Characterising organic carbon sources in Anthropocene affected Arctic upland lake catchments, Disko Island, West Greenland

Mark A. Stevenson[1,2*], Suzanne McGowan[1], Emma J. Pearson[3], George E.A. Swann[1], Melanie J. Leng[4,5], Vivienne. J. Jones[6], Joseph J. Bailey[1,7#], Xianyu Huang[8], Erika Whiteford[9,10$]

[1]Centre for Environmental Geochemistry, School of Geography, University of Nottingham, University Park, Nottingham, NG7 2RD, UK
[2]School of Natural and Environmental Sciences, Newcastle University, Newcastle-upon-Tyne, NE1 7RU, UK
[3]School of Geography, Politics and Sociology, Newcastle University, Newcastle-upon-Tyne, NE1 7RU, UK
[4]National Environmental Isotope Facility, British Geological Survey, Keyworth, Nottingham, NG12 5GG, UK
[5]Centre for Environmental Geochemistry, School of Biosciences, University of Nottingham, Sutton Bonington Campus, Leicestershire, LE12 5RD, UK
[6]Environmental Change Research Centre, Department of Geography, University College London, London, WC1E 6BT, UK
[7]Geography Department, York St John University, YO31 7EX, UK
[8]State Key Laboratory of Biogeology and Environmental Geology and School of Geography and Information Engineering, China University of Geosciences, Wuhan 430078, China
[9]Department of Geography, Loughborough University, Loughborough, LE11 3TU, UK
[10]School of Science and Technology, Nottingham Trent University, Nottingham, NG11 8NS, UK
*Present address: School of Natural and Environmental Sciences, Newcastle University, Newcastle-upon-Tyne, NE1 7RU, UK
#Present address: Geography Department, York St John University, YO31 7EX, UK
$Present address: School of Science and Technology, Nottingham Trent University, Nottingham, NG11 8NS, UK

Correspondence to: Mark A. Stevenson (mark.stevenson@newcastle.ac.uk)

Abstract. The Arctic is rapidly changing, disrupting biogeochemical cycles and the processing, delivery and sedimentation of carbon (C), in linked terrestrial-aquatic systems. In this investigation, we coupled a hydrogeomorphic assessment of catchment soils, sediments and plants with a recent lake sediment sequence to understand the source and quality of organic carbon present in three Arctic upland lake catchments on Disko Island, located just south of the Low-High Arctic transition zone. This varied permafrost landscape has exposed soils with less vegetation cover at higher altitudes, and all lakes received varying extent of glacial meltwater inputs. We provide improved isotope and biomarker source identifications for palaeolimnological studies in high latitude regions, where terrestrial vegetation is at or close to its northerly and altitudinal range limit. The poorly developed catchment soils lead to lake waters with low dissolved organic carbon (DOC) concentrations ($\leq$1.5 mg L^{-1}). Sedimentary Carbon/Nitrogen (C/N) ratios, the C isotope composition of organic matter ($\delta^{13}C_{org}$) and biomarker ratios (*n*-alkanes, *n*-alkanols, *n*-alkanoic acids and sterols) showed that sedimentary organic matter (OM) in these lakes is mostly derived from aquatic sources (algae and macrophytes). We used a ^{210}Pb dated sediment core to determine how carbon cycling in a lake-catchment system (Disko 2) had changed over recent centuries. Recent warming since the end of the Little Ice Age (LIA ~1860 AD), which accelerated after ca. 1950, led to melt of glacier ice and permafrost releasing nutrients and DOC to the lake, stimulating pronounced aquatic algal production, as shown by a > 10 fold increase in β-carotene, indicative of a major regime shift. Our findings highlight that in Arctic lakes with sparsely developed catchment vegetation and soils, recent Anthropocene warming results in pronounced changes to in-lake C processing and the deposition of more reactive, predominately autochthonous C, compared with extensively vegetated low Arctic systems.

45   **Keywords.** carbon cycling; lipid biomarkers; carbon isotopes; hydrogeomorphology; wetlands; lakes; sediments; plants; permafrost; Greenland; Arctic

1 Introduction

The carbon (C) stored and cycled in permafrost, soils and lake sediments of Arctic landscapes is a vital component of the terrestrial C budget (Hugelius et al., 2014; Anderson et al., 2019), but Arctic ecosystems are changing
50   rapidly (Saros et al., 2019). Climate warming in Arctic regions (Smol and Douglas, 2007) can change the ecosystem structure of catchments and lakes with probable implications for landscape-scale C cycling (Anderson et al., 2018). In particular, changes in vegetation quantity and quality associated with long-term climate shifts can influence soil development (Wookey et al., 2009) and C transfer between terrestrial and aquatic environments. "Arctic greening", where warmer temperatures have enhanced terrestrial vegetation growth (Arndt et al., 2019)
55   or "Arctic browning" where changing climate regimes such as drought and winter warming can reduce vegetation productivity (Phoenix and Bjerke, 2016), have the potential to alter primary production in lakes (McGowan et al., 2018), with associated implications for C cycling. Warming also leads to glacial recession increasing meltwater discharge into Arctic watersheds (Slemmons et al., 2013). Arctic regions lying close to the High Arctic - Low Arctic transition zone (below 75° N latitude in western Greenland (Daniels and De Molenaar, 1993)) are likely to
60   experience particularly marked shifts in C cycling. Characterised by the presence (Low Arctic) or absence (High Arctic) of vegetation, warming may push marginal ecosystems into different ecological states (Corell et al., 2013).

Accurate methods for tracing how the quality and transfer of C is changing across catchment-lake systems are required for predicting broader scale future impacts on the Arctic C cycle (with global implications), for which geochemical analyses of lake-catchment samples are especially insightful (Karlsson et al., 2009;
65   Lapierre and del Giorgio, 2012; McGowan et al., 2018). Previous studies in Greenland have used $\delta^{13}C_{org}$ and C/N to track particulate catchment inputs (Anderson et al., 2018; Leng et al., 2012), but due to their narrow ranges with some specimens, using these techniques in isolation cannot always distinguish source identities (Lacey et al., 2018; Holtvoeth et al., 2016). Here we additionally use lipid biomarker chain length compositions and the C isotopic composition of fatty acid methyl esters (FAMEs) as a potential solution to this challenge (Castañeda and
70   Schouten, 2011). These techniques, which have been rarely applied in high latitude lake catchments, can provide additional understanding because shorter chains are typically indicative of algae and photosynthetic bacteria, mid-chains for macrophytes and longer chains of terrestrial plant input (Cranwell et al., 1987; Meyers, 2003; Ficken et al., 2000), while pigments (e.g. β-carotene) are effective autotroph biomarkers (McGowan, 2013). The type and quality of terrestrial OC supplied to lakes changes microbial communities and may ultimately regulate whether
75   lakes are CO_2 sources or sinks (Jansson et al., 2000; Kortelainen et al., 2013).

Palaeoenvironmental records from lakes integrate carbon from the lake and catchment and allow the reconstruction of variability of C cycling on timescales that would otherwise be unobtainable (Leavitt et al., 1989; Leavitt et al., 2009). Palaeolimnology of Arctic lakes can be used to help estimate C burial rates over the Holocene (Anderson et al., 2009; Anderson et al., 2019) and to understand how lakes in different regions process terrestrial
80   OM (McGowan et al., 2018). Investigations of carbon cycling across the transition between the cooler Little Ice Age (LIA) and warmer recent conditions in the Arctic provide a way of understanding how Arctic systems respond to climate change. Lacustrine records from close to the Jakobshavn Isfjord, near Ilulissat, east of Disko suggest that the lowest Holocene temperatures occurred here in the 19th century (LIA) (Axford et al., 2013; Briner et al.,

2016), rising in the early 20th century (Box et al., 2009; Yamanouchi, 2011) and accelerating with recent warming
85   (Hanna et al., 2012).

Qeqertarsuaq (Disko Island, West Greenland) is situated in a Low Arctic region, just south of the High
Arctic transition zone, and has a heterogeneous cover of herbaceous plants and bryophytes and several glaciers.
This paper investigates the organic geochemistry of OM in the catchments and lake sediments on Disko Island to
identify how terrestrial and aquatic $\delta^{13}C_{org}$ biomarkers (C/N ratios and $\delta^{13}C_{org}/\delta^{13}C_{FAMEs}$, chlorophyll and
90   carotenoid pigments) can be interpreted as palaeoenvironmental proxies in this sparsely vegetated Arctic region.
By investigating sites spanning different catchment sizes, vegetation cover, and proximity to glacial features we
aim to understand how these terrestrial factors influenced lake water quality and OC composition to ultimately be
archived in the sediment record. We use this information to guide the interpretation of a sediment sequence from
one lake spanning the transition from the LIA towards warmer conditions after 1950 to help understand how
95   pronounced climate warming can alter terrestrial-aquatic carbon cycling.

2 Materials and methods

2.1 Study area

Qeqertarsuaq (Disko Island) (Fig. 1) is located within Disko Bugt (Bay), a large marine embayment between
69º15'N - 70º20'N and 51º50'W - 55º00'W. Disko is the largest island to the west of Greenland (8,575 km²) and
100   is extensively glaciated with main Sermersuaq (Storbræ) and Bræpasset ice caps covering the central plateaus.
The geology is composed of 5,000 m deep and 60 M year old Tertiary basalts, part of the Disko-Nussuaq surge
cluster (Chalmers et al., 1999; Humlum, 1996). There are prominent U-shaped valleys, cirques, talus slopes and
fjords. Vegetation at a low altitude includes verdant dwarf shrub heaths in well-drained sites, and small mires in
wet or snowpack-fed localities (Bennike, 1995). *Salix glauca* is the dominant shrub and sparser fell-field
105   vegetation is present at higher elevations (Callaghan et al., 2011).

Disko Island has been subject to recent climate change, with a warming trajectory since the end of the
LIA and at an increasing rate since the 1990s (Box, 2002; Hansen et al., 2006). This has been confirmed locally
by tree ring evidence of winter warming (Hollesen et al., 2015) and both locally and regionally by instrumental
records (Hanna et al., 2012; Hansen et al., 2006). Mechanistically, warming has contributed to the destabilisation
110   of permafrost soils (Rowland et al., 2010) and retreat of catchment ice (including glaciers) (Anderson et al., 2017),
releasing nutrients into downstream lakes. Old C released from melting catchment permafrost (Pautler et al., 2010;
Schuur et al., 2009) could be one such source of C entering the lakes (as Disko is in a known permafrost active
layer monitoring zone (Humlum et al., 1999)), along with minor contributions of catchment bryophytes and plant
vegetation, evidenced by lipid ratios contributing to sedimentary C.

2.1 Hydro-geomorphological survey and catchment sampling

Three lakes (named Disko 1, 2 & 4), spanning a range of altitudes (214m to 575m) and catchment sizes (358 ha
to 1,829 ha) were visited in April and August 2013 (Fig. 1c). In April, catchment soils and plants were sampled
from exposed snow-free areas using a trowel into plastic bags. Further sampling was conducted in August 2013
to collect additional catchment soils, plants, benthic rock-scrape algae and lake edge water samples (lake edge,
120   ~3-5 m) for nutrient, DOC and pigment analysis.

2.1.1 Vegetation composition surveys

Field notes, aerial imagery (LANDSAT & QuickBird through Google Earth) and mapping (Disko Island, Qeqertarsuaq 1:100,000 Hiking Map) were combined to estimate the percentage cover of alpine vegetation, bare ground, snow cover and glaciers present in each lake catchment (including defining catchment boundaries) by
125   manually delineating the areas using the 2D area function in Google Earth Pro 7. In each lake catchment up to five 10x10 m plots were surveyed for vegetation cover, within which five 1x1 m quadrats were placed randomly, photographed and the percentage cover of plant functional types (defined as: total moss/lichen, total plants & total bare ground) were estimated, with key plant species identified following Rune (2011).

2.1.2 Water chemistry analysis

130   Water samples were filtered in the field using Whatman G/C filter papers, refrigerated and taken back to the laboratory for analysis. Filter papers were wrapped in foil, sealed in polythene and stored at −20ºC for chlorophyll and carotenoid (pigment) and trichromatic chlorophyll a analyses. TP (total phosphorus), SRP (soluble reactive phosphorus) and NH_4-N (ammonium) concentrations were measured by colorimetry using the methods in Mackereth et al. (1978). DOC was measured using a Shimadzu TOC-VCSN analyser, with DOC calculated as the
135   sum of organically bound C present in water that originate from compounds which can pass through a 0.45 μm filter. Total nitrogen (TN) was additionally analysed for the summer 2013 samples using a Ganimede TN analyser by alkaline digestion with peroxodisulphate.

2.2 Sediment core collection and chronology

In April 2013 sediments were sampled from Disko 2 and Disko 4 (Table 1) by drilling through lake ice using a
140   HON-Kajak corer. Disko 2 was selected for intensive study due to its well-dated sediment sequence. Sediment samples were extruded at 0.5 cm intervals and water samples were taken under the lake ice at a depth of 1 m. The core (Disko 2 K1; length of core 31.5 cm) was analysed for ^{210}Pb, ^{226}Ra, ^{137}Cs and ^{241}Am by direct gamma assay using an ORTEC HPGe GWL Series well-type coaxial low background germanium detector at the Environmental Change Research Centre, University College London. Total ^{210}Pb was measured by gamma emissions at 46.5keV,
145   with ^{226}Ra by the combined daughter isotope emissions of 295keV and 325keV to infer 'supported' ^{210}Pb. ^{137}Cs and ^{241}Am were measured at 662keV and 59.5keV (Appleby et al., 1986), with absolute efficiencies determined by comparison with standards. The constant rate of ^{210}Pb supply (CRS) model was used because slight departures from a monotonic declining trend in ^{210}Pb activity indicated variable sediment supply. A composite age-depth model was produced using the 1963 peak in ^{137}Cs and ^{241}Am from radioactive fallout at 3.25 cm, as the ^{210}Pb
150   placed this slightly earlier at 5 cm and so needed correcting (Appleby, 2001). Estimated ages in the extrapolated part of the core (below 10 cm) are assumed to have the same error (± 18 years) as final measured sample in 1845 AD.

