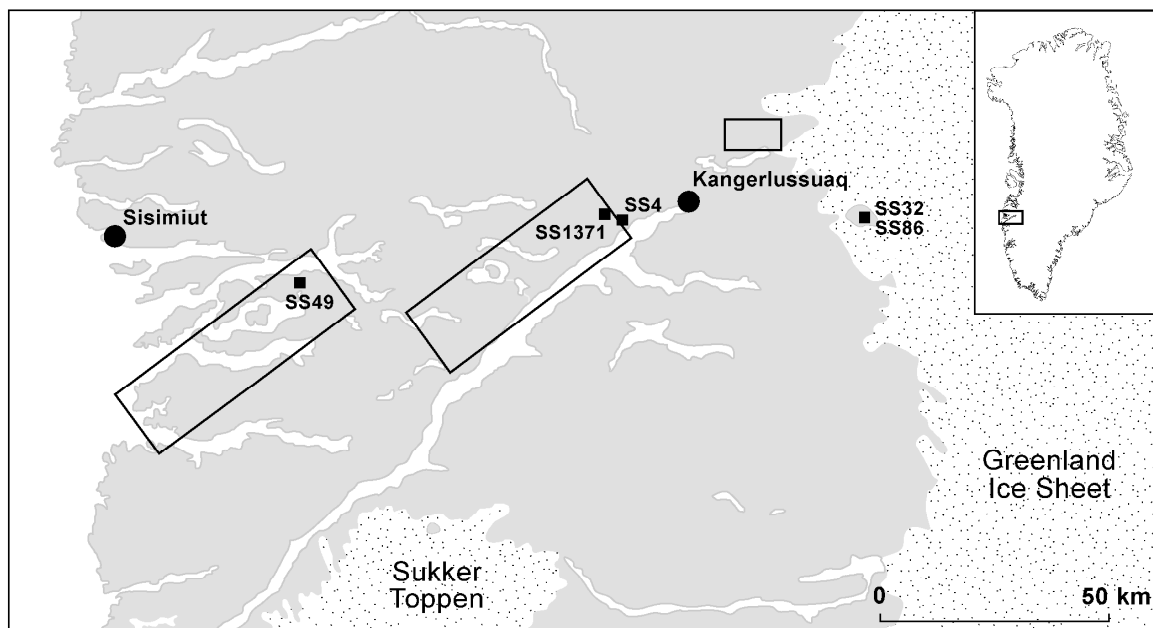
Figure 4. Sediment pigment record and geochemical proxies of the study lakes. Biogenic silica (BSi), organic carbon (OC), and total nitrogen (TN) presented as weight percentage, OC/TN as molar ratio. Chlorophylls (chlorophyll-a and b, pheophytins-a and b) and carotenoids (the remainder of the pigments) as concentration in  $\mu\text{mol g}^{-1}\text{OC}$ . The ratio of chl

1  
2  
3 a/ppn a is included as a degradation signal. Note different scales on x-axis. A) Coastal lake  
4 SS49. B) Oligosaline lake SS1371, Okenone total include the cis- and trans-peak,  
5 pheophytins-a and b include pheophytin and pyro-pheophytin as they are both important  
6 degradation products in this lake. C) Nunatak lake SS86, Bchl-e total includes 8 homologs of  
7 bacteriochlorophyll-e.  
8  
9  
10  
11  
12  
13  
14  
15  
16

17 Figure 5. A:  $\delta^{15}\text{N}$  and pigment PCA-1 at SS49 plotted against  $^{210}\text{Pb}$  timescale; B:  $\delta^{15}\text{N}$  and  
18  $\text{NO}_3$  in the Greenland ice core (Hastings *et al.*, 2009) and C: mean annual temperature at  
19 Nuuk, showing representative trends in air temperature for SW Greenland since 1860.  
20  
21  
22  
23  
24  
25  
26  
27

