

Anthropocene climate warming enhances autochthonous carbon cycling in an upland Arctic lake, Disko Island, West Greenland

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

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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-amounts 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 ($\leq 1.5 \text{ mg L}^{-1}$). Sedimentary Carbon/Nitrogen (C/N) ratios, the C isotope composition of organic matter ($\delta^{13}\text{C}_{\text{org}}$) 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 ²¹⁰Pb 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. We also demonstrate that recent increases in catchment terrestrial vegetation cover contributed to the

45 **autochthonous response.** 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, **when** compared with extensively vegetated low Arctic systems.

Keywords. carbon cycling; lipid biomarkers; carbon isotopes; hydrogeomorphology; wetlands; lakes; sediments;
50 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
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)
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.,
60 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
experience particularly marked shifts in C cycling. Characterised by the presence (Low Arctic) or absence (High
65 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;
Lapierre and del Giorgio, 2012; McGowan et al., 2018). Previous studies in Greenland have used $\delta^{13}\text{C}_{\text{org}}$ and C/N
70 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
Schouten, 2011). These techniques, which have been rarely applied in high latitude lake catchments, can provide
75 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
lakes are CO₂ sources or sinks (Jansson et al., 2000; Kortelainen et al., 2013).

80 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

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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 (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 $\delta^{13}\text{C}_{\text{org}}$ biomarkers (C/N ratios and $\delta^{13}\text{C}_{\text{org}}/\delta^{13}\text{C}_{\text{FAMES}}$, chlorophyll and 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 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 69°15'N - 70°20'N and 51°50'W - 55°00'W. Disko is the largest island to the west of Greenland (8,575 km²) and is extensively glaciated with main Sermersuaq (Storbræe) 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 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 (Holleisen 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 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

120 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, ~3-5 m) for nutrient, DOC and pigment analysis.

2.1.1 Vegetation composition surveys

125 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 manually delineating the areas using the 2D area function in Google Earth Pro 7. In each lake catchment up to 130 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

135 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 -20°C for chlorophyll and carotenoid (pigment) and trichromatic chlorophyll *a* analyses. TP (total phosphorus), SRP (soluble reactive phosphorus) and $\text{NH}_4\text{-N}$ (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 sum of organically bound C present in water that originate from compounds which can pass through a $0.45\ \mu\text{m}$ 140 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 HON-Kajak corer. Disko 2 was selected for intensive study due to its well-dated sediment sequence. Sediment 145 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, with ^{226}Ra by the combined daughter isotope emissions of 295keV and 325keV to infer 'supported' ^{210}Pb (Fig S1 150 a). ^{137}Cs and ^{241}Am were measured at 662keV and 59.5keV (Appleby et al., 1986), with absolute efficiencies determined by comparison with standards (Fig. S1 b). 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 placed this slightly earlier at 5 cm and so ~~needed correcting~~ was corrected by 1.75 cm at this point in line with radiometric dating convention (Appleby, 2001) (Fig. 6). Estimated ages in the extrapolated part 155

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of the core (below 10 cm) are assumed to have the same error (± 18 years) as final measured sample in 1845 AD.

[The organic carbon mass accumulation rate \(CMAR\) was obtained by multiplying the dry mass accumulation rate \(DMAR\) by the decimal of TOC \(SI Fig. 3\).](#)

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2.3 Geochemical analyses

160 2.3.1 Chlorophyll and carotenoid pigment analysis

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; 5 μm 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).

170 2.3.2 Bulk $\delta^{13}\text{C}$ and C/N analysis

$\delta^{13}\text{C}_{\text{org}}$ 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 $\delta^{13}\text{C}_{\text{org}}$ 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. $\delta^{13}\text{C}$ 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 $\delta^{13}\text{C}_{\text{org}}$ was to within $\pm 0.1\text{‰}$ (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. [An isotopic mixing model based on a Bayesian framework was developed using SIAR V4 \(Stable Isotope Analysis in R\) \(Inger et al., 2010\) in R \(v3.5.2\) \(R Core Team, 2020\) to couple catchment \$\delta^{13}\text{C}_{\text{org}}\$ and C/N ratio as source data with down-core measurements \(grouped by sedimentary zone\). Proportional boxplots were produced for both broad catchment groups \(terrestrial other, vascular plant and aquatics\) and at the finer species level \(SI Fig. 4-7\). Trophic enrichment factors were set at a mean of 1.63 ‰ and standard deviation of 0.63 ‰ for \$\delta^{13}\text{C}\$ \(Inger et al., 2010\), with a value of zero used for C/N ratio.](#)

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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

195 from the neutral fraction using CH₂Cl₂ extracted ultrapure H₂O. 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
200 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 C₆H₁₄. 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 °C in CH₂Cl₂ using a HP DB5-MS (30m x 0.25 mm
210 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

205 Compound-specific δ¹³C was obtained by injecting FAME acid fractions spiked with squalene (temperature
290°C) 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 δ¹³C values (Chiron, Norway) and daily
210 combined isotopic reference samples from C₁₂ to C₃₂ (Indiana University). δ¹³C values were relative to the VPDB
(‰) and corrections were made for the additional C atoms introduced by BF₃-MeOH derivatisation (Boschker et
al., 1999).