2.3 Geochemical analyses

2.3.1 Chlorophyll and carotenoid pigment analysis

155   Chlorophyll a (chl a) was measured spectrophotometrically against an extraction solvent blank, after extracting Whatman G/C filter residues in acetone overnight (Jeffrey and Humphrey, 1975). Filtered residues from Whatman

GF/F (0.45 μm pore size) and freeze-dried sediments were extracted in an acetone, methanol, and water mixture (80:15:5), filtered using a 0.22 μm polytetrafluoroethylene (PTFE) filter, dried under N_2 gas and re-dissolved into injection solution, prior to analysis on an Agilent 1200 series high-performance liquid chromatography (HPLC) unit using a ODS Hypersill column (250x4.6mm; 5μm particle size) and a photo-diode array detector (350-750nm). Separation conditions are detailed in McGowan et al. (2012). Pigments were identified and quantified from the chromatograms by comparing retention times and spectral characteristics with calibration standards (DHI Denmark).

2.3.2 Bulk $\delta^{13}C$ and C/N analysis

$\delta^{13}C_{org}$ and C/N analyses were conducted on sediment core, soil and vegetation samples. Sediment core and soil samples were pre-treated in excess 5% HCl (hydrochloric acid), dried and homogenised with an agate pestle and mortar. Plants and bryophyte samples were placed in 5% HCl for 5 minutes to remove carbonates, checked for a visible reaction, washed in deionised H_2O, dried and ground to powder by freezer milling using liquid nitrogen. TOC, TN (from which we calculated C/N ratios) and $\delta^{13}C_{org}$ analyses were performed at the British Geological Survey online using a Costech ECS4010 elemental analyser (EA) coupled to a VG Triple Trap and a VG Optima dual-inlet mass spectrometer. $\delta^{13}C$ values were calibrated to the VPDB (Vienna Pee Dee Belemnite) scale using within-run laboratory standards (BROC2 and SOILB) calibrated against NBS-18, NBS-19 and NBS-22. Analytical precision of $\delta^{13}C_{org}$ was to within ±<0.1‰ (1 SD). C/N is presented as the weight ratio. To facilitate statistical analysis samples were grouped into aquatics, core sediments, terrestrial vascular plants and 'terrestrial other', with significant differences examined using one-way ANOVA. Post-hoc Tukey HSD was confirmed using Welch's t-test.

2.3.3 Lipid biomarker analysis

Lipid compounds were extracted from freeze dried and homogenised plant, benthic macroalgae, soil and sediment samples from Disko 2 using 15 mL HPLC grade 3:1 $CH_2Cl_2:CH_3OH$ and a MARS 5 microwave system (CEM Microwave Technology, UK). The total lipid extracts (TLEs) were dried under N_2, saponified using 6% KOH in CH_3OH, and the neutral compounds separated using hexane (after Pearson et al., 2011). Excess salts were removed from the neutral fraction using CH_2Cl_2 extracted ultrapure H_2O. The neutral fractions were derivatised using bis(trimethylsilyl)trifluoroacetamide (BSTFA) to form trimethylsilyl esters prior to analysis by gas chromatography mass spectrometry (GC-MS). Following the removal of the neutral compounds, the remaining extract was acidified with HCL and the acid fraction subsequently separated using hexane. Prior to analysis by GC-MS the isolated acid fraction was esterified using BF3/MeOH and the fatty acid methyl esters (FAMEs) extracted using C_6H_{14}. Neutral and FAME samples were analysed using an Agilent 7890A GC coupled to a 5975C MS. Samples were injected in pulsed splitless mode at 280 ºC in CH_2Cl_2 using a HP DB5-MS (30m x 0.25 mm i.d: 0.25μm film thickness) column and with a temperature program, compound identification and quantification following Pearson et al. (2007). Diagnostic lipid ratios and equations were selected based on those which were most informative in the catchment study to interpret changes in source inputs (Fig. 5) and down-core changes in OM source and reactivity (Fig. 7b).

2.3.4 Compound-specific isotope analysis

Compound-specific $\delta^{13}C$ was obtained by injecting FAME acid fractions spiked with squalene (temperature
195 290ºC) in splitless mode into a Thermo Finnigan Trace GC coupled to a Thermo Finnigan Delta Plus XP isotope
ratio mass spectrometer using a combustion interface (GC-C-IRMS) according to conditions in Huang et al.
(2018). Samples were run in at least duplicate, with reproducibility achieved to at least ±0.5 ‰ (SD). An *n*-alkane
standard was used as a within-run laboratory standard with known $\delta^{13}C$ values (Chiron, Norway) and daily
combined isotopic reference samples from C_{12} to C_{32} (Indiana University). $\delta^{13}C$ values were relative to the VPDB
200 (‰) and corrections were made for the additional C atoms introduced by BF_3-MeOH derivatisation (Boschker et
al., 1999).

3 Results

3.1 Hydro-geomorphology and vegetation surveys of lake catchments

The highest elevation cirque lake (Disko 2) had the least imagery-derived catchment vegetation and the smallest
205 catchment: lake area (CA:LA) ratio (Table 1, Fig. 2). The U-shaped valley bottom lake (Disko 1) with a
moderately-sized CA:LA ratio had the highest proportion of imagery-derived vegetation cover. The lowest
elevation valley-end lake (Disko 4) was located <2 km from to the coast and had moderate imagery-derived
vegetation cover and the highest CA:LA ratio. All lakes had permanent glacial ice present in the catchment, but
this coverage was greatest in the catchment of Disko 1 which is fed directly by the Lyngmarksbræen ice cap (Fig.
210 1c). Close to the lake margins, plant cover (assessed by vegetation transects) was highest at Disko 4 and lowest at
Disko 2 (Table 1). Detailed vegetation composition surveys are available in Table S1.

3.2 Water chemistry

In all lakes, soluble reactive phosphorus (SRP) concentrations were higher in April than in August, with the
highest levels in Disko 1 and lowest in Disko 2 (Table 2). Seasonal total phosphorus (TP) concentrations in Disko
215 1 were similar in April (67.9 µg L^{-1}) and August (67.8 µg L^{-1}), but declined in Disko 2 (from 37.5 µg L^{-1} in April
to 2.9 µg L^{-1} in August) and Disko 4 (164.8 µg L^{-1} in April to 2.9 µg L^{-1} in August). Dissolved inorganic nitrogen
(DIN) was present predominantly as nitrate (NO_3-N) which was highest in Disko 1 (0.05 µg L^{-1} in August) and
lowest in Disko 2 (0.01 µg L^{-1} in April) whereas NH_4^+-N concentrations were below detection limits, except in
August in Disko 1 and Disko 2 (11.54 µg L^{-1} and 3.15 µg L^{-1} and respectively). DOC was low in all lakes in both
220 April and August; the highest values were recorded in April for Disko 4 (1.5 mg L^{-1}). Biomarker pigments in the
water column of all lakes in August (ice-free conditions) were characterised by high concentrations of pheophytin
b (0.95 nmol L^{-1}), a chlorophyll *b* derivative from chlorophytes, euglenophytes and higher plants and lower
concentrations of diatoxanthin from siliceous algae. Diatoxanthin concentrations were much higher in Disko 1
(0.63 nmol L^{-1}) than in the other lakes (Disko 2 – 0.05 nmol L^{-1}; Disko 4 0.08 nmol L^{-1}).

225 ### 3.3 TOC, C/N ratios and $\delta^{13}C$ values (bulk and compound specific) of modern samples

TOC content varied between a maximum of 66.3% in herbaceous plants to 0.1% in soil and 0.5% in lake surface
sediment (Fig. 3a). C/N ratios varied most in lichens (from 25 to nearly 200) and terrestrial plants (~15 to ~130)
and were lowest in algae (9-17), aquatic macrophytes (9-36) and lake surface sediment samples (7.6-8.6) (Fig.

3b). $\delta^{13}C_{org}$ ranged between −6.4‰ and −33.6 ‰ (Fig. 3c). Comparing the C/N ratio and $\delta^{13}C_{org}$ values, most
230   samples plotted across the ranges typical of C_3 land plants and lacustrine algae (Leng et al., 2012; Meyers and
Teranes, 2001; Anderson et al., 2018), with one outlying aquatic macrophyte sample (Disko 2 *Potamogeton sp.*
−6.4 ‰) and some lichen, moss and soil samples that had higher $\delta^{13}C_{org}$ values (~ −20 to −23 ‰) (Fig. 4). Benthic
macroalgae samples had $\delta^{13}C_{org}$ values around −25 ‰. Lake sediment core samples from Disko 2 & 4 had low
C/N ratios (7.5 to 18.4) and $\delta^{13}C_{org}$ values ranged from −28 to −20 ‰. Statistical analyses demonstrated that C/N
235   ratios of terrestrial vascular plants were significantly different from aquatics and lake sediments, and terrestrial
'other' also differed from lake sediments (Table 3). There was more overlap in $\delta^{13}C_{org}$ and so terrestrial vascular
plant measurements were only significantly different to core sediments and terrestrial 'other' (Table 3; Fig. 4).
Compound-specific $\delta^{13}C_{FAMEs}$ values tended to become progressively lower (more negative) for higher chain
length fatty acids than shorter chains, compared with bulk $\delta^{13}C_{org}$ (SI Fig. 1).

240   **3.4 Lipid distributions and ratios from the Disko 2 catchment**

Lipid histograms of *n*-alkane, *n*-alkanol and *n*-alkanoic acid distributions from Disko 2 (Fig. 5; SI 3) showed that
n-alkane samples displayed an odd over even predominance whereas *n*-alkanols and *n*-alkanoic acids were even-
dominated, as expected for naturally-derived samples (Bianchi and Canuel, 2011). Proportionately (given
limitations of single ion filtering) *n*-alkanoic acids were the most abundant (mean 56.7%; range 11.7-98.8%)
245   followed by *n*-alkanols (mean 31.5%; range 0.8-83.5%) and *n*-alkanes (mean 11.8%; range 0.5-59.0%).
Differentiation between catchment samples was possible, for example among *n*-alkanes the algal benthic rock
scrape sample was dominated by *n*-C_{23}, while for *Potamogeton* sp. and green moss, *n*-C_{31} was dominant.
Herbaceous plants like *Harrimanella hypnoides* tended to be dominated by *n*-C_{29} or *n*-C_{31} while dominant chain
lengths were slightly shorter in *Salix arctica* (*n*-C_{27}).
250   Soil sample *n*-alkane distributions were either clearly terrestrially influenced (e.g. catchment summit
soil, dominant in *n*-C_{31}) or more mixed (e.g. Plot C soil sample, taken close to the Disko 2 shoreline) and surface
sediments also reflected mixed inputs with a slight odd predominance and an extended range. *n*-Alkanols could
also be used to differentiate between samples, for example, dominance of *n*-C_{26} in the algal benthic rock scrape
but *n*-C_{22} in *Potamogeton* sp. Higher plants were typically mid-chain dominant in *n*-alkanols (e.g. *Harrimanella*
255   *hypnoides* with a maximum at either *n*-C_{26} or *n*-C_{22} and *Salix arctica* *n*-C_{24}), and the bimodal distribution of lake
surface sediments suggesting multiple inputs. Although the algal benthic rock scrape and one *H. hypnoides* sample
had similar *n*-C_{26} alkanol dominance, *H. hypnoides* also has higher chain length compounds which the algal
benthic rock scrape does not and the degree of dominance in the algal rock scrape (*n*-C_{26} ca. 20%) is different
compared with *H. hypnoides* (*n*-C_{26} ca. 9%), making these compositions distinguishable. The *n*-alkanoic acid *n*-
260   C_{16} was ubiquitous and abundant in all samples, but distributions still provided some differentiation, for example
a secondary abundance in green moss around *n*-C_{24}, *n*-C_{22} in the unidentified herbaceous plant sample and *n*-C_{24}
in the *Salix arctica* (leaf) and *n*-C_{18} in the plot C soil sample.
Lipid ratios for catchment and surface sediment samples are provided in Table 4 with interpretation of
source attribution described in Table S2. In this system the *n*-alkane ratios CPI 2 appears to be a good indicator
265   of terrestrial plants, TAR_{HC} is an excellent indicator of leafy vascular plants, P_{WAX} indicates multiple terrestrial
inputs and *n*-C_{27} is a good indicator of key plant species like *Salix arctica* and *Chamerium latifolium*. Among *n*-
alkanoic acids CPI_T is a good indicator of aquatic macrophytes and mosses and *n*-C_{30} of non-woody herbaceous

terrestrial inputs. n-Alkanol indicator ratios are n-C_{16} for aquatic macrophytes and n-C_{24} for woody terrestrial plants. The ratio of brassicasterol/ total sterols is a clear indicator of algae and moss.