28 Figure 6. Conceptual interpretation of the relative importance of energy (E) and mass (*m*) in  
29 controlling lake response (summarized by PCA axis 1 and BSi) to environmental change  
30 according to Leavitt *et al.*, 2009. While direct influx of *m* through precipitation is inferred to  
31 have a strong control of all lakes before ~1000 cal yr BP (A), direct influx of E is inferred to  
32 have a greater influence in closed basin systems (SS1371 and SS86 after ~1000 cal yr  
33 BP)(B). SS49 has not been included in the plot due to no change in the main controlling  
34 factor during the investigated time period. Supplementary palaeolimnological proxies have  
35 been included for comparison; the temperature reconstruction from the Greenland GRIP and  
36 Dye-3 ice cores (Dahl-Jensen *et al.*, 1998), regional lake-levels (Aebly & Fritz, 2009), and  
37 diatom-inferred conductivity from SS4 (McGowan *et al.*, 2003).  
38  
39  
40  
41  
42  
43  
44  
45  
46  
47  
48  
49  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60

Figure 1, Reuss et al.



for Review

1  
2  
3  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13  
14  
15  
16  
17  
18  
19  
20  
21  
22  
23  
24  
25  
26  
27  
28  
29  
30  
31  
32  
33  
34  
35  
36  
37  
38  
39  
40  
41  
42  
43  
44  
45  
46  
47  
48  
49  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60

Figure 2. Reuss et al.