3 Results

3.1 Hydro-geomorphology and vegetation surveys of lake catchments

215 The highest elevation cirque lake (Disko 2) had the least imagery-derived catchment vegetation and the smallest
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
220 this coverage was greatest in the catchment of Disko 1 which is fed directly by the Lyngmarksbræen ice cap (Fig.
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

225 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
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 (NO₃-N) which was highest in Disko 1 (0.05 µg L⁻¹ in August) and

lowest in Disko 2 (0.01 $\mu\text{g L}^{-1}$ in April) whereas $\text{NH}_4^+\text{-N}$ concentrations were below detection limits, except in August in Disko 1 and Disko 2 (11.54 $\mu\text{g L}^{-1}$ and 3.15 $\mu\text{g L}^{-1}$ and respectively). DOC was low in all lakes in both 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}).

3.3 TOC, C/N ratios and $\delta^{13}\text{C}$ 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). $\delta^{13}\text{C}_{\text{org}}$ ranged between -6.4‰ and -33.6 ‰ (Fig. 3c). Comparing the C/N ratio and $\delta^{13}\text{C}_{\text{org}}$ values, most 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 $\delta^{13}\text{C}_{\text{org}}$ values (~-20 to -23 ‰) (Fig. 4). Benthic macroalgae samples had $\delta^{13}\text{C}_{\text{org}}$ values around -25 ‰. Lake sediment core samples from Disko 2 & 4 had low C/N ratios (7.5 to 18.4) and $\delta^{13}\text{C}_{\text{org}}$ values ranged from -28 to -20 ‰. Statistical analyses demonstrated that C/N 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 $\delta^{13}\text{C}_{\text{org}}$ and so terrestrial vascular plant measurements were only significantly different to core sediments and terrestrial 'other' (Table 3; Fig. 4). Compound-specific $\delta^{13}\text{C}_{\text{FAMES}}$ values tended to become progressively lower (more negative) for higher chain length fatty acids than shorter chains, compared with bulk $\delta^{13}\text{C}_{\text{org}}$ (SI Fig. 34).

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%) 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}).

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 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

270 surface sediments suggesting multiple inputs. Although the algal benthic rock scrape and one *H. hypnoides* sample had similar $n\text{-C}_{26}$ 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 ($n\text{-C}_{26}$ ca. 20%) is different compared with *H. hypnoides* ($n\text{-C}_{26}$ ca. 9%), making these compositions distinguishable. The n -alkanoic acid $n\text{-C}_{16}$ was ubiquitous and abundant in all samples, but distributions still provided some differentiation, for example a secondary abundance in green moss around $n\text{-C}_{24}$, $n\text{-C}_{22}$ in the unidentified herbaceous plant sample and $n\text{-C}_{24}$ in the *Salix arctica* (leaf) and $n\text{-C}_{18}$ in the plot C soil sample.

275 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 of terrestrial plants, TAR_{HC} is an excellent indicator of leafy vascular plants, P_{WAX} indicates multiple terrestrial inputs and $n\text{-C}_{27}$ 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 $n\text{-C}_{30}$ of non-woody herbaceous terrestrial inputs. n -Alkanol indicator ratios are $n\text{-C}_{16}$ for aquatic macrophytes and $n\text{-C}_{24}$ 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

285 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 $\delta^{13}\text{C}_{\text{org}}$ 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 $\delta^{13}\text{C}_{\text{org}}$ 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).

290 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 $\delta^{13}\text{C}_{\text{org}}$ decreased from ~ -23.5 to -28 ‰ by ~1940 AD where it remained stable to ~2003 (± 2) AD (Fig. 7a). $\delta^{13}\text{C}_{28:0}$ 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). Both DMAR and CMAR were characterised by a clear pulsed increase to higher levels between ~1930 and 1960 AD (SI Fig. 3).

300 SIAR isotopic mixing model analysis linking the catchment with down-core $\delta^{13}\text{C}$ and C/N ratios revealed higher proportional densities (expressed in both histograms and boxplots) for vascular plants in group 1 (zone A 2013-1830 AD) than groups 2 (zone B 1830-1530 AD) and 3 (zone C 1530 -1300 AD) (Fig SI 4 & 5). At the species level although mixed, changes in *Betula nana*, *Chamerion latifolium*, *Eriophorum*, guano and soil explain

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the most variation in group 1 (zone A 2013-1830 AD) the most evidenced by proportional histograms and boxplots (Fig SI 5 & 6). For both groups 2 (zone B 1830-1530 AD) and 3 (zone C 1530-1300 AD), although mixed, the aquatic group is the most important with, benthic algae the greatest contributor.

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4 Discussion

310 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 ($\leq 1.5 \text{ mg L}^{-1}$, 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 $13.8 \text{ } \mu\text{g L}^{-1}$, TP $67.9 \text{ } \mu\text{g L}^{-1}$ and NH_4^+ $11.5 \text{ } \mu\text{g L}^{-1}$) 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

345 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.

350 **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
355 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 N₂ 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
360 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 δ¹³C_{org} there was more overlap in values between surface sediment and
365 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 δ¹³C_{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,
370 2001). In contrast to higher plants, bryophytes lack stomata and their isotope fractionation is closer to atmospheric
CO₂ (Smith and Griffiths, 1996) (Hanson et al., 2014; Glime, 2007). On the other hand, variability in δ¹³C_{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 CO₂. Higher
values of δ¹³C_{org} in these lake sediments indicate either greater contributions of algae, phytobenthos and
375 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?