3.5 Down core changes in bulk and compound-specific isotopic and geochemical parameters

270

Stratigraphic zones determined using optimal sum of squares partitioning were from 2010 – 1833 AD (Zone A), 1833 – 1530 AD (Zone B) and 1530 – 1334 AD (Zone C). In zone C (1300-1530 (±18) AD) TOC generally declined from ~3 to 1.4 %, except for a peak to ~5.7 % around ~1440 (±18) AD and $\delta^{13}C_{org}$ ranged between −20.3 ‰ and −24.1 ‰ (Fig. 7a). C/N ratio ranged between 9 and 12 (reaching higher value ~1510 (±18) AD)), while β-carotene ranged between 1.1 and 33.3 nmoles pigment g^{-1} TOC with a peak around ~1440 (±18) AD (Fig. 7a). Zone B (1530 – 1833 (±18) AD) proxies were relatively stable, except for an increase to ~5% TOC ~1640 (±18) AD, where $\delta^{13}C_{org}$ was high (~ −20.2 ‰) and β-carotene reached ~24.1 nmoles pigment g^{-1} TOC (Fig. 7a). In zone B around ~1643 (±18) AD there were peaks in CPI 2 and decreases in P_{WAX} and the C_{26} n-alkanes, with increases in TAR_{HC} around 1762 (±18) AD and ~1653/1798 (±18) AD for the C_{30} n-alkanoic acids (Fig.7b).

280

The most pronounced changes in all geochemical proxies was in zone A (1833 (±18) – 2013 (±2) AD) which includes the ~1850 (± 18) AD end of the LIA cooling period (Fig.7). TOC increased from <2.3 to 7.4 % by the top of the core, while $\delta^{13}C_{org}$ decreased from ~ −23.5 to −28 ‰ by ~1940 AD where it remained stable to ~2003 (±2) AD (Fig. 7a). $\delta^{13}C_{28:0}$ FAME also decreased from −26.3 ‰ in ~ 1895 AD to −35.6 ‰ by ~2003 (±2) AD (Fig. 7a). In contrast, C/N increased fluctuations ~1923 (±6) AD (12.4) followed by a steep decline to ~7.5 by ~1987 (±3) AD, while β-carotene increased 70-fold (from 0.9 nmoles pigment g^{-1} TOC in 1906 (±7) AD to 70.4 nmoles pigment g^{-1} by 2003 (±2) AD) (Fig. 7a). In zone A P_{WAX}, CPI_T, C_{16} n-alkanol and the ratio of brassicasterol/total sterols increased to the top of the core, whilst the C_{27} n-alkane, C_{30} n-alkanoic acid and C_{24} n-alkanols declined, and CPI 2 and TAR_{HC} were both generally stable until the uppermost sample (Fig. 7b).

4 Discussion

290

4.1 How is catchment and aquatic carbon structured on Disko?

The soil TOC content of the catchments was highly variable (Fig. 3a). Local decomposition mediated by temperature and moisture can regulate Arctic soil loss or retention, and litter quality (Wookey et al., 2009). In Arctic regions generally, patchy organic soils indicate faster recycling rates or specific local sources of OM controlled by the topographically varied dynamic glacio-fluvial geomorphic system (Conant et al., 2011; Castellano et al., 2015; Anderson et al., 2017). Vegetation changed with elevation, with the highest lake (Disko 2) catchment having more 'bare ground' and rock/gravel substrates (Table 1). The terrestrial plant species we observed are common on Disko (e.g. Callaghan et al., 2011; Daugbjerg, 2003) and other parts of West Greenland (Fredskild, 2000), but the plant communities were less species-rich than in other lowland areas of Disko (Table 1, Table SI 1) (Callaghan et al., 2011). The woody species *Betula nana* and *Juniperus communis* were uncommon, and only present in the Disko 4 catchment (Table S1) probably because it is more sheltered with better drained soils than the other sites, and located closer to the high diversity of the coast (Callaghan et al., 2011; Hollesen et al., 2015).

Differences in vegetation cover, soil development and glacial meltwaters appeared to influence water chemistry and aquatic production. DOC concentrations were low in all lakes (≤1.5 mg L^{-1}, Table 2), reflecting

limited soil maturity and development in all three catchments, as observed in high Arctic lakes with similar topographies (Michelutti et al., 2002). The DOC concentrations are much lower than other low Arctic sites in Alaska or Norway which have more vegetated catchments (McGowan et al., 2018). Sparser vegetation cover of the Disko 2 (13%) catchment compared with Disko 1 (39%) probably reduced organic matter leachates from soils or permafrost, reducing microbial-induced phosphorus leaching (Buckeridge et al., 2015). Hence, TP and SRP concentrations were lower in Disko 2 than 4 (Table 2) during the spring thaw period. The extensive wetlands in the Disko 1 and 4 catchments are typical of Arctic environments where waterlogging leads to terrestrial OM transfer, leading to slightly higher C/N ratios in Disko 4 compared with Disko 2 which is located in a cirque where bare earth is predominant (Hobbie et al., 2000) (Table 1; Fig. 1, 4).

Permanent ice/glaciers accounted for between 3.6 and 14% of catchment cover (Table 1), and glacial inputs are known to influence waterbodies via effects of seasonal meltwater release on lake water residence time and nutrient release from glacial melt (Anderson et al., 2017; Slemmons et al., 2013). Of the three lakes, Disko 1 stands out with the highest glacier cover as part of the Lyngmarksbræen ice cap is present in the catchment and so the water chemistry is different to Disko 2 and 4. High summer nutrient conditions in Disko 1 (Table 2: SRP 13.8 µg L^{-1}, TP 67.9 µg L^{-1} and NH$_4^+$ 11.5 µg L^{-1}) are probably linked to the catchment glacier source, which can influence algal production and community composition (Slemmons and Saros, 2012). Glacier outwash into Disko 1 appears to profoundly affect phytoplankton composition, with the degradation products (pheophytin b, a chlorophyll b derivative) being most abundant in Disko 1, whereas chlorophyll a was dominant at other sites (Table 2). Chlorophytes also grow on glaciers (Williamson et al., 2018) and degrade into pheophytin b by UVR damage to algae (Leavitt et al., 2003), which can be released to downstream waterbodies via ice melt. High diatoxanthin from siliceous algae in Disko 1 can be explained by photo-degradation due to low DOC (high water clarity) and shallow depth (<5.9 m), without refuge from UVR (Sommaruga, 2001), while high chlorophyll a in Disko 4 can be explained by nutrient inputs from a large catchment with extensive plant cover (Table 1). Overall, catchment characteristics such as glaciers, soil type and plant cover appear to be tightly coupled to water quality, giving the potential for allochthonous inputs to influence autochthonous production.

4.2 Can catchment and in-lake composition of modern bulk OM discriminate between C sources and sinks?

Despite the potential for terrestrial-aquatic linkages the bulk geochemical composition of catchment vegetation and soils was very different to the surface and down-core lake sediments (Fig. 3 & 4), suggesting that lake OM was either predominantly derived from in-lake/ autochthonous rather than catchment sources, or was extensively chemically altered prior to deposition in the lakes. (Table 3). The low C/N ratios of surface sediments (range 7.6 to 8.6) also suggest an algal (low cellulose, aquatic) source rather than terrestrial plants with much higher values: lichen (30.0 – 196.0), herbaceous plants (12.2 – 133.6) and moss (35.2 – 127.9) (Fig. 3b) (Meyers and Teranes, 2001). C/N variation in lichens is likely as some can be N$_2$ fixing. Degradation of terrestrial vegetation by microbial and physical processes can result in substantial DOC loss from catchments (Moody et al., 2013). However, the low OM abundance in these catchments likely leads to low DOC fluxes, as evidenced by low lake water DOC (all ≤1.5 mg L^{-1}). The OM sedimented in lakes is a combination of autochthonous and allochthonous sourced C often with varying susceptibility to degradation (Catalán et al., 2013). The low C/N values are similar to values measured in lakes in the Low Arctic area of Sisimiut in south-west Greenland (Leng et al., 2012; Anderson et al., 2018).

Compared with C/N, for $\delta^{13}C_{org}$ there was more overlap in values between surface sediment and
345 terrestrial samples, making differentiation more challenging (Fig. 3c) probably due to the competing effects of
productivity and source in influencing C isotope composition (Hodell and Schelske, 1998; Meyers and Teranes,
2001). Despite the overlap we found the $\delta^{13}C_{org}$ of terrestrial vascular plants was statistically distinct from core
sediments, but also from samples in the terrestrial 'other' category (bryophytes and lichens) (Table 3), suggesting
that autochthonous algae, and not terrestrial vegetation are the main source of sedimentary C (Meyers and Teranes,
350 2001). In contrast to higher plants, bryophytes lack stomata and their isotope fractionation is closer to atmospheric
CO_2 (Smith and Griffiths, 1996) (Hanson et al., 2014; Glime, 2007). On the other hand, variability in $\delta^{13}C_{org}$ of
aquatic algae and macrophyte samples (−6.4 ‰ to −30.7 ‰) is expected to be high because aquatic plants can
access C (and DIC) from different lake habitats which are in variable equilibrium with atmospheric CO_2. Higher
values of $\delta^{13}C_{org}$ in these lake sediments indicate either greater contributions of algae, phytobenthos and
355 macrophytes, or increases in productivity. The few low C/N ratios (approaching ~12 in some terrestrial specimens
e.g. *Salix arctica, Chamerion latifolium*) are probably linked to new plant growth (with less lignin) or differences
in sample response to inorganic C cleaning during acidification (Brodie et al., 2011). Low values in soils may be
caused by soil algae which are common in Arctic areas (Pushkareva et al., 2016) and loss of carbon by diagenesis.

4.3 Does the molecular composition of modern samples help distinguish C sources and sinks?

360 Several molecular ratio indicators were particularly useful markers of terrestrial samples (see Table S3 for
interpretation summary). For *n*-alkanes, higher CPI 2 values were a good indicator of leafy catchment plants (e.g.
Harrimanella hypnoides, Chamerion latifolium) (Table 4) (Marzi et al., 1993), although they made a small
contribution to the total organic carbon pool. The low *n*-alkane CPI 2 values in soil and surface sediment reflect
both the possibility of degradation of any minor terrestrial sources and in sediments, and the predominant aquatic
365 source in the lake sediments evidenced by C/N ratios (Meyers and Ishiwatari, 1993) (Fig. 4). Similarly, the *n*-
alkane TAR_{HC} ratio, commonly used as an indicator of terrestrial (higher) to aquatic algae (lower) inputs
(Bourbonniere and Meyers, 1996), was a robust indicator of the ericaceous shrub *Harrimanella hypnoides* and
other herbaceous plants. The TAR_{HC} ratio had moderate values in aquatic macrophytes such as *Potamogeton* sp.,
but only minor values in *Salix arctica*. Highest *n*-alkane P_{WAX} index values (Zheng et al., 2007) were found in
370 vascular plants such as *Harrimanella hypnoides*, catchment soil samples, the aquatic macrophyte *Potamogeton*
sp. and green moss. Although the P_{WAX} ratio in this system cannot distinguish among specific terrestrial inputs,
the low contributions from the algal benthic rock scrape sample, make the index a good indicator of terrestrial
versus aquatic inputs. C_{30} *n*-alkanoic acid contributions were highest in the *Harrimanella hypnoides* (2) sample
and the Plot C soil sample, and so this ratio appears to be a good indicator of non-woody vascular plant and soil
375 terrestrial inputs. The C_{24} *n*-alkanol ratio was a good indicator of lignin rich higher plants like *Salix arctica* and
Chamerion latifolium.

Other molecular ratio indicators were useful markers of aquatic algae, macrophytes, bryophytes and soils
samples (see Table S3 for interpretation summary). The CPI_T index for *n*-alkanoic acids was a good indicator of
how degraded (or petrogenic) the C is (Matsuda and Koyama, 1977), with highest contributions in *Potamogeton*
380 sp., black moss and the algal benthic rock scrape indicating high proportions of non-degraded C (Table 4). The
C_{16} *n*-alkanol was most abundant in surface sediment, the aquatic macrophyte *Potamogeton* sp. and Plot C soil,
but can also be interpreted as a bulk productivity indicator (Cranwell, 1981). Brassicasterol is known to be present

in samples containing cyanobacteria, algal mats, diatoms and Brassicaceae (Pearson et al., 2007; Volkman, 2003; Volkman et al., 1998; Rampen et al., 2010), but is commonly used in lakes as a diatom biomarker due to its high abundance (Nelson and Sachs, 2014). The markedly higher brassicasterol contribution in the Disko 2 surface sediments points to a pelagic algal source (mainly diatoms) of this compound, with the next highest contributions in the algal benthic rock scrape sample, moss samples and the Plot C soil sample. The C_{27} ratio to total saturated n-alkanes was also a good indicator of some plants such as the *Salix arctica* leaf fragment and *Chamerion latifolium*. Combined, these markers not only distinguish terrestrial from aquatic, but also infer more subtle changes in the type of terrestrial cover.