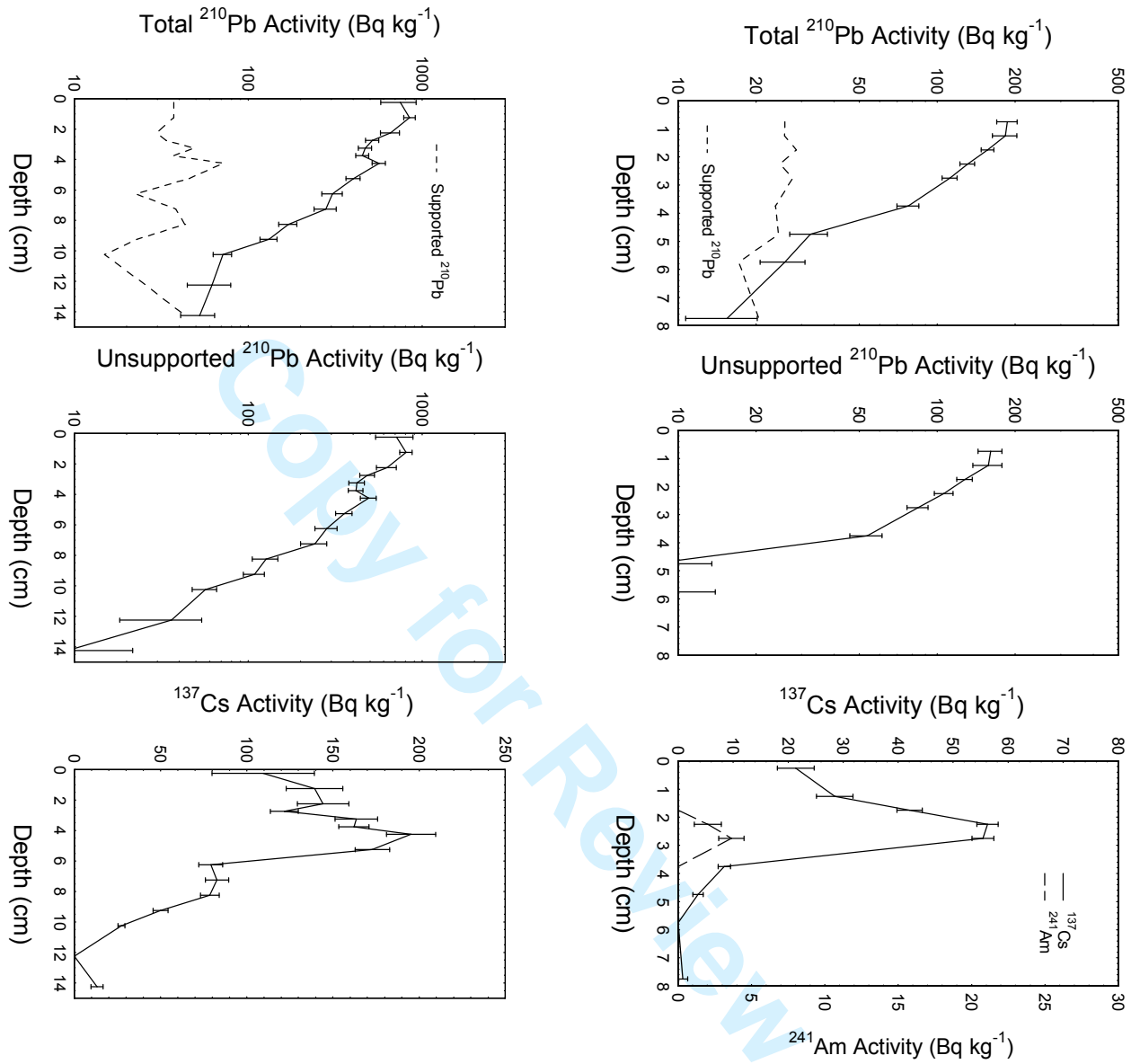
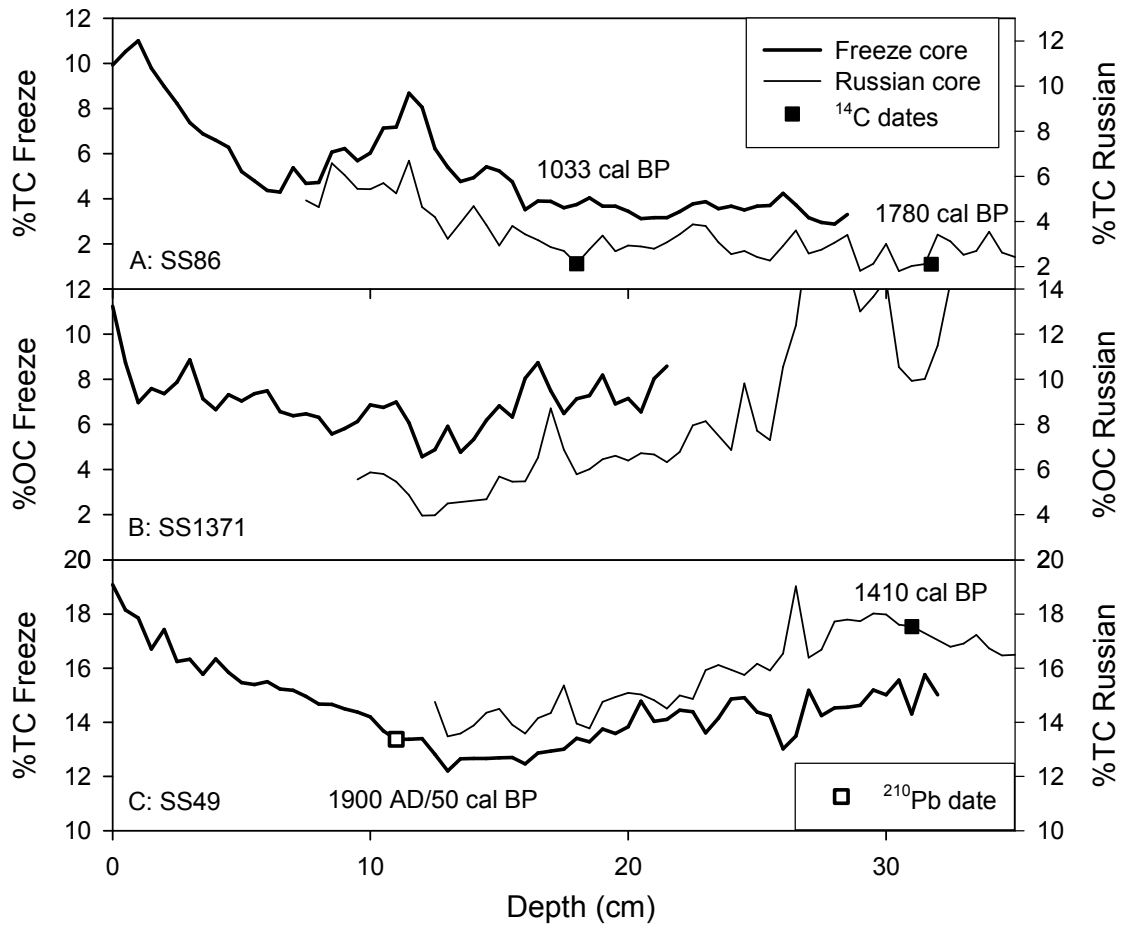


Figure 3, Reuss et al.