380 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 CPI₂ values in soil and surface sediment reflect both the possibility of degradation of any minor terrestrial sources and in sediments, and the predominant aquatic 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 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. C₃₀ *n*-alkanoic acid contributions were highest in 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 terrestrial inputs. The C₂₄ *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 soil 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* sp., black moss and the algal benthic rock scrape indicating high proportions of non-degraded C (Table 4). The C₁₆ *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 C₂₇ 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), C₁₆ *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*-C₂₇ and *n*-C₃₀ 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

420 after 1950 to understand how catchment mediated climate changes affect C cycling in this snowpack-fed cirque lake.

425 Aside from brief increases in % TOC and β -carotene around ~1450 AD and ~1650 AD, there was a major shift in these proxies after from ~1860 AD (corresponding to the end of the LIA), to the present day with a >10-fold increase in β -carotene (from ~1910 AD to recent maximum) 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., 430 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 by responding in a two-stage, pulsed manner. This contrasts with many Low Arctic systems, where catchment 435 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 $\delta^{13}\text{C}_{\text{org}}$ in the most recent sediments in Disko 2 is opposite to the expectation that higher productivity from nutrient enrichment leads to higher $\delta^{13}\text{C}_{\text{org}}$ (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 $\delta^{13}\text{C}_{\text{org}}$ probably results from a source change such as increased algae and macrophyte abundance. Combined, these geochemical markers 440 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 recent warming and regional Disko glacier retreat (since ~1950 AD) (Yde and Knudsen, 2007; Hollesen et al., 2015; Box, 2002). The major two-step threshold shift since the end of the LIA (Fig. 7), is likely regulated by catchment processes associated with a glacier and/or permanent ice processes (e.g. nivation hollows, other periglacial processes) in the Disko 2 catchment, releasing N and P to the lake, shifting nutrient conditions and stimulating an algal response. Although 2013 surveys revealed only a small amount of permanent ice in this catchment, it is likely that in the LIA there was a larger cirque glacier in the catchment. Glaciers can respond not only to temperature but also precipitation which regulates their hydrological connectivity and subsequent release of N, P and C to lakes. During retreat, ice-entrained nutrition can be discharged and soil surfaces exposed, contributing to threshold shifts in algal species and abundance (Slemmons et al., 2017; Slemmons et al., 2013; Colombo et al., 2019). One possible source of N and P accumulation on glaciers is via aeolian transport of dust, which has many potential sources on Disko, an actively glaciated environment, with likely greater permanent ice cover in the LIA. Mechanistically, regional dust variability in Greenland is known to be a key determinant of sediment supply (Bullard and Mockford, 2018) and there is evidence that dust supplied P can stimulate Arctic lake algal response (Anderson et al., 2017). This is because

Commented [MS9]: To reflect comment from reviewer #1 on timing of some proxies response which is somewhat more recent than the end of the LIA.

Commented [MS10]: To reflect comment from reviewer #1 on the threshold response in multiple geochemical indicators.

Commented [MS11]: This helps address comment from reviewer #1 who recommended we consider the origin of the threshold response of C/N and $\delta^{13}\text{C}_{\text{org}}$ linked to nutrients in the water column and catchment filters which may have delayed the lacustrine response.

460 dust is known to stimulate microbial and algal abundance on the glacier surface (Stibal et al., 2015), which subsequently becomes entrained in glacier ice and released to downstream lakes on retreat.

465 [The timing of the most marked lacustrine response (Fig. 7) appears to be from the mid-20th century onwards, slightly later than the changes after the end of the LIA. Evidence for recent regional warming includes significant Greenland summit annual surface temperature increase between 1982 and 2011 (McGrath et al., 2013), increased runoff from melt from the Greenland ice sheet in response to climate warming (Hanna et al., 2008), recent warming at west Greenland coastal weather stations between 1981 and 2011/12 (Hanna et al., 2012), significant increases in the Greenland Blocking Index (GBI) since 1981 in all seasons and annually (Hanna et al., 2016) and significant warming of the polar water layer in Disko Bay, responsible (in part) for the retreat of the Jakobshavn Isbrae at Ilulissat (Myers and Ribergaard, 2013). Although recent warming (last ~40 years) is known to be the most pronounced, it is challenging to interpret if warming was more pronounced in the middle 20th century than ~100 years earlier, when the LIA ended due to limitations from instrumental temperature records. Taking a palaeoclimate approach Overpeck et al. (1997) summarised multiple records suggesting that peak 20th century temperature in 1945 AD across the Arctic was around 1.2 °C more than in 1910. This suggests that mid-20th century warmth in the Arctic overall was probably greater than at the end of the LIA, but due to the spatial heterogeneity of these climate changes in different localities of the Arctic it is hard to compare these time periods specifically on Disko Island. However, these indicators are not direct temperature or climate proxies (Fig 7), but rather indicators of carbon cycling which are indirectly related to both catchment processes and climate, releasing nutrients and carbon and stimulating the recent algal response. Temperature is typically only one of many factors which drive changes in ecological thresholds in Greenland lakes (Anderson et al., 2018; Law et al., 2015; McGowan et al., 2018; Saros et al., 2019; Axford et al., 2013). Therefore the ecological response to regional warming is probably indirect through changes in catchment and shifts in the terrestrial carbon cycle, rather than solely a direct lake-water algal temperature response. Interestingly, both DMAR and CMAR increased in a pulsed manner between ~1930 and 1960 AD, suggesting processes in the Disko 2 catchment may have been particularly active during the mid-20th century, likely associated with glacier retreat or changes in catchment periglacial processes.