4.4 Evidence for changes in C cycling through the LIA and recent warming periods

The dominant feature of the lake sediments across all catchments is the low C/N ratios (Fig. 4), suggesting that most sedimentary OM in these lakes is autochthonous in origin, corroborated by high brassicasterol (algal indicator), C_{16} n-alkanols (macrophyte indicator) and n-alkanoic acid CPI_T ratio (indicator of both young, fresh OM and shorter-chain algal source) in the uppermost sediments (Table 4). This is mirrored by declines after ~1750 AD in both long chain n-C_{27} and n-C_{30} total n-alkanoic acids ratios (Fig.7b) which highlight that proportionally shorter chain lengths indicative of aquatic algae increased (Cranwell et al., 1987). Dated sediments from Disko 2 therefore allow shifts in predominantly autochthonous C sources to be tracked through the LIA cooling period (LIA ~1300 AD to ~1850 AD; Miller et al., 2012; Matthews and Briffa, 2005) and subsequent warmer period after 1950 to understand how catchment mediated climate changes affect C cycling in this snowpack-fed cirque lake.

Aside from brief increases in % TOC and β-carotene around ~1450 AD and ~1650 AD, there was a major shift from ~1860 AD, to the present day with a >10-fold increase in β-carotene indicating a major increase in phototrophic production. One scenario is that the catchment thawed and the permanent ice cover (snowpack, cirque glacier) reduced, resulting in nutrients being released into the lake stimulating algal growth (Slemmons and Saros, 2012; Slemmons et al., 2013). Increases in the length of the ice-free period have been correlated with diatom responses in High Arctic locations (Michelutti et al., 2020; Griffiths et al., 2017). Although changes in the ice-free period have been associated with biological and chemical changes in Low Arctic lakes (Saros et al., 2019; Hobbs et al., 2010), it is rare to see the major directional shifts in Low Arctic biota in response to warming that are more commonly observed in High Arctic sites.

The high elevation of Disko 2 (575 m.a.s.l) and its limited catchment vegetation appear to suggest that the lake response to warming is simple, directional and fundamentally changed carbon cycling in the lake. This contrasts with many Low Arctic systems, where catchment vegetation is more established (Anderson et al., 2019), leading to more complex whole-lake responses to climate. The probable loss of a cirque glacier in the catchment after the LIA would have changed meltwater regimes, reducing lake water dilution and turbidity, making the lake more suited to algal production after ~1900 AD. Decreases in C/N ratio and increases in β-carotene after ~1900 AD (Meyers and Teranes, 2001) indicate increased algal production, similar to post-LIA increases in pigments in Baffin Island lake sediments (Florian, 2016; Florian et al., 2015; Michelutti et al., 2005). The decrease in $\delta^{13}C_{org}$ in the most recent sediments in Disko 2 is opposite to the expectation that higher productivity from nutrient enrichment leads to higher $\delta^{13}C_{org}$ (Hodell and Schelske, 1998). This decline of ~1.4 ‰ over the last ~50 years may be partly accounted for by the Suess effect from fossil fuel contamination (Verburg, 2007; Tans et al., 1979).

However, most of the reduction in $\delta^{13}C_{org}$ probably results from a source change such as increased algae and macrophyte abundance. Combined, these geochemical markers show that following the end of the LIA cooling, algal production increased markedly in Disko 2, with this shift becoming more pronounced with the onset of
425   recent warming and regional Disko glacier retreat (since ~1950 AD) (Yde and Knudsen, 2007; Hollesen et al., 2015; Box, 2002).

      Although terrestrial OM was unlikely to be the dominant source of OM to Disko 2, there are indications that the type and magnitude of terrestrial OM fluxes have subtly varied over time. For example, increases in the C/N ratio around ~1900 AD suggest catchment dieback, facilitated by warming during melt season exposure from
430   reduced ice cover (Leng et al., 2012). The composition of catchment-derived C also shifted after ~1895 AD with a decline in $\delta^{13}C_{28:0}$ FAME from terrestrial plants, indicating a shift in terrestrial species with lower $\delta^{13}C_{28:0}$ values as the catchment thawed and nutrient availability increased. $\delta^{13}C_{FAME}$ values of Disko 2 catchment samples were lower (isotopically-depleted) in higher plants (e.g. *Harrimanella hypnoides* & *Chamerion latifolium*) (Table S2). Increases in *n*-alkane CPI 2 index between 1700 and 1600 AD probably reflect an earlier period of increased
435   herbaceous plant input as a result of erosive freeze-thaw action in the catchment (Fig.7b). Despite some degradation in the OM, the CPI 2 value is consistently >2 down core which suggests *n*-alkane diagenesis is not a key process, with relatively fresh material potentially sourced from release of preserved OM from permafrost thaw in the catchment from climate warming (Schuur et al., 2009; Pautler et al., 2010). The peak in *n*-alkane TAR$_{HC}$ (index strongly biased towards the ericaceous shrub *Harrimanella hyponoides* and other herbaceous
440   plants) in the 1760s is probably linked to catchment input of terrestrial material brought in during the LIA, which is probably partly old organic C, fluvially released from soils (Vonk et al., 2010). The steep topography surrounding Disko 2 is suited to the delivery of old OM, with visible seasonal thaw slope failure (Fig 2.a) confirmed by occasional gravel layers in other cores in this lake (Stevenson, 2017) which are a probable C delivery source (Lamoureux and Lafrenière, 2014). However, the *n*-alkane P$_{WAX}$, an indicator of waxy hydrocarbons which
445   are particularly abundant in terrestrial plants, was highest in the most recent ~70 years implying recent delivery of allochthonous material. Although the TAR$_{HC}$ and P$_{WAX}$ records disagree, they both suggests that the type of terrestrial material brought into the lake in the most recent period has changed and could be related to the sensitivity of TAR$_{HC}$ to *Harrimanella hypnoides* and other herbaceous plants, or their expansion (Table 4). The ratio of *n*-C$_{24}$ to total *n*-alkanols declined only in the uppermost samples (post~2003 AD), perhaps influenced by
450   the most recent changes in catchment vegetation on Disko (Hollesen et al., 2015), or dilution by recent abundant short chain *n*-alkanols. Increases in P$_{WAX}$ in the most recent sediments (Fig. 7b) highlights (through comparison with modern samples, Fig. 4, Table S3) that there are some recent increases in terrestrial contributions (but of a different source to that explained by TAR$_{HC}$) contemporaneous with the recent increases in algal material (Fig. 7).

455   **4.5 Implications for C cycling in a coupled lake-catchment system**

The limited terrestrial vegetation and poorly developed soils on Disko Island, result in minor allochthonous inputs of C into lakes relative to autochthonous sources, especially in high elevation lakes. In systems where DOC fluxes from catchment soils are limited, this means in-lake autotrophic production is closely coupled and responsive to nutrient release from the catchment. Based on C/N and $\delta^{13}C_{org}$ the net carbon balance in these lakes on Disko
460   Island is more autochthonous than other low Arctic lakes in coastal south-west Greenland where catchment

vegetation is more developed (Anderson et al., 2018). Instead, Disko lakes seem to be similar in character to those on Baffin Island (Florian, 2016; Florian et al., 2015; Michelutti et al., 2002; Michelutti et al., 2005) owing to similarities in latitude (and hence seasonal variability), proximity to oceanic influence and geomorphology. The relatively minor DOC concentrations in lakes on Disko is also in marked contrast to more vegetated Arctic lake-

465   catchment systems in Alaska and Sweden (McGowan et al., 2018). Despite limited terrestrial-aquatic C fluxes in Disko lakes, there is probably a substantial minerogenic input related to the steep topography and proximity to active glacial and periglacial processes, probably influencing sedimentation rate to a greater extent than in lakes in a less rugged terrain.

       In-lake changes in C cycling also appear to be mediated by close coupling of these lakes to permanent

470   ice in the catchments through nutrients, DOC and outwash from glacial and periglacial processes, and potentially also length of the ice-free period. From a lake metabolic balance (or C flux) perspective, autotrophic production increases with temperatures, and may be associated with the length of the ice-free period (Michelutti et al., 2020; Griffiths et al., 2017; Saros et al., 2019). Increases in temperature also induced modest changes in catchment C fluxes. Together, these changes probably contribute to a modest but regionally important enhanced C store in

475   similar lakes across the region. Our results are consistent with changes to the Arctic C cycle more broadly across the Northern Hemisphere (McGuire et al., 2009; Post et al., 2009). This confirms that even in poorly vegetated catchments, changes in the character of plant cover and, more recently, increases in algal production, have occurred coincident with climate-mediated catchment change, likely coupled to glacier and permafrost/soil nutrient and C cycling.

480   **5 Conclusions**

Our study demonstrates that Arctic lakes in catchments with limited vegetation and at the transition between the Low-High Arctic boundary can respond rapidly to recent warming throughout the Anthropocene. This results in pronounced increases in phototrophic production, indicative of a major shift in autochthonous OC cycling. Specifically, we found:

485   i.   C in lake sediments in sparsely vegetated catchments mostly derives from autochthonous algal sources with minor terrestrial catchment carbon inputs;

     ii.   low lake water DOC concentrations suggest limited terrestrial-aquatic C transfer in comparison to other Low Arctic areas (e.g Sweden, Alaska) with denser vegetation cover;

     iii.   ice and glacier retreat appeared to regulate lake nutrient inputs and subsequently phototrophic

490   productivity, especially since the end of the LIA;

     iv.   increases in the length of the ice-free period during recent warming may also help to explain a recent regime shift evidenced by marked increases in autochthonous algae.

       Principally our findings illustrate how lakes with limited allochthonous C input respond more directly to

495   climate-related changes in comparison to the catchments that are typically in more extensively vegetated low Arctic systems. This is important because future anticipated warming will likely cause extensive, widespread major regime shifts in similar high-moderate elevation transitional Low-High Arctic catchment-lake systems where catchment buffering in poorly vegetated catchments is limited.

Data availability

500   Datasets have been submitted to PANGAEA (PDI-25602).

Author contribution

MAS wrote the initial manuscript and carried out geochemical analyses. SMcG and GEA supervised nutrient,
pigment and isotope preparation at U of N. EJP supervised lipid analyses at NU, MJL isotope analyses at BG, VJJ
age-depth estimations at UCL and XH CSIA at CUG. JJB assisted with statistical analyses and summer 2013
505   fieldwork. MAS, EJP and EW carried out spring 2013 sediment coring. All authors revised and approved the
manuscript.

Competing interests

The authors declare that they have no conflict of interest.

Acknowledgements

510   Mark Stevenson gratefully acknowledges the receipt of a NERC/ESRC studentship (ES/J500100/1).   We
acknowledge grants IP-1393-1113 & IP-1516-1114 from the NERC Isotope Geosciences laboratory (NIGL) for
the analysis of $\delta^{13}C_{org}$ & C/N ratios on sediment, soil and plant samples. Lipid and water chemistry analyses were
funded by the Freshwater Biological Association's 2015 Gilson Le Cren Memorial Award to Mark Stevenson.
We thank Teresa Needham, Christopher Kendrick, Julie Swales, Ian Conway, Graham Morris, Bernard Bowler,
515   Paul Donohoe, Qingwei Song and Jiantao Xue for technical support. We acknowledge the support of Handong
Yang for radiometric dating and the drafting of Fig 6a. Financial support for fieldwork was awarded via the
INTERACT transnational access scheme (grant agreement No 262693) under the European Community's Seventh
Framework Programme and UK RI NERC grant NE/K000276/1. Logistical support is acknowledged from
University of Copenhagen Arktisk Station including Ole Stecher, Kjeld Mølgaard and Erik.

References

520

Anderson, N. J., D'Andrea, W., and Fritz, S. C.: Holocene carbon burial by lakes in SW Greenland, Global Change
Biol., 15, 2590-2598, 10.1111/j.1365-2486.2009.01942.x, 2009.

Anderson, N. J., Saros, J. E., Bullard, J. E., Cahoon, S. M. P., McGowan, S., Bagshaw, E. A., Barry, C. D., Bindler,
R., Burpee, B. T., Carrivick, J. L., Fowler, R. A., Fox, A. D., Fritz, S. C., Giles, M. E., Hamerlik, L., Ingeman-
525   Nielsen, T., Law, A. C., Mernild, S. H., Northington, R. M., Osburn, C. L., Pla-Rabès, S., Post, E., Telling, J.,
Stroud, D. A., Whiteford, E. J., Yallop, M. L., and Yde, J. C.: The Arctic in the Twenty-First Century: Changing
Biogeochemical Linkages across a Paraglacial Landscape of Greenland, Bioscience, 67, 118-133,
10.1093/biosci/biw158, 2017.

Anderson, N. J., Leng, M. J., Osburn, C. L., Fritz, S. C., Law, A. C., and McGowan, S.: A landscape perspective
530   of Holocene organic carbon cycling in coastal SW Greenland lake-catchments, Quaternary Science Reviews, 202,
98-108, 10.1016/j.quascirev.2018.09.006, 2018.

Anderson, N. J., Appleby, P. G., Bindler, R., Renberg, I., Conley, D. J., Fritz, S. C., Jones, V. J., Whiteford, E. J., and Yang, H.: Landscape-Scale Variability of Organic Carbon Burial by SW Greenland Lakes, Ecosystems, 22, 1706-1720, 10.1007/s10021-019-00368-8, 2019.

535 Arndt, K. A., Santos, M. J., Ustin, S., Davidson, S. J., Stow, D., Oechel, W. C., Tran, T. T. P., Graybill, B., and Zona, D.: Arctic greening associated with lengthening growing seasons in Northern Alaska, Environmental Research Letters, 14, 125018, 10.1088/1748-9326/ab5e26, 2019.

Axford, Y., Losee, S., Briner, J. P., Francis, D. R., Langdon, P. G., and Walker, I. R.: Holocene temperature history at the western Greenland Ice Sheet margin reconstructed from lake sediments, Quaternary Science

540 Reviews, 59, 87-100, 10.1016/j.quascirev.2012.10.024, 2013.

Bennike, O.: Palaeoecology of two lake basins from Disko, West Greenland, JQS, 10, 149-155, 10.1002/jqs.3390100205, 1995.