1  
2  
3  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13  
14  
15  
16  
17  
18  
19  
20  
21  
22  
23  
24  
25  
26  
27  
28  
29  
30  
31  
32  
33  
34  
35  
36  
37  
38  
39  
40  
41  
42  
43  
44  
45  
46  
47  
48  
49  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60



Figure 4, Reuss et al.

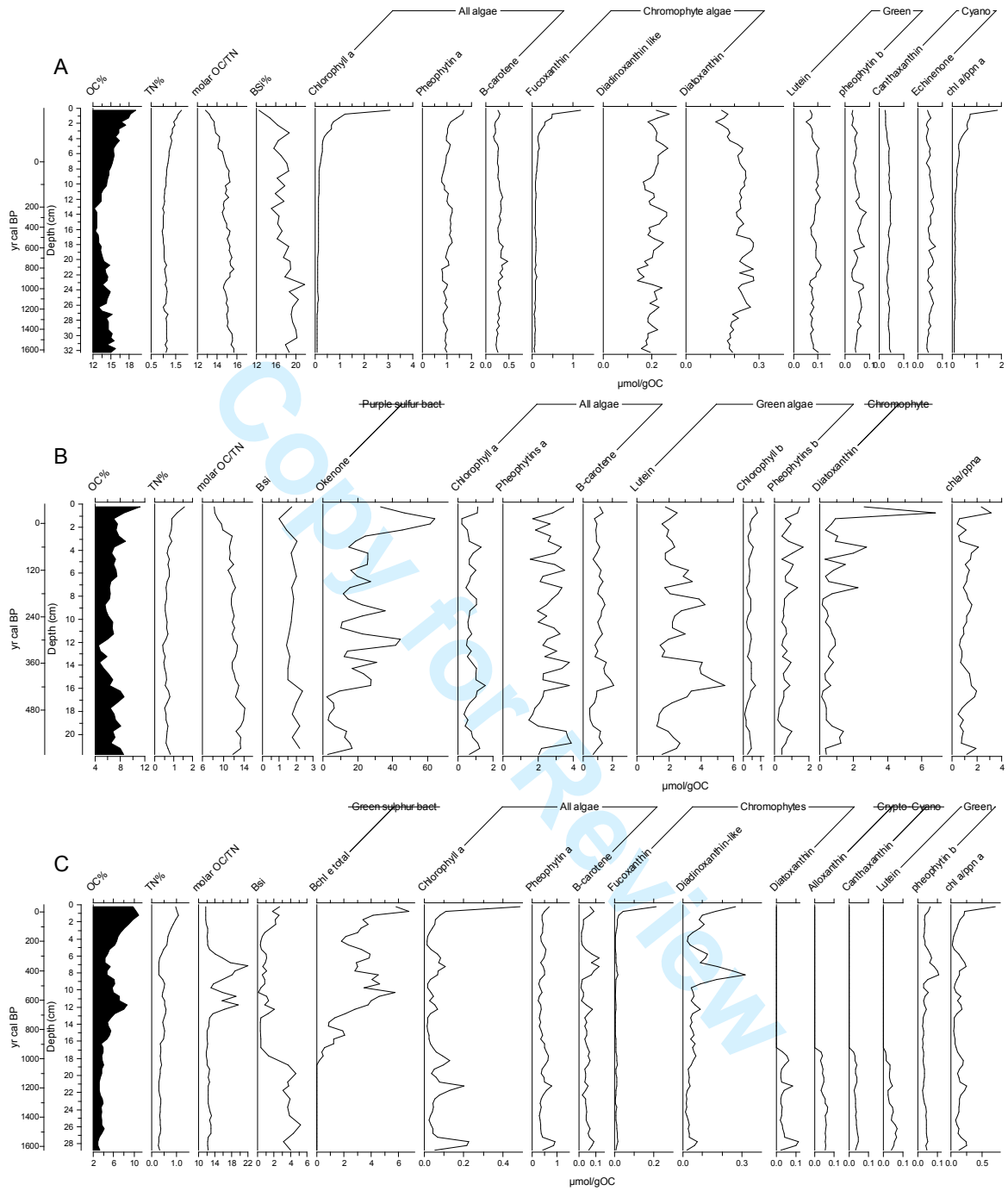


Figure 5, Reuss et al.