485 Although terrestrial OM was unlikely to be the dominant source of OM to Disko 2 sediments, there are indications that the type and magnitude of terrestrial OM fluxes have subtly varied over time and that recent changes in the catchment may be reflected in changes in the sediment. For example, increases in the C/N ratio around ~1900 AD suggest catchment dieback, facilitated by warming during melt season exposure from reduced ice cover (Leng et al., 2012). The composition of catchment-derived C also shifted after ~1895 AD with a decline in $\delta^{13}\text{C}_{28:0}$ FAME from terrestrial plants, indicating a shift in terrestrial species with lower $\delta^{13}\text{C}_{28:0}$ values as the catchment thawed and nutrient availability increased. $\delta^{13}\text{C}_{\text{FAME}}$ values of Disko 2 catchment samples were lower (isotopically-depleted) in higher plants (e.g. *Harrimanella hypnoides* & *Chamerion latifolium*) (Table-Fig. S32). Increases in *n*-alkane CPI 2 index between 1700 and 1600 AD probably reflect an earlier period of increased 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 hypnoides* and other herbaceous

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Commented [MS13]: This helps address comment from reviewer #1 who recommended we consider the timing of the main lacustrine response which appears to take place after the end of the LIA in the middle 20th century and assess when warming was most pronounced and discuss interrelationships between warming and changes in the catchment.

Commented [MS14]: This helps address comment from reviewer #1 who recommended we consider total C plotted as flux, rather than just %. Presence of pulse in CMAR and DMAR in the mid-20th Century underscores the close link between the lake and catchment.

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500 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 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 suggest 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*-C₂₄ to total *n*-alkanols declined only in the uppermost samples (post-2003 AD), perhaps influenced by the most recent changes in catchment vegetation on Disko (Holleisen 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). Interestingly, despite limited vegetation cover in the Disko 2 catchment, SIAR isotopic mixing model analysis linking catchment with down-core C/N and $\delta^{13}C_{org}$ shows higher proportion densities for vascular plants in group 1 (zone A 2013-1830AD) than further down the core, supporting the idea that changes in terrestrial vegetation cover were contemporaneous with changes in the sediment (Fig. SI 4 and 5). At a finer scale, highly mixed *Betula nana*, *Chamerion latifolium*, *Eriophorum*, guano and soil explain the most variation in zone A, whereas in earlier sediments (zone B 1830-1530 AD and zone C 1530 -1300 AD) benthic algae explain the most variation in down-core C/N and $\delta^{13}C_{org}$ (Fig. SI 6 and 7). This supports the idea that changes in the catchment vegetation linked to warming probably also played a role in the major autochthonous response in the Disko 2 lake.

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

525 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 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-catchment systems in Alaska and Sweden (McGowan et al., 2018). Despite limited terrestrial-aquatic C fluxes in Disko lakes, there is probably likely 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.

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

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540 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
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
545 occurred coincident with climate-mediated catchment change, likely coupled to glacier and permafrost/soil
nutrient and C cycling.

5 Conclusions

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

- i. C in lake sediments in sparsely vegetated catchments mostly derives from autochthonous algal sources
with minor terrestrial catchment carbon inputs, likely involved in stimulating recent change;
- 555 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
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.

560 Principally our findings illustrate how lakes with limited allochthonous C input respond more directly to
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
565 where catchment buffering in poorly vegetated catchments is limited.

Data availability

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

Author contribution

570 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
fieldwork. MAS, EJP and EW carried out spring 2013 sediment coring. All authors revised and approved the
manuscript.

Commented [MS18]: This is now much more certain given the helpful outcome from SIAR analysis, recommended by reviewer #2.

Competing interests

575 The authors declare that they have no conflict of interest.

Acknowledgements

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 $\delta^{13}\text{C}_{\text{org}}$ & C/N ratios on sediment, soil and plant samples. Lipid and water chemistry analyses were
580 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, 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
585 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

- 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.
- 590 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-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
595 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 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.
- 600 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.
- 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.
- 605 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 Reviews*, 59, 87-100, doi.org/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.
- 610 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 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.
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- 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.
- 620 Box, J. E.: Survey of Greenland instrumental temperature records: 1873–2001, *IJCLI*, 22, 1829-1847, 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.
- 625 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 climate change in Arctic Canada and Greenland, *Quaternary Science Reviews*, 147, 340-364, 10.1016/j.quascirev.2016.02.010, 2016.
- 630 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, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of bulk organic matter, and on environmental interpretation, from acid treatment methods, *Quaternary Science Reviews*, 30, 3076–3087, 2011.
- 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.
- 635 Bullard, J. E., and Mockford, T.: Seasonal and decadal variability of dust observations in the Kangerlussuaq area, west Greenland, *Arct. Antarct. Alp. Res.*, 50, S100011, 10.1080/15230430.2017.1415854, 2018.
- 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.
- 640 Castañeda, I. S., and Schouten, S.: A review of molecular organic proxies for examining modern and ancient 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.
- 645 Catalán, N., Obrador, B., Felip, M., and Pretus, J. L.: Higher reactivity of allochthonous vs. autochthonous DOC 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.
- 650 Colombo, N., Bocchiola, D., Martin, M., Confortola, G., Salerno, F., Godone, D., D'Amico, M. E., and Freppaz, M.: High export of nitrogen and dissolved organic carbon from an Alpine glacier (Indren Glacier, NW Italian Alps), *Aquat. Sci.*, 81, 74, 10.1007/s00027-019-0670-z, 2019.
- 655 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., Lavalley, J. M., Leifeld, J., Parton, W. J., 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.
- 660 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 Book 2013: Emerging issues in our global environment, United Nations Environment Programme, Nairobi, Kenya, 19-33, 2013.
- 665 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 sediments—II, *Organic Geochemistry*, 11, 513-527, 10.1016/0146-6380(87)90007-6, 1987.