Bianchi, T. S., and Canuel, E. A.: Chemical biomarkers in aquatic ecosystems, Princeton University Press, 2011.

Boschker, H. T. S., de Brouwer, J. F. C., and Cappenberg, T. E.: The contribution of macrophyte-derived organic

545 matter to microbial biomass in salt-marsh sediments: Stable carbon isotope analysis of microbial biomarkers, Limnology and Oceanography, 44, 309-319, 10.4319/lo.1999.44.2.0309, 1999.

Bourbonniere, R. A., and Meyers, P. A.: Anthropogenic influences on hydrocarbon contents of sediments deposited in eastern Lake Ontario since 1800, Environ. Geol., 28, 22-28, 10.1007/s002540050074, 1996.

Box, J. E.: Survey of Greenland instrumental temperature records: 1873–2001, IJCli, 22, 1829-1847,

550 10.1002/joc.852, 2002.

Box, J. E., Yang, L., Bromwich, D. H., and Bai, L.-S.: Greenland Ice Sheet Surface Air Temperature Variability: 1840–2007*, J. Clim., 22, 4029-4049, 10.1175/2009jcli2816.1, 2009.

Briner, J. P., McKay, N. P., Axford, Y., Bennike, O., Bradley, R. S., de Vernal, A., Fisher, D., Francus, P., Fréchette, B., Gajewski, K., Jennings, A., Kaufman, D. S., Miller, G., Rouston, C., and Wagner, B.: Holocene

555 climate change in Arctic Canada and Greenland, Quaternary Science Reviews, 147, 340-364, 10.1016/j.quascirev.2016.02.010, 2016.

Brodie, C. R., Casford, J. S., Lloyd, J. M., Leng, M. J., Heaton, T. H., Kendrick, C. P., and Yongqiang, Z.: Evidence for bias in C/N, δ^{13}C and δ^{15}N values of bulk organic matter, and on environmental interpretation, from acid treatment methods, Quaternary Science Reviews, 30, 3076–3087, 2011.

560 Buckeridge, K. M., Schaeffer, S. M., and Schimel, J. P.: Vegetation Leachate During Arctic Thaw Enhances Soil Microbial Phosphorus, Ecosystems, 19, 477-489, 10.1007/s10021-015-9947-9, 2015.

Callaghan, T., Christensen, T., and Jantze, E.: Plant and Vegetation Dynamics on Disko Island, West Greenland: Snapshots Separated by Over 40 Years, AMBIO, 40, 624-637, 10.1007/s13280-011-0169-x, 2011.

Castañeda, I. S., and Schouten, S.: A review of molecular organic proxies for examining modern and ancient

565 lacustrine environments, Quaternary Science Reviews, 30, 2851-2891, 10.1016/j.quascirev.2011.07.009, 2011.

Castellano, M. J., Mueller, K. E., Olk, D. C., Sawyer, J. E., and Six, J.: Integrating plant litter quality, soil organic matter stabilization, and the carbon saturation concept, Global Change Biol., 21, 3200-3209, 10.1111/gcb.12982, 2015.

Catalán, N., Obrador, B., Felip, M., and Pretus, J. L.: Higher reactivity of allochthonous vs. autochthonous DOC

570 sources in a shallow lake, Aquat. Sci., 75, 581-593, 10.1007/s00027-013-0302-y, 2013.

Chalmers, J. A., Pulvertaft, T. C. R., Marcussen, C., and Pedersen, A. K.: New insight into the structure of the Nuussuaq Basin, central West Greenland, Mar. Pet. Geol., 16, 197-224, 10.1016/S0264-8172(98)00077-4, 1999.

Conant, R. T., Ryan, M. G., Ågren, G. I., Birge, H. E., Davidson, E. A., Eliasson, P. E., Evans, S. E., Frey, S. D., Giardina, C. P., Hopkins, F. M., Hyvönen, R., Kirschbaum, M. U. F., Lavallee, J. M., Leifeld, J., Parton, W. J.,

575   Megan Steinweg, J., Wallenstein, M. D., Martin Wetterstedt, J. Å., and Bradford, M. A.: Temperature and soil organic matter decomposition rates – synthesis of current knowledge and a way forward, Global Change Biol., 17, 3392-3404, 10.1111/j.1365-2486.2011.02496.x, 2011.

Corell, R., Barry, T., Eamer, J., Hislop, L., Kullerud, L., Melillo, J., Nellemann, C., Neretin, L., Reiersen, L.-O., and Samseth, J.: The view from the top, in: Searching for responses to a rapidly changing Arctic in UNEP Year

580   Book 2013: Emerging issues in our global environment, United Nations Environment Programme, Nairobi, Kenya, 19-33, 2013.

Cranwell, P. A.: Diagenesis of free and bound lipids in terrestrial detritus deposited in a lacustrine sediment, Organic Geochemistry, 3, 79-89, 10.1016/0146-6380(81)90002-4, 1981.

Cranwell, P. A., Eglinton, G., and Robinson, N.: Lipids of aquatic organisms as potential contributors to lacustrine

585   sediments—II, Organic Geochemistry, 11, 513-527, 10.1016/0146-6380(87)90007-6, 1987.

Daniels, F. A., and De Molenaar, J.: Dry coastal ecosystems of Greenland, in: Ecosystems of the World 2A, Dry Coastal Ecosystems. Polar Regions and Europe, edited by: van der Maarel, E., Elsevier, Amsterdam, 39-50, 1993.

Daugbjerg, N.: Some Vascular Plants in the Vicinity of Godhavn, Disko Island - July 2002, University of Copenhagen, 93-101, 2003.

590   Ficken, K. J., Li, B., Swain, D. L., and Eglinton, G.: An n-alkane proxy for the sedimentary input of submerged/floating freshwater aquatic macrophytes, Organic Geochemistry, 31, 745-749, 10.1016/S0146-6380(00)00081-4, 2000.

Florian, C. R., Miller, G. H., Fogel, M. L., Wolfe, A. P., Vinebrooke, R. D., and Geirsdóttir, Á.: Algal pigments in Arctic lake sediments record biogeochemical changes due to Holocene climate variability and anthropogenic

595   global change, J. Paleolimnol., 54, 53-69, 10.1007/s10933-015-9835-5, 2015.

Florian, C. R.: Multi-Proxy Reconstructions of Holocene Environmental Change and Catchment Biogeochemistry Using Algal Pigments and Stable Isotopes Preserved in Lake Sediment from Baffin Island and Iceland, Geological Sciences University of Colorado, Boulder, 2016.

Fredskild, B.: The Holocene vegetational changes on Qeqertarsuatsiaq, a West Greenland island, Geografisk

600   Tidsskrift-Danish Journal of Geography, 100, 7-14, 10.1080/00167223.2000.10649434, 2000.

Glime, J.: Bryophyte Ecology. Volume 1. Physiological Ecology. Ebook sponsored by Michigan Technological University and the International Association of Bryologists. Accessed on 18.08. 2020. 2007.

Griffiths, K., Michelutti, N., Sugar, M., Douglas, M. S. V., and Smol, J. P.: Ice-cover is the principal driver of ecological change in High Arctic lakes and ponds, PLOS ONE, 12, e0172989, 10.1371/journal.pone.0172989,

605   2017.

Hanna, E., Mernild, S. H., Cappelen, J., and Steffen, K.: Recent warming in Greenland in a long-term instrumental (1881–2012) climatic context: I. Evaluation of surface air temperature records, Environmental Research Letters, 7, 045404, 10.1088/1748-9326/7/4/045404, 2012.

Hansen, B. U., Elberling, B., Humlum, O., and Nielsen, N.: Meteorological trends (1991–2004) at Arctic Station,
610   Central West Greenland (69°15'N) in a 130 years perspective, Geografisk Tidsskrift-Danish Journal of
       Geography, 106, 45-55, 10.1080/00167223.2006.10649544, 2006.
       Hanson, D. T., Renzaglia, K., and Villarreal, J. C.: Diffusion Limitation and CO_2 Concentrating Mechanisms in
       Bryophytes, in: Photosynthesis in Bryophytes and Early Land Plants, edited by: Hanson, T. D., and Rice, K. S.,
       Springer Netherlands, Dordrecht, 95-111, 2014.
615   Hobbie, S. E., Schimel, J. P., Trumbore, S. E., and Randerson, J. R.: Controls over carbon storage and turnover
       in high-latitude soils, Global Change Biology, 6, 196-210, 10.1046/j.1365-2486.2000.06021.x, 2000.
       Hobbs, W. O., Telford, R. J., Birks, H. J. B., Saros, J. E., Hazewinkel, R. R. O., Perren, B. B., Saulnier-Talbot,
       É., and Wolfe, A. P.: Quantifying Recent Ecological Changes in Remote Lakes of North America and Greenland
       Using Sediment Diatom Assemblages, PLOS ONE, 5, e10026, 10.1371/journal.pone.0010026, 2010.
620   Hodell, D. A., and Schelske, C. L.: Production, sedimentation, and isotopic composition of organic matter in Lake
       Ontario, Limnology and Oceanography, 43, 200-214, 10.4319/lo.1998.43.2.0200, 1998.
       Hollesen, J., Buchwal, A., Rachlewicz, G., Hansen, B. U., Hansen, M. O., Stecher, O., and Elberling, B.: Winter
       warming as an important co-driver for Betula nana growth in western Greenland during the past century, Global
       Change Biol., 21, 2410-2423, 10.1111/gcb.12913, 2015.
625   Holtvoeth, J., Rushworth, D., Copsey, H., Imeri, A., Cara, M., Vogel, H., Wagner, T., and Wolff, G. A.: Improved
       end-member characterisation of modern organic matter pools in the Ohrid Basin (Albania, Macedonia) and
       evaluation of new palaeoenvironmental proxies, Biogeosciences, 13, 795-816, 10.5194/bg-13-795-2016, 2016.
       Huang, X., Pancost, R. D., Xue, J., Gu, Y., Evershed, R. P., and Xie, S.: Response of carbon cycle to drier
       conditions in the mid-Holocene in central China, Nature Communications, 9, 1369, 10.1038/s41467-018-03804-
630   w, 2018.
       Hugelius, G., Strauss, J., Zubrzycki, S., Harden, J. W., Schuur, E. A. G., Ping, C. L., Schirrmeister, L., Grosse,
       G., Michaelson, G. J., Koven, C. D., O'Donnell, J. A., Elberling, B., Mishra, U., Camill, P., Yu, Z., Palmtag, J.,
       and Kuhry, P.: Estimated stocks of circumpolar permafrost carbon with quantified uncertainty ranges and
       identified data gaps, Biogeosciences, 11, 6573-6593, 10.5194/bg-11-6573-2014, 2014.
635   Humlum, O.: Origin of rock glaciers: Observations from Mellemfjord, Disko Island, Central West Greenland,
       Permafrost   and   Periglacial   Processes,   7,   361-380,   10.1002/(sici)1099-1530(199610)7:4<361::Aid-
       ppp227>3.0.Co;2-4, 1996.
       Humlum, O., Hansen, B. U., Nielsen, N., and Christiansen, H. H.: Meteorological observations 1998 at the arctic
       station, Qeqertarsuaq (69°15'N), Central West Greenland/Active layer monitoring in two Greenlandic permafrost
640   areas: Zackenberg and Disko Island, Geografisk Tidsskrift-Danish Journal of Geography, 99, 113-121,
       10.1080/00167223.1999.10649428, 1999.
       Jansson, M., Bergström, A.-K., Blomqvist, P., and Drakare, S.: Allochthonous organic carbon and
       phytoplankton/bacterioplankton production relationships in lakes, Ecology, 81, 3250-3255, 10.1890/0012-
       9658(2000)081[3250:aocapb]2.0.co;2, 2000.
645   Jeffrey, S. W., and Humphrey, G. F.: New spectrophotometric equations for determining chlorophylls a, b, $c1$, $c2$
       in higher plants, algae, and natural phytoplankton, Biochemie und Physiologie der Pflanzen, 167, 191-194,
       citeulike-article-id:906609, 1975.

Karlsson, J., Byström, P., Ask, J., Ask, P., Persson, L., and Jansson, M.: Light limitation of nutrient-poor lake ecosystems, Nature, 460, 506-509, 10.1038/nature08179, 2009.

650   Kortelainen, P., Rantakari, M., Pajunen, H., Huttunen, J. T., Mattsson, T., Juutinen, S., Larmola, T., Alm, J., Silvola, J., and Martikainen, P. J.: Carbon evasion/accumulation ratio in boreal lakes is linked to nitrogen, Global Biogeochemical Cycles, 27, 363-374, 10.1002/gbc.20036, 2013.

Lacey, J. H., Leng, M. J., Vane, C. H., Radbourne, A. D., Yang, H., and Ryves, D. B.: Assessing human impact on Rostherne Mere, UK, using the geochemistry of organic matter, Anthropocene, 21, 52-65,

655   10.1016/j.ancene.2018.02.002, 2018.

Lamoureux, S. F., and Lafrenière, M. J.: Seasonal fluxes and age of particulate organic carbon exported from Arctic catchments impacted by localized permafrost slope disturbances, Environmental Research Letters, 9, 045002, 10.1088/1748-9326/9/4/045002, 2014.