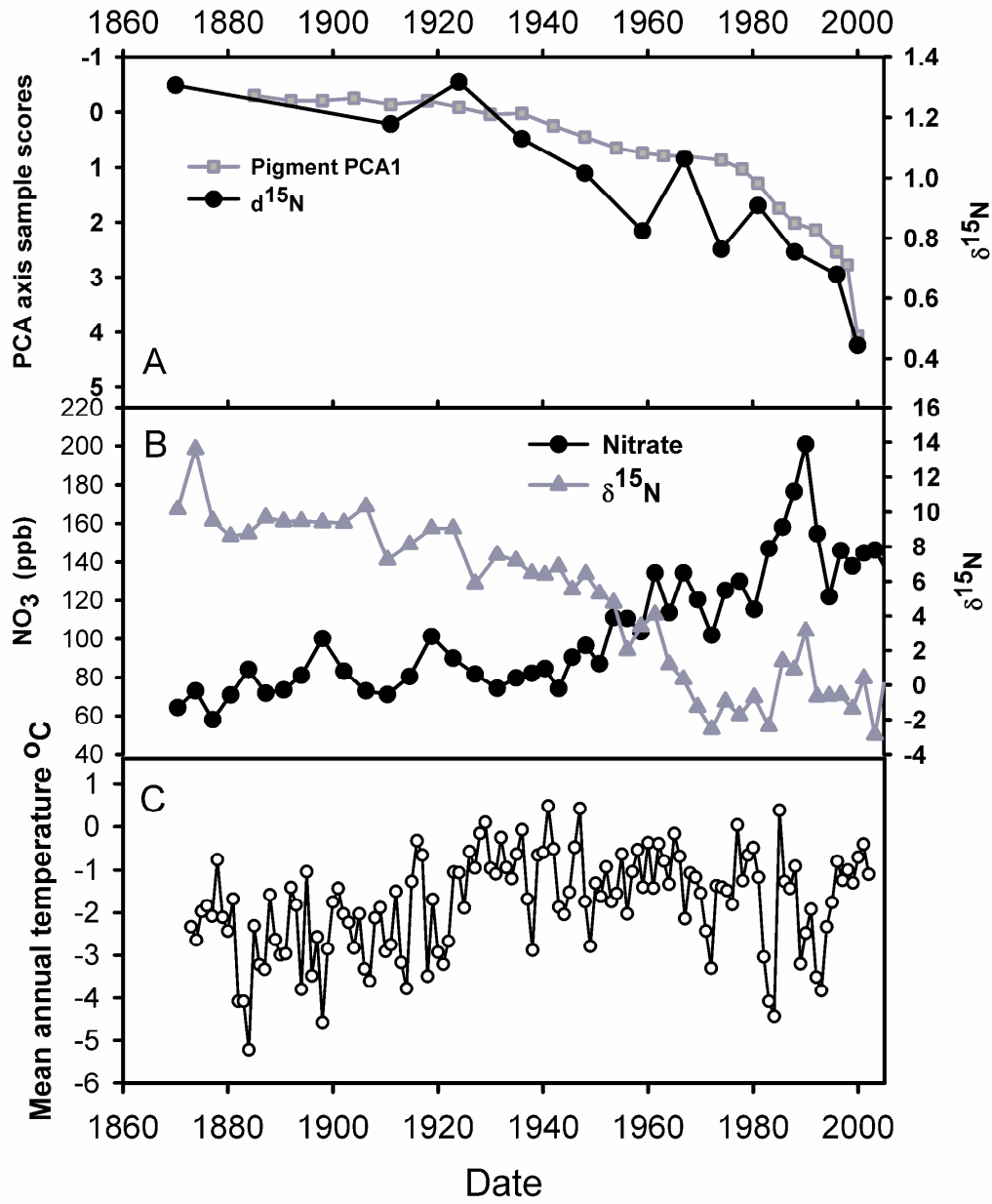


Figure 6, Reuss et al.