- 670 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.
- 675 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 global change, *J. Paleolimnol.*, 54, 53-69, 10.1007/s10933-015-9835-5, 2015.
- 680 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 Qeqertarsuaq, a West Greenland island, *Geografisk Tidsskrift-Danish Journal of Geography*, 100, 7-14, 10.1080/00167223.2000.10649434, 685 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.
- 690 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, 2017.
- Hanna, E., Huybrechts, P., Steffen, K., Cappelen, J., Huff, R., Shuman, C., Irvine-Fynn, T., Wise, S., and Griffiths, M.: Increased Runoff from Melt from the Greenland Ice Sheet: A Response to Global Warming, *J. Clim.*, 21, 331-341, 10.1175/2007jcli1964.1, 2008.
- 695 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.
- Hanna, E., Cropper, T. E., Hall, R. J., and Cappelen, J.: Greenland Blocking Index 1851–2015: a regional climate change signal, *IJCli*, 36, 4847-4861, doi.org/10.1002/joc.4673, 2016.
- 700 Hansen, B. U., Elberling, B., Humlum, O., and Nielsen, N.: Meteorological trends (1991–2004) at Arctic Station, 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 CO₂ Concentrating Mechanisms in Bryophytes, in: *Photosynthesis in Bryophytes and Early Land Plants*, edited by: 705 Hanson, T. D., and Rice, K. S., Springer Netherlands, Dordrecht, 95-111, 2014.
- 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.
- 710 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.
- 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, 715 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.
- 720 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-w, 2018.
- 725 Hugelius, G., Strauss, J., Zubrzycki, S., Harden, J. W., Schuur, E. A. G., Ping, C. L., Schirmer, 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.
- 730 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.
- 735 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 areas: Zackenberg and Disko Island, *Geografisk Tidsskrift-Danish Journal of Geography*, 99, 113-121, 10.1080/00167223.1999.10649428, 1999.
- Inger, R., Jackson, A., Parnell, A., and Bearhop, S.: SIAR v4 (Stable Isotope Analysis in R): an ecologist's guide, Dublin, Ireland, 2010.
- 740 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.
- 745 Jeffrey, S. t., and Humphrey, G.: New spectrophotometric equations for determining chlorophylls a, b, c1 and c2 in higher plants, algae and natural phytoplankton, *Biochemie und physiologie der pflanzen*, 167, 191-194, 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.
- 750 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, 10.1016/j.ancene.2018.02.002, 2018.
- 755 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 pCO₂ versus DOC relationship across northern landscapes, *Journal of Geophysical Research: Biogeosciences*, 117, 10.1029/2012jg001945, 2012.
- 760 Law, A. C., Anderson, N. J., and McGowan, S.: Spatial and temporal variability of lake ontogeny in south-western Greenland, *Quaternary Science Reviews*, 126, 1-16, doi.org/10.1016/j.quascirev.2015.08.005, 2015.
- 765 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.
- 770 Leavitt, P. R., Cumming, B. F., Smol, J. P., Reasoner, M., Pienitz, R., and Hodgson, D. A.: Climatic control of ultraviolet radiation effects on lakes, *Limnology and Oceanography*, 48, 2062-2069, 10.4319/lo.2003.48.5.2062, 2003.
- 775 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 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, Freshwater Biological Association, 1978.
- 780 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 Cosmochimica Acta*, 41, 1825-1834, 10.1016/0016-7037(77)90214-9, 1977.
- 785 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.1365-2427.2011.02689.x, 2012.
- 790 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.
- 795 McGrath, D., Colgan, W., Bayou, N., Muto, A., and Steffen, K.: Recent warming at Summit, Greenland: Global context and implications, *Geophysical Research Letters*, 40, 2091-2096, doi.org/10.1002/grl.50456, 2013.
- 800 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.
- 805 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.
- 810 Michelutti, N., Douglas, M. S. V., Muir, D. C. G., Wang, X., and Smol, J. P.: Limnological Characteristics of 38 Lakes and Pondson 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.
- 815 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., Antoniadou, D., Lehnher, 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.
- 820 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.
- 825 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.
- Myers, P. G., and Ribergaard, M. H.: Warming of the Polar Water Layer in Disko Bay and Potential Impact on Jakobshavn Isbrae, *Journal of Physical Oceanography*, 43, 2629-2640, 10.1175/jpo-d-12-051.1, 2013.
- 830 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.

- Overpeck, J., Hughen, K., Hardy, D., Bradley, R., Case, R., Douglas, M., Finney, B., Gajewski, K., Jacoby, G., Jennings, A., Lamoureux, S., Lasca, A., MacDonald, G., Moore, J., Retelle, M., Smith, S., Wolfe, A., and Zielinski, G.: Arctic Environmental Change of the Last Four Centuries, *Science*, 278, 1251-1256, 10.1126/science.278.5341.1251, 1997.
- 835 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.
- 840 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.
- 845 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., 850 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.
- 855 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.
- R Core Team: R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria, 2020.
- 860 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.
- 865 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.
- 870 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.
- 875 Slemmons, K. E., Medford, A., Hall, B. L., Stone, J. R., McGowan, S., Lowell, T., Kelly, M., and Saros, J. E.: Changes in glacial meltwater alter algal communities in lakes of Scoresby Sund, Renland, East Greenland throughout the Holocene: Abrupt reorganizations began 1000 years before present, *The Holocene*, 27, 929-940, 10.1177/0959683616678468, 2017.
- 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.
- 880 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.
- 885 Smith, E. C., and Griffiths, H.: The occurrence of the chloroplast pyrenoid is correlated with the activity of a CO₂-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.