Lapierre, J.-F., and del Giorgio, P. A.: Geographical and environmental drivers of regional differences in the lake

660   pCO2 versus DOC relationship across northern landscapes, Journal of Geophysical Research: Biogeosciences, 117, 10.1029/2012jg001945, 2012.

Leavitt, P. R., Carpenter, S. R., and Kitchell, J. F.: Whole-lake experiments: The annual record of fossil pigments and zooplankton, Limnology and Oceanography, 34, 700-717, 10.4319/lo.1989.34.4.0700, 1989.

Leavitt, P. R., Cumming, B. F., Smol, J. P., Reasoner, M., Pienitz, R., and Hodgson, D. A.: Climatic control of

665   ultraviolet radiation effects on lakes, Limnology and Oceanography, 48, 2062-2069, 10.4319/lo.2003.48.5.2062, 2003.

Leavitt, P. R., Fritz, S. C., Anderson, N. J., Baker, P. A., Blenckner, T., Bunting, L., Catalan, J., Conley, D. J., Hobbs, W. O., Jeppesen, E., Korhola, A., McGowan, S., RÜhland, K., Rusak, J. A., Simpson, G. L., Solovieva, N., and Werne, J.: Paleolimnological evidence of the effects on lakes of energy and mass transfer from climate

670   and humans, Limnology and Oceanography, 54, 2330-2348, 10.4319/lo.2009.54.6_part_2.2330, 2009.

Leng, M. J., Wagner, B., Anderson, N. J., Bennike, O., Woodley, E., and Kemp, S. J.: Deglaciation and catchment ontogeny in coastal south-west Greenland: implications for terrestrial and aquatic carbon cycling, Journal of Quaternary Science, 27, 575-584, 10.1002/jqs.2544, 2012.

Mackereth, F. J. H., Heron, J., and Talling, J. F.: Water analysis: some revised methods for limnologists,

675   Freshwater Biological Association, 1978.

Marzi, R., Torkelson, B. E., and Olson, R. K.: A revised carbon preference index, Organic geochemistry, 20, 1303-1306, 10.1016/0146-6380(93)90016-5, 1993.

Matsuda, H., and Koyama, T.: Early diagenesis of fatty acids in lacustrine sediments—II. A statistical approach to changes in fatty acid composition from recent sediments and some source materials, Geochimica et

680   Cosmochimica Acta, 41, 1825-1834, 10.1016/0016-7037(77)90214-9, 1977.

Matthews, J. A., and Briffa, K. R.: The 'Little Ice Age': Re-evaluation of an evolving concept, Geografiska Annaler: Series A, Physical Geography, 87, 17-36, 10.1111/j.0435-3676.2005.00242.x, 2005.

McGowan, S., Barker, P., Haworth, E. Y., Leavitt, P. R., Maberley, S. C., and Pates, J.: Humans and climate as drivers of algal community change in Windermere since 1850, Freshwat. Biol., 57, 260-277, doi:10.1111/j.1365-

685   2427.2011.02689.x, 2012.

McGowan, S.: PALEOLIMNOLOGY | Pigment Studies, in: Encyclopedia of Quaternary Science (Second Edition), edited by: Elias, S. A., and Mock, C. J., Elsevier, Amsterdam, 326-338, 2013.

McGowan, S., Anderson, N. J., Edwards, M. E., Hopla, E., Jones, V., Langdon, P. G., Law, A., Solovieva, N., Turner, S., van Hardenbroek, M., Whiteford, E. J., and Wiik, E.: Vegetation transitions drive the autotrophy–heterotrophy balance in Arctic lakes, Limnology and Oceanography Letters, 3, 246-255, 10.1002/lol2.10086, 2018.

McGuire, A. D., Anderson, L. G., Christensen, T. R., Dallimore, S., Guo, L., Hayes, D. J., Heimann, M., Lorenson, T. D., Macdonald, R. W., and Roulet, N.: Sensitivity of the carbon cycle in the Arctic to climate change, Ecol. Monogr., 79, 523-555, 10.1890/08-2025.1, 2009.

Meyers, P. A., and Ishiwatari, R.: Lacustrine organic geochemistry—an overview of indicators of organic matter sources and diagenesis in lake sediments, Organic Geochemistry, 20, 867-900, 10.1016/0146-6380(93)90100-P, 1993.

Meyers, P. A., and Teranes, J. L.: Sediment Organic Matter, in: Tracking Environmental Change Using Lake Sediments. Volume 2: Physical and Geochemical Methods, edited by: Last, W. M., and Smol, J. P., Developments in Paleoenvironmental Research, Springer Netherlands, 239-269, 2001.

Meyers, P. A.: Applications of organic geochemistry to paleolimnological reconstructions: a summary of examples from the Laurentian Great Lakes, Organic Geochemistry, 34, 261-289, 10.1016/S0146-6380(02)00168-7, 2003.

Michelutti, N., Douglas, M. S. V., Muir, D. C. G., Wang, X., and Smol, J. P.: Limnological Characteristics of 38 Lakes and Pond son Axel Heiberg Island, High Arctic Canada, Int. Rev. Hydrobiol., 87, 385-399, 10.1002/1522-2632(200207)87:4<385::aid-iroh385>3.0.co;2-3, 2002.

Michelutti, N., Wolfe, A. P., Vinebrooke, R. D., Rivard, B., and Briner, J. P.: Recent primary production increases in arctic lakes, Geophysical Research Letters, 32, 10.1029/2005gl023693, 2005.

Michelutti, N., Douglas, M. S. V., Antoniades, D., Lehnherr, I., St. Louis, V. L., St. Pierre, K., Muir, D. C. G., Brunskill, G., and Smol, J. P.: Contrasting the ecological effects of decreasing ice cover versus accelerated glacial melt on the High Arctic's largest lake, Proceedings of the Royal Society B: Biological Sciences, 287, 20201185, doi:10.1098/rspb.2020.1185, 2020.

Miller, G. H., Geirsdóttir, Á., Zhong, Y., Larsen, D. J., Otto-Bliesner, B. L., Holland, M. M., Bailey, D. A., Refsnider, K. A., Lehman, S. J., Southon, J. R., Anderson, C., Björnsson, H., and Thordarson, T.: Abrupt onset of the Little Ice Age triggered by volcanism and sustained by sea-ice/ocean feedbacks, Geophysical Research Letters, 39, 10.1029/2011gl050168, 2012.

Moody, C. S., Worrall, F., Evans, C. D., and Jones, T. G.: The rate of loss of dissolved organic carbon (DOC) through a catchment, JHyd, 492, 139-150, 10.1016/j.jhydrol.2013.03.016, 2013.

Nelson, D. B., and Sachs, J. P.: The influence of salinity on D/H fractionation in dinosterol and brassicasterol from globally distributed saline and hypersaline lakes, Geochimica et Cosmochimica Acta, 133, 325-339, 10.1016/j.gca.2014.03.007, 2014.

Pautler, B. G., Austin, J., Otto, A., Stewart, K., Lamoureux, S. F., and Simpson, M. J.: Biomarker assessment of organic matter sources and degradation in Canadian High Arctic littoral sediments, Biogeochemistry, 100, 75-87, 10.1007/s10533-009-9405-x, 2010.

Pearson, E. J., Farrimond, P., and Juggins, S.: Lipid geochemistry of lake sediments from semi-arid Spain: Relationships with source inputs and environmental factors, Organic Geochemistry, 38, 1169-1195, 10.1016/j.orggeochem.2007.02.007, 2007.

Pearson, E. J., Juggins, S., Talbot, H. M., Weckström, J., Rosén, P., Ryves, D. B., Roberts, S. J., and Schmidt, R.: A lacustrine GDGT-temperature calibration from the Scandinavian Arctic to Antarctic: Renewed potential for the application of GDGT-paleothermometry in lakes, Geochimica et Cosmochimica Acta, 75, 6225-6238, 10.1016/j.gca.2011.07.042, 2011.

Phoenix, G. K., and Bjerke, J. W.: Arctic browning: extreme events and trends reversing arctic greening, Global Change Biol., 22, 2960-2962, 10.1111/gcb.13261, 2016.

Post, E., Forchhammer, M. C., Bret-Harte, M. S., Callaghan, T. V., Christensen, T. R., Elberling, B., Fox, A. D., Gilg, O., Hik, D. S., Høye, T. T., Ims, R. A., Jeppesen, E., Klein, D. R., Madsen, J., McGuire, A. D., Rysgaard, S., Schindler, D. E., Stirling, I., Tamstorf, M. P., Tyler, N. J. C., van der Wal, R., Welker, J., Wookey, P. A., Schmidt, N. M., and Aastrup, P.: Ecological Dynamics Across the Arctic Associated with Recent Climate Change, Science, 325, 1355-1358, 10.1126/science.1173113, 2009.

Pushkareva, E., Johansen, J. R., and Elster, J.: A review of the ecology, ecophysiology and biodiversity of microalgae in Arctic soil crusts, Polar Biology, 39, 2227-2240, 10.1007/s00300-016-1902-5, 2016.

Rampen, S. W., Abbas, B. A., Schouten, S., and Sinninghe Damste, J. S.: A comprehensive study of sterols in marine diatoms (Bacillariophyta): Implications for their use as tracers for diatom productivity, Limnology and Oceanography, 55, 91-105, 10.4319/lo.2010.55.1.0091, 2010.

Rowland, J., Jones, C., Altmann, G., Bryan, R., Crosby, B., Hinzman, L., Kane, D., Lawrence, D., Mancino, A., and Marsh, P.: Arctic landscapes in transition: responses to thawing permafrost, Eos, Transactions American Geophysical Union, 91, 229-230, 2010.

Rune, F.: Wild Flowers of Greenland, Gyldenlund Publishing, Hillerød, Denmark, 350 pp., 2011.

Saros, J. E., Anderson, N. J., Juggins, S., McGowan, S., Yde, J. C., Telling, J., Bullard, J. E., Yallop, M. L., Heathcote, A. J., Burpee, B. T., Fowler, R. A., Barry, C. D., Northington, R. M., Osburn, C. L., Pla-Rabes, S., Mernild, S. H., Whiteford, E. J., Andrews, M. G., Kerby, J. T., and Post, E.: Arctic climate shifts drive rapid ecosystem responses across the West Greenland landscape, Environmental Research Letters, 14, 074027, 10.1088/1748-9326/ab2928, 2019.

Schuur, E. A. G., Vogel, J. G., Crummer, K. G., Lee, H., Sickman, J. O., and Osterkamp, T. E.: The effect of permafrost thaw on old carbon release and net carbon exchange from tundra, Nature, 459, 556-559, 10.1038/nature08031, 2009.

Slemmons, K. E. H., and Saros, J. E.: Implications of nitrogen-rich glacial meltwater for phytoplankton diversity and productivity in alpine lakes, Limnology and Oceanography, 57, 1651-1663, 10.4319/lo.2012.57.6.1651, 2012.

Slemmons, K. E. H., Saros, J. E., and Simon, K.: The influence of glacial meltwater on alpine aquatic ecosystems: a review, Environmental Science: Processes & Impacts, 15, 1794-1806, 10.1039/C3EM00243H, 2013.

Smith, E. C., and Griffiths, H.: The occurrence of the chloroplast pyrenoid is correlated with the activity of a CO_2-concentrating mechanism and carbon isotope discrimination in lichens and bryophytes, Planta, 198, 6-16, 10.1007/bf00197580, 1996.

Smol, J. P., and Douglas, M. S.: From controversy to consensus: making the case for recent climate change in the Arctic using lake sediments, Front. Ecol. Environ., 5, 466-474, 10.1890/060162, 2007.

Sommaruga, R.: The role of solar UV radiation in the ecology of alpine lakes, J. Photochem. Photobiol. B: Biol., 62, 35-42, 10.1016/S1011-1344(01)00154-3, 2001.

Stevenson, M. A.: Carbon cycling in Arctic lakes: sedimentary biomarker reconstructions from Disko Island, West Greenland, University of Nottingham, 2017.

Tans, P. P., De Jong, A. F. M., and Mook, W. G.: Natural atmospheric ^{14}C variation and the Suess effect, Nature, 280, 826-828, 10.1038/280826a0, 1979.

Verburg, P.: The need to correct for the Suess effect in the application of δ13C in sediment of autotrophic Lake Tanganyika, as a productivity proxy in the Anthropocene, J. Paleolimnol., 37, 591-602, 10.1007/s10933-006-9056-z, 2007.

Volkman, J.: Sterols in microorganisms, Appl. Microbiol. Biotechnol., 60, 495-506, 10.1007/s00253-002-1172-8, 2003.

Volkman, J. K., Barrett, S. M., Blackburn, S. I., Mansour, M. P., Sikes, E. L., and Gelin, F.: Microalgal biomarkers: A review of recent research developments, Organic geochemistry, 29, 1163-1179, 10.1016/S0146-6380(98)00062-X, 1998.

Vonk, J. E., van Dongen, B. E., and Gustafsson, Ö.: Selective preservation of old organic carbon fluvially released from sub-Arctic soils, Geophysical Research Letters, 37, 10.1029/2010gl042909, 2010.

Williamson, C. J., Anesio, A. M., Cook, J., Tedstone, A., Poniecka, E., Holland, A., Fagan, D., Tranter, M., and Yallop, M. L.: Ice algal bloom development on the surface of the Greenland Ice Sheet, FEMS Microbiol. Ecol., 94, 10.1093/femsec/fiy025, 2018.