890 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.

Stibal, M., Gözdereliler, E., Cameron, K. A., Box, J. E., Stevens, I. T., Gokul, J. K., Schostag, M., Zarsky, J. D., Edwards, A., Irvine-Fynn, T. D. L., and Jacobsen, C. S.: Microbial abundance in surface ice on the Greenland Ice Sheet, *Frontiers in Microbiology*, 6, 10.3389/fmicb.2015.00225, 2015.

895 Tans, P. P., De Jong, A. F. M., and Mook, W. G.: Natural atmospheric ¹⁴C 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.

900 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.

905 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.

915 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.

920 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.

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

	Disko 2		Disko 1		Disko 4	
Lake coordinates	69°23.342'N, 53°24.085' W		69°21.204'N, 53°29.421'W		69°17.841'N, 53°48.548'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 – <i>more marked channel ~50 m d/s</i>		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 – <i>more marked channel ~50 m downstream</i>		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	
	April	August	April	August	April	August
2013 sampling						
SRP ($\mu\text{g L}^{-1}$)	10.5	0.7	59.8	13.8	16.3	1.9
TP ($\mu\text{g L}^{-1}$)	37.5	2.9	67.9	67.8	164.8	2.9
NO ³⁻ ($\mu\text{g L}^{-1}$)	0.01	0.04	0.05	0.03	0.02	0.04
NH ₄ ⁺ ($\mu\text{g L}^{-1}$)	BDL	3.2	BDL	11.5	BDL	BDL
Spectrophotometric Chl <i>a</i> ($\mu\text{g L}^{-1}$)	0.9	0.7	1.0	0.4	0.1	1.0 935
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 ($\text{nmol pigment L}^{-1}$)	–	0.05	–	0.63	–	0.08
Chlorophyll <i>a</i> ($\text{nmol pigment L}^{-1}$)	–	0.55	–	0.46	–	0.85
Pheophytin <i>b</i> ($\text{nmol pigment L}^{-1}$)	–	0.33	–	0.95	–	0.35
Date of collection	19/04	03/08	17/04	02/08	21/04	07/08

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

	ANOVA post-hoc Tukey p adj	Man-Whitney U p-value	Significant for both parametric and non- parametric tests?
C/N			
Core sediments vs aquatics	0.48	0.01	X
Terrestrial 'other' vs aquatics	<0.01	0.31^	X
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^	X
δ¹³C_{org}			
Core sediments vs aquatics	0.51	0.74	X
Terrestrial 'other' vs aquatics	0.15	1^	X
Terrestrial vascular plants vs aquatics	<0.001	0.46	X
Terrestrial 'other' vs core sediments	0.22	0.03	X
Terrestrial vascular plants vs core sediments	<0.001	<0.001	✓
Terrestrial vascular plants vs terrestrial 'other'	<0.001	<0.001	✓

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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 (Brassicasterol /total sterols) *1000
		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	
<i>Harrimanella hypnoides</i> (1)	43.0	39.6	18899.2	1.2	14.4	43.2	0.7	0.1	9.8	6.4
<i>Harrimanella hypnoides</i> (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
<i>Potamogeton</i> 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
<i>Chamerion latifolium</i>	33.1	16.4	129.9	0.7	26.0	27.5	0.7	0.0	29.3	3.5
<i>Salix arctica</i> (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

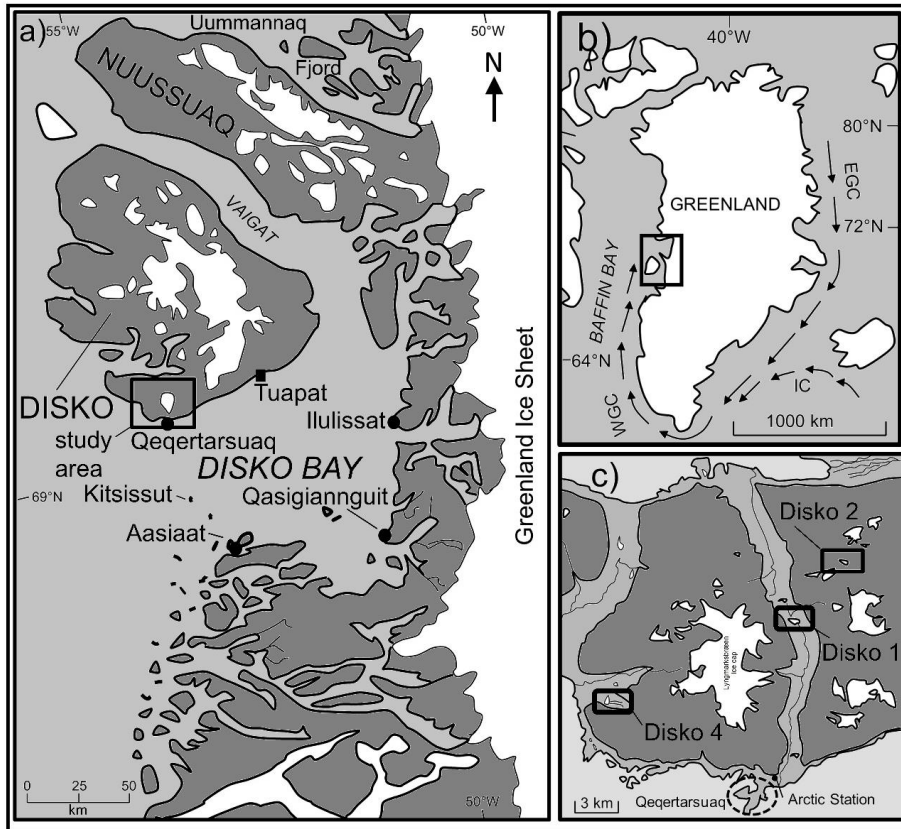


Figure 1: a) Location of Disko Island and study area (indicated) in West Greenland. b) Location of Disko Island relative to Greenland. c) Location of study catchments to the north of Qeqertarsuaq and Disko Island. See Stevenson (2017) for local catchment maps.

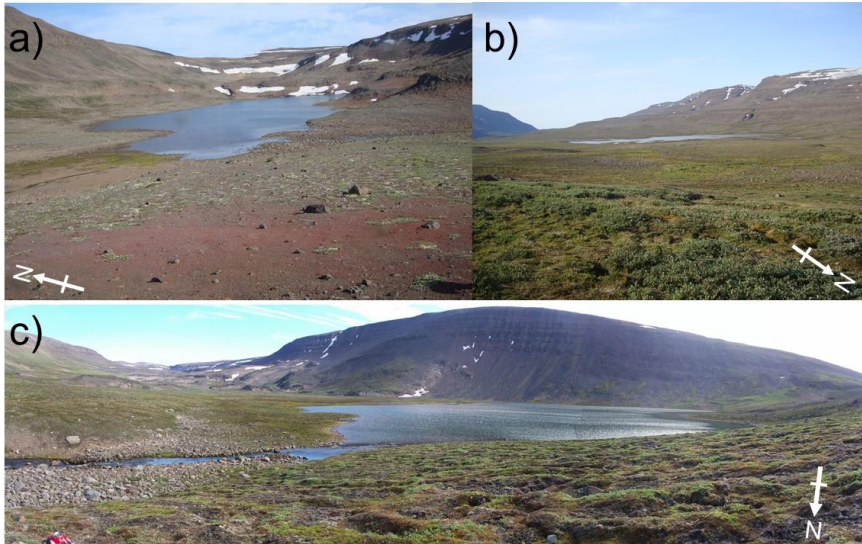


Figure 2: Images of study catchments and lakes on Disko Island in August 2013: a) Lake Disko 2 (M.Stevenson); b) Lake Disko 1 (M.Stevenson); c) Lake Disko 4 (J.Bailey).

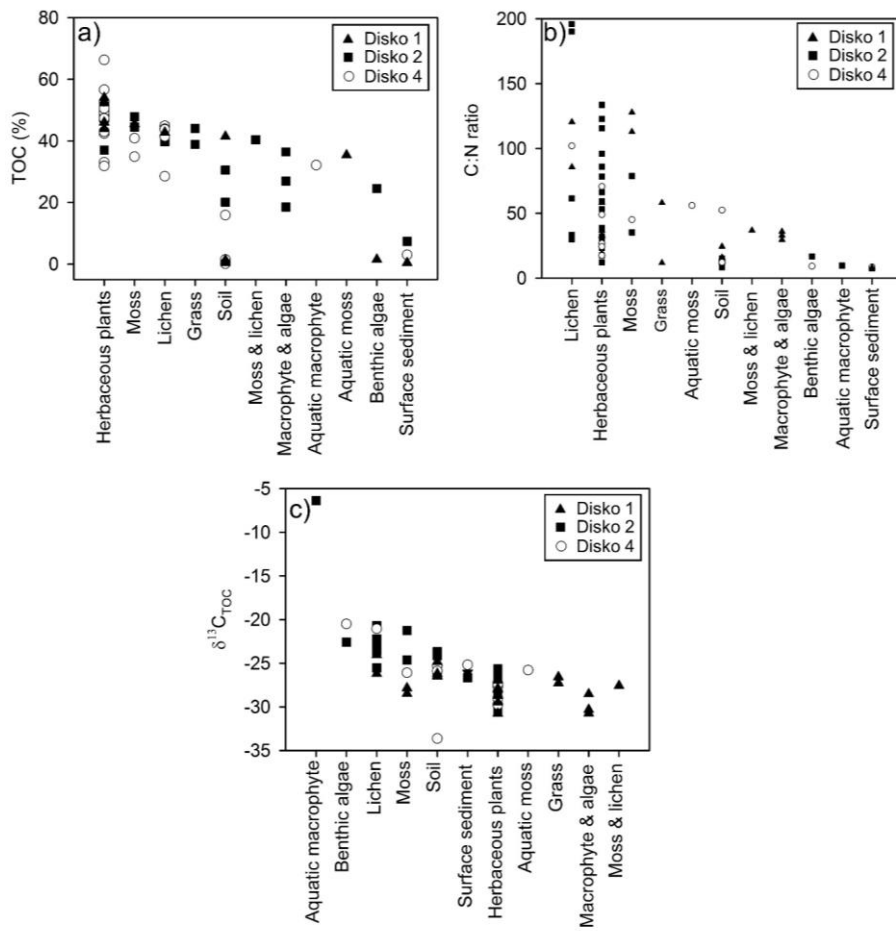
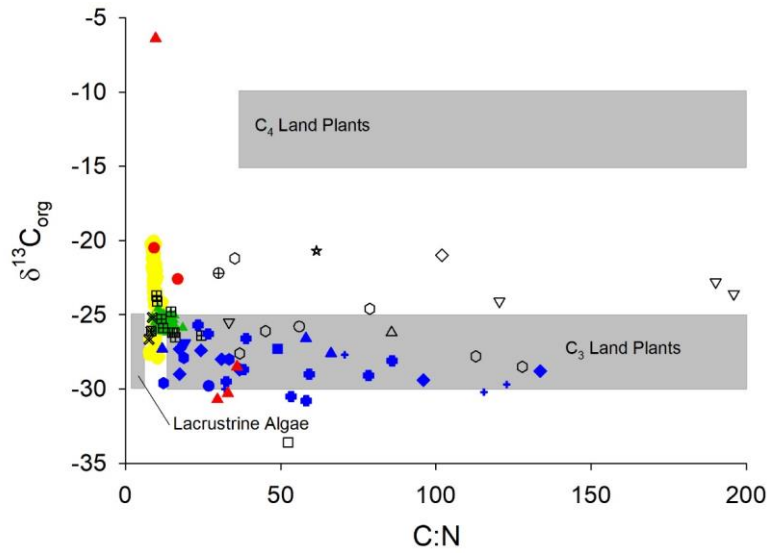