Wookey, P. A., Aerts, R., Bardgett, R. D., Baptist, F., Bråthen, K. A., Cornelissen, J. H. C., Gough, L., Hartley, I. P., Hopkins, D. W., Lavorel, S., and Shaver, G. R.: Ecosystem feedbacks and cascade processes: understanding their role in the responses of Arctic and alpine ecosystems to environmental change, Global Change Biol., 15, 1153-1172, 10.1111/j.1365-2486.2008.01801.x, 2009.

Yamanouchi, T.: Early 20th century warming in the Arctic: A review, Polar Science, 5, 53-71, 10.1016/j.polar.2010.10.002, 2011.

Yde, J. C., and Knudsen, N. T.: 20th-century glacier fluctuations on Disko Island (Qeqertarsuaq), Greenland, Annals of Glaciology, 46, 209-214, 10.3189/172756407782871558, 2007.

Zheng, Y., Zhou, W., Meyers, P. A., and Xie, S.: Lipid biomarkers in the Zoigê-Hongyuan peat deposit: Indicators of Holocene climate changes in West China, Organic Geochemistry, 38, 1927-1940, 10.1016/j.orggeochem.2007.06.012, 2007.

Tables

Table 1: Key lake characteristics. Presented in descending elevation order.

	Disko 2		Disko 1		Disko 4	
Lake coordinates	69º23.342'N, 53º24.085' W		69º21.204'N, 53º29.421'W		69º17.841'N, 53º48.548'W	
Elevation (m a.s.l)	575		299		214	
Lake type	Cirque lake		U-shaped valley bottom lake		Valley-end lake	
Outflow type	Over scree covered shallow wetlands		Wide braided wetland outflow – *more marked channel ~50 m d/s*		Direct outflow	
Total catchment area (ha)	358		1,455		1,829	
Imagery derived vegetation (ha)	46	13%	569	39%	497	27%
Imagery derived bare earth/rock (ha)	284	79%	650	45%	1,250	68%
Imagery derived permanent ice/glacier (ha)	17	5%	203	14%	67	3.6%
Vegetation survey derived total moss/lichen	27.3%		37.5%		37.0%	
Vegetation survey derived total plants	19.2%		32.7%		44.4%	
Vegetation survey derived total bare ground	53.5%		29.8%		18.6%	
Catchment: lake area ratio	45:1		66:1		122:1	
Outflow type	Over scree covered shallow wetlands		Wide braided wetland outflow – *more marked channel ~50 m downstream*		Direct outflow	

Table 2: Selected water chemistry spot samples from Disko Island lakes in April and August 2013. Dashes denoting missing values, BDL = below detection limit.

Lake	Disko 2		Disko 1		Disko 4	
2013 sampling	April	August	April	August	April	August
SRP (μg L^{-1})	10.5	0.7	59.8	13.8	16.3	1.9
TP (μg L^{-1})	37.5	2.9	67.9	67.8	164.8	2.9
NO^{3-} (μg L^{-1})	0.01	0.04	0.05	0.03	0.02	0.04
NH$_4^+$ (μg L^{-1})	BDL	3.2	BDL	11.5	BDL	BDL
Spectrophotometric Chl *a* (μg L^{-1})	0.9	0.7	1.0	0.4	0.1	1.0
Total N (mg L^{-1})	–	BDL	–	BDL	–	BDL
DOC (mg L^{-1})	1.0	0.6	0.6	0.9	1.5	0.8
Diatoxanthin (nmol pigment L^{-1})	–	0.05	–	0.63	–	0.08
Chlorophyll *a* (nmol pigment L^{-1})	–	0.55	–	0.46	–	0.85
Pheophytin *b* (nmol pigment L^{-1})	–	0.33	–	0.95	–	0.35
Date of collection	19/04	03/08	17/04	02/08	21/04	07/08

805

Table 3: Significant differences between pairings using parametric ANOVA post-hoc Tukey and non-parametric Mann-Whitney U

	ANOVA post-hoc Tukey	Man-Whitney U	Significant for both parametric and non-parametric tests?
	p adj	p-value	
C/N			
Core sediments vs aquatics	0.48	0.01	✗
Terrestrial 'other' vs aquatics	<0.01	0.31^	✗
Terrestrial vascular plants vs aquatics	0.01	0.03	✓
Terrestrial 'other' vs core sediments	<0.001	<0.001	✓
Terrestrial vascular plants vs core sediments	<0.001	<0.001	✓
Terrestrial vascular plants vs terrestrial 'other'	0.94	0.30^	✗
$\delta^{13}C_{org}$			
Core sediments vs aquatics	0.51	0.74	✗
Terrestrial 'other' vs aquatics	0.15	1^	✗
Terrestrial vascular plants vs aquatics	<0.001	0.46	✗
Terrestrial 'other' vs core sediments	0.22	0.03	✗
Terrestrial vascular plants vs core sediments	<0.001	<0.001	✓
Terrestrial vascular plants vs terrestrial 'other'	<0.001	<0.001	✓

Significant differences (p < 0.05) shaded

^Cannot compute exact p-value with ties

Table 4: Selected lipid ratios for catchment samples from Disko 2 analysed using GC-MS.

Sample	TOC %	*n*-alkane ratios				*n*-alkanoic acid (FAMEs) ratios		*n*-alkanol ratios		Sterols
		CPI 2[1]	TAR HC[2]	P_{WAX}[3]	*n*-C$_{27}$/ total *n*-alkanes	CPI$_T$[4]	*n*-C$_{30}$ / total *n*-alkanoic acids	*n*-C$_{16}$ / total *n*-alkanols	*n*-C$_{24}$ total *n*-alkanols	(Brassicasterol /total sterols) *1000
Harrimanella hypnoides (1)	43.0	39.6	18899.2	1.2	14.4	43.2	0.7	0.1	9.8	6.4
Harrimanella hypnoides (2)	42.4	27.6	2980.3	1.0	5.5	15.3	4.2	0.7	16.3	16.3
Unidentified herbaceous plant	49.2	25.9	1523.1	1.1	10.8	24.3	0.6	0.3	8.5	7.1
Black moss	40.9	3.7	3.0	0.7	11.2	69.7	0.0	8.2	11.0	56.6
Potamogeton sp.	32.1	3.6	68.5	0.9	7.7	74.3	0.0	12.4	9.8	14.5
Green moss	34.9	5.4	8.0	0.9	10.3	32.4	0.1	0.1	26.2	44.2
Algal benthic rock scrape	24.5	3.5	6.2	0.5	7.7	40.6	0.0	0.8	25.5	57.8
Chamerion latifolium	33.1	16.4	129.9	0.7	26.0	27.5	0.7	0.0	29.3	3.5
Salix arctica (leaf)	47.0	18.5	19.6	0.9	31.3	17.3	0.0	0.2	42.2	0.4
Catchment – Soil[5]	0.3	5.2	357.7	1.1	11.1	15.1	0.2	2.0	21.6	20.4
PLOT C – Soil[6]	0.1	1.3	18.9	0.8	10.2	11.7	2.0	4.9	12.3	44.7
Surface sediment	7.4	2.4	5.1	0.8	6.0	24.2	0.1	16.7	10.8	608.7

[1]CPI 2 – Carbon Preference Index 2 (Marzi et al., 1993) = $((C_{23}+C_{25}+C_{27})+(C_{25}+C_{27}+C_{29}))/2*(C_{24}+C_{26}+C_{28})$

[2]TAR$_{HC}$ – Terrigenous to Aquatic Ratio (Bourbonniere and Meyers, 1996) = $(C_{27}+C_{29}+C_{31})/(C_{15}+C_{17}+C_{19})$

[3]P_{WAX} – Index of waxy *n*-alkanes to total hydrocarbons (Zheng et al., 2007) = $(C_{27}+C_{29}+C_{31})/(C_{23}+C_{25}+C_{29}+C_{31})$

[4]CPI$_T$ – Carbon Preference Index (for the entire range) (Matsuda and Koyama, 1977) =
$0.5*((C_{12}+C_{14}+C_{16})+(C_{22}+C_{24}+C_{26}+C_{28}+C_{30}))+((C_{14}+C_{16}+C_{18})+(C_{24}+C_{26}+C_{28}+C_{30}+C_{32}))/((C_{13}+C_{15}+C_{17})+(C_{23}+C_{25}+C_{27}+C_{29}+C_{31}))$

Brassicasterol = 24-methylcholesta-5,22-dien-3β-ol
[5]from close to the highest point in the catchment 893 m (catchment summit)
[6]from close to shoreline (PLOT C)

Figures
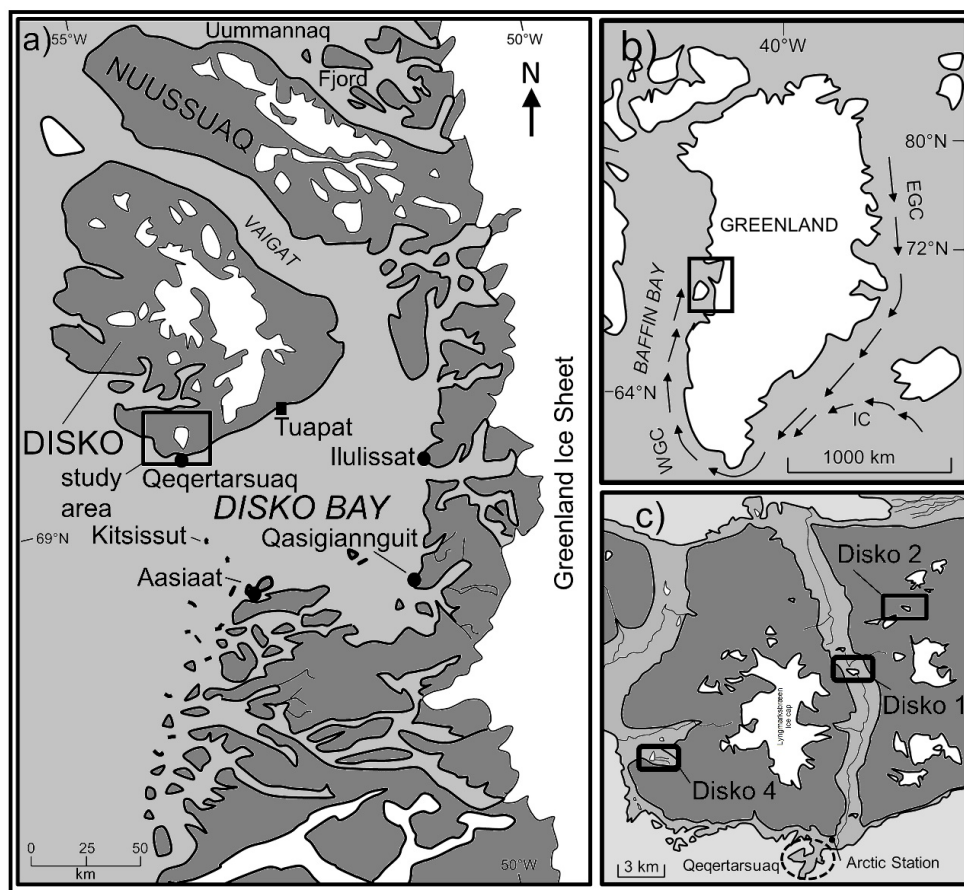
Figure 1: a) Location of Disko Island and study area (indicated) in West Greenland. b) Location of Disko Island relative to Greenland. c) Location of study catchments to the north of Qeqertarsuaq and Disko Island. See Stevenson (2017) for local catchment maps.
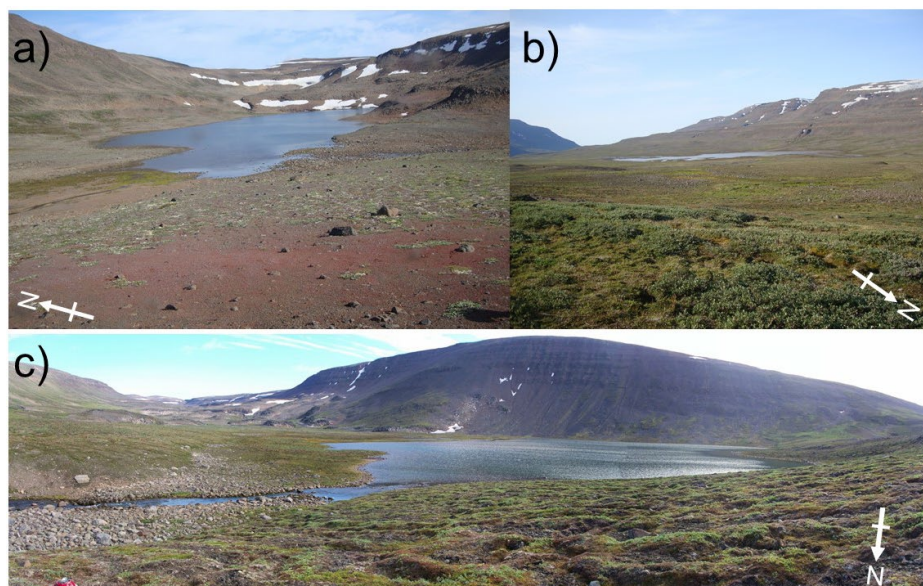
Figure 2: Images of study catchments and lakes on Disko Island in August 2013: a) Lake Disko 2 (M.Stevenson); b) Lake Disko 1 (M.Stevenson); c) Lake Disko 4 (J.Bailey).
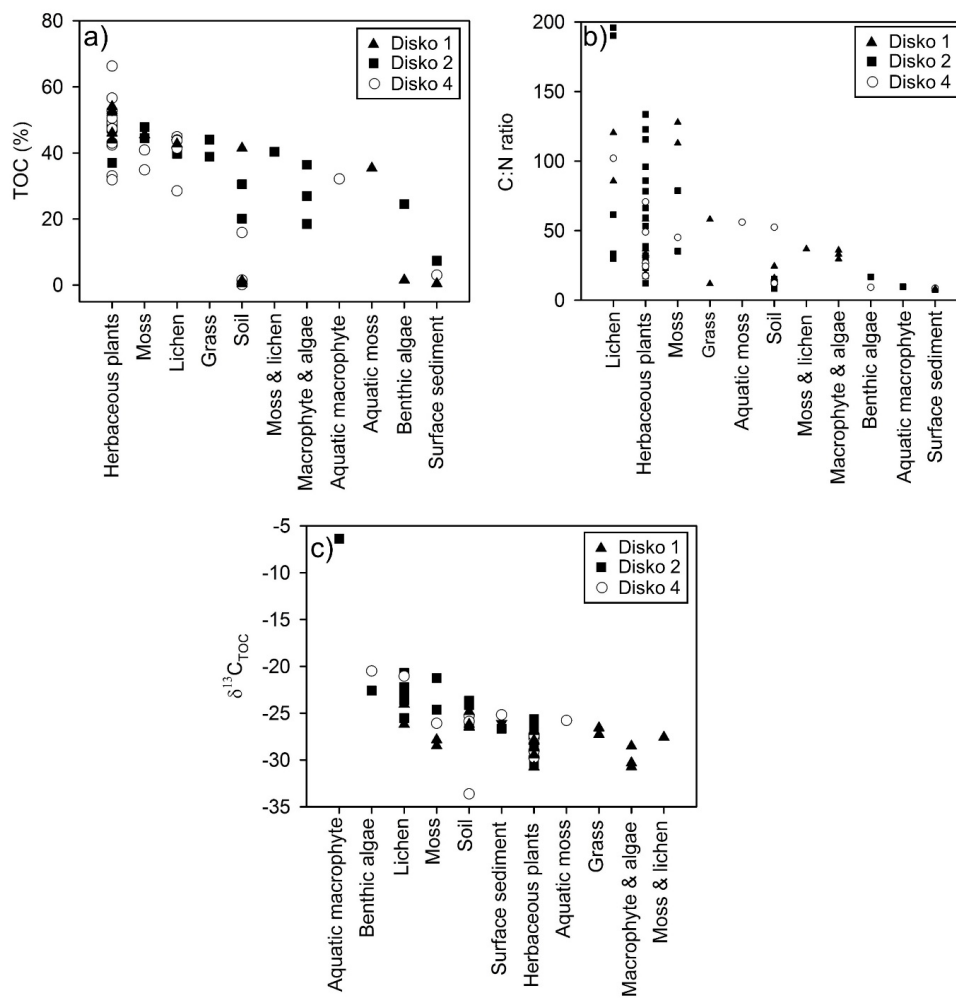
Figure 3: a) TOC (%); b) C$_{org}$:N ratio; and c) δ^{13}C$_{org}$ plotted against sample type, separated by lake catchment.

Figure 4: C_{org}/N vs $\delta^{13}C_{org}$ for catchment end-member samples and sediments from all three Disko catchment study areas. Grey shaded regions show expected zones for C_4 land plants, C_3 land plants and lacustrine algae according to Meyers (2003).
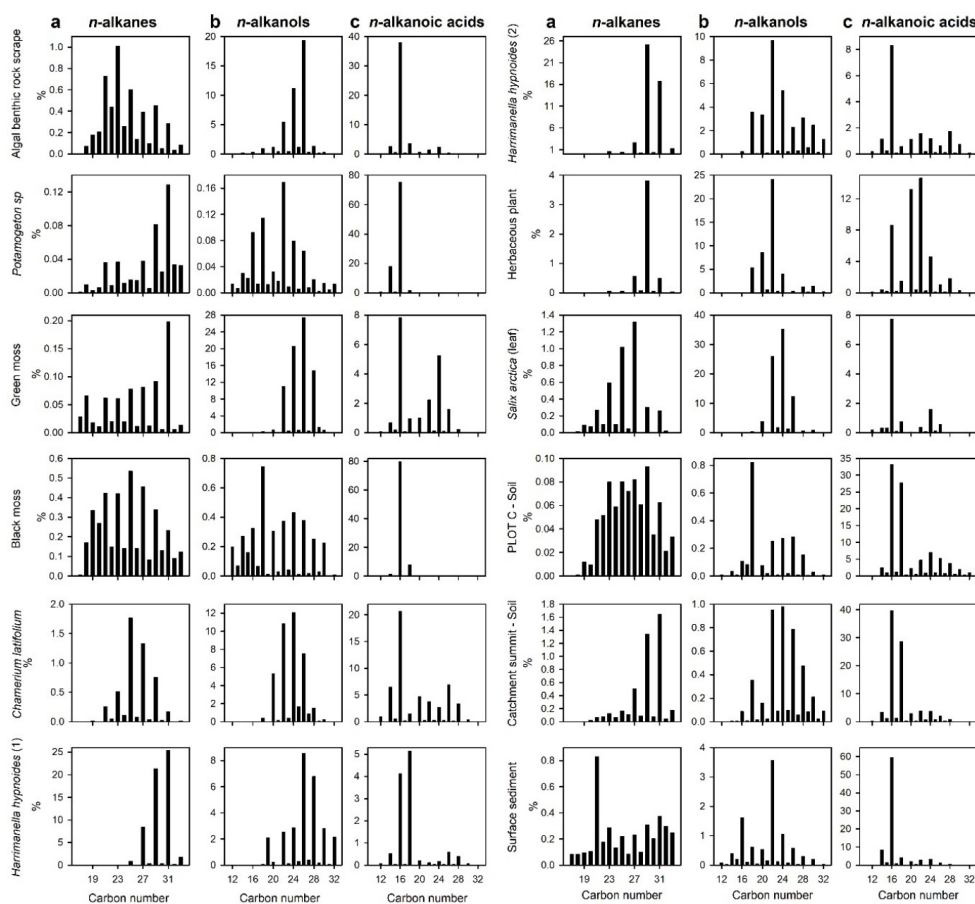
Figure 5: Lipid distribution histograms for a) *n*-alkanes, **b)** *n*-alkanols **and c)** *n*-alkanoic acids for catchment plant, soil and surface sediment samples from Disko 2 lake and catchment analysed using GC-MS. Expressed as relative proportion of total saturated *n*-alkanes, *n*-alkanols and *n*-alkanoic acids. Soil samples are taken from close to the Disko 2 shoreline (PLOT C) and close to the highest point in the catchment 893 m (catchment summit). Locations detailed in Stevenson (2017). Numbers (1) and (2) indicate replicates of separate samples from the Disko 2 catchment with the same identification.
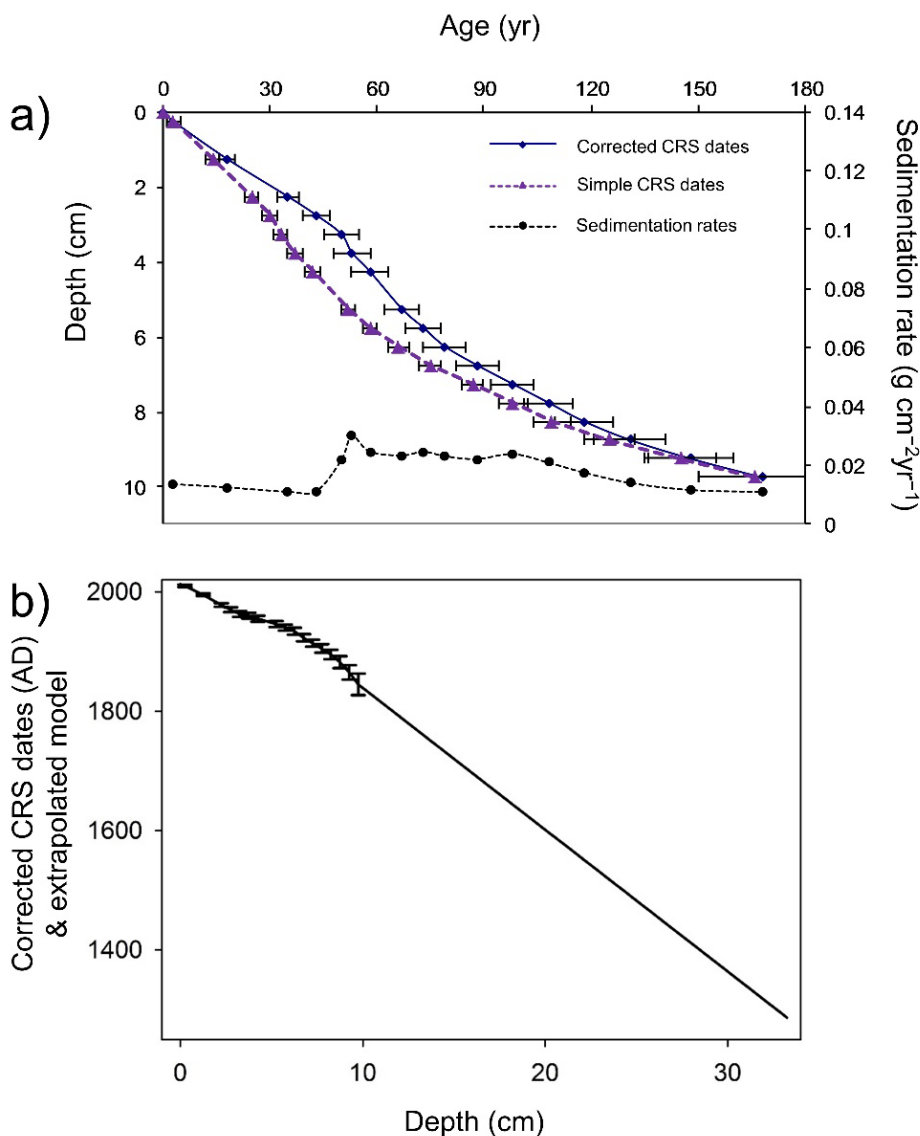
Figure 6: a) Age depth models and sedimentation rates using corrected CRS model for the Disko 2 K1 core. The simple CRS (constant rate of supply) model derived from ^{210}Pb dating only is indicated by purple triangles. The corrected CRS (constant rare of supply) model is derived from a ^{210}Pb base, with manual splice corrections based on the ^{137}Cs 1963 peak indicated by blue circles. b) Extrapolated model for Disko 2 K1 short core with linear extrapolation of dates between 1940 and 1845 AD used to extend the chronology to the base of the core. Error bars indicate measured ^{210}Pb samples to 1σ standard error.
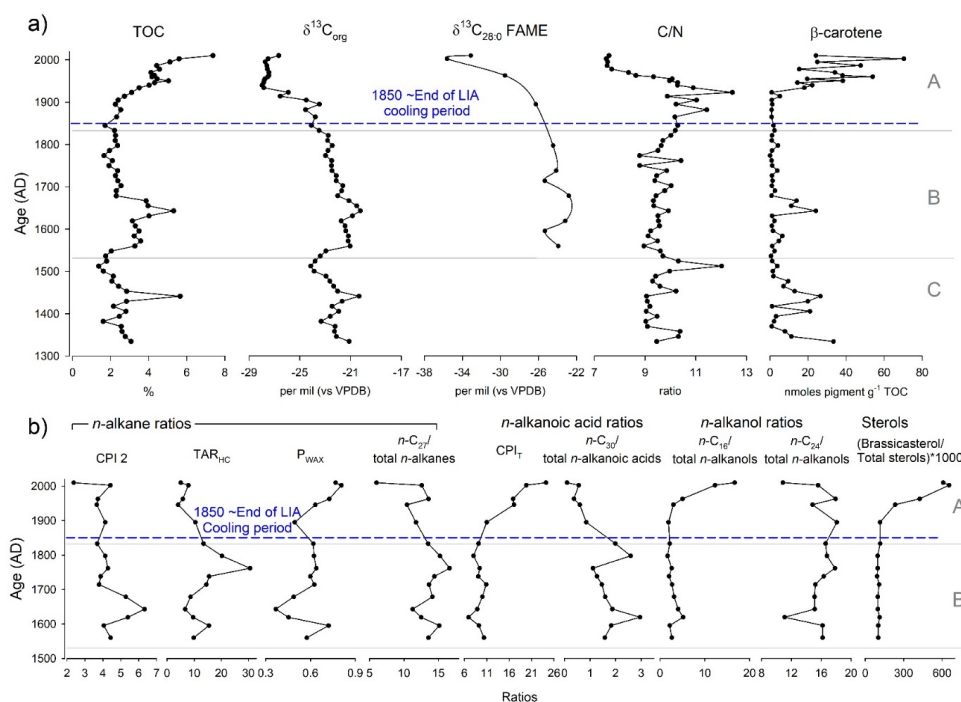
Figure 7: a) Down-core bulk and compound-specific isotopic geochemical parameters from the Disko 2 (K1) short core, presented against modelled age (AD). b) Down-core lipid geochemical ratios and equations including CPI 2, TAR$_{HC}$, P$_{WAX}$, n-C$_{27}$/total n-alkanes, CPI$_T$, n-C$_{30}$/total n-alkanoic acids, n-C$_{16}$/total n-alkanols, n-C$_{24}$/total n-alkanols and (brassicasterol/total sterols)*1000. For abbreviations see Table 4.