Figure 3: a) TOC (%); b) $C_{org}:N$ ratio; and c) $\delta^{13}C_{org}$ plotted against sample type, separated by lake catchment.



Key:			
Aquatics		Terrestrial vascular plants	
● Benthic algae rock scrape	● <i>Betula nana</i>	○ <i>Calliergon giganteum</i>	
▲ <i>Potamogeton</i>	● <i>Chamerion latifolium</i>	▽ <i>Cetraria nivalis</i>	
	▼ <i>Eriophorum</i>	□ Guano	
	▲ Grass	◇ Lichen	
	+ <i>Harrimanella hypnoides</i>	△ Mix of <i>Cladonia arbuscula</i> & <i>Cladonia rangiferina</i>	
	■ <i>Juniperus communis</i>	○ Moss	
	● Other Plants (unidentified)	⊞ Soil	
	◆ <i>Salix arctica</i>	⊕ <i>Stereocaulon alpinum</i>	
		× Surface sediment	
		★ <i>Umbilicaria</i> -type lichen	
Core sediments			
● Disko 2 core sediments			
▲ Disko 4 core sediments			

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).

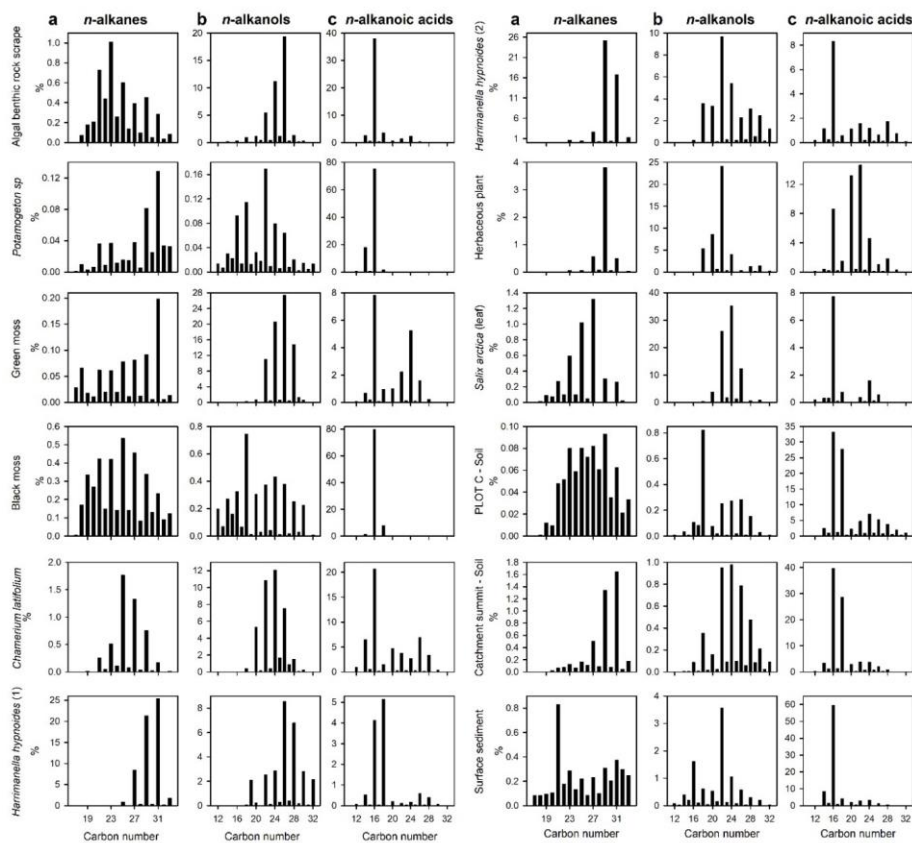


Figure 5: Lipid distribution histograms for a) *n*-alkanes, b) *n*-alkanols and c) *n*-alkanoic acids for catchment plant, soil and surface sediment samples from Disko 2 lake and catchment analysed using GC-MS. Expressed as relative proportion of total saturated *n*-alkanes, *n*-alkanols and *n*-alkanoic 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.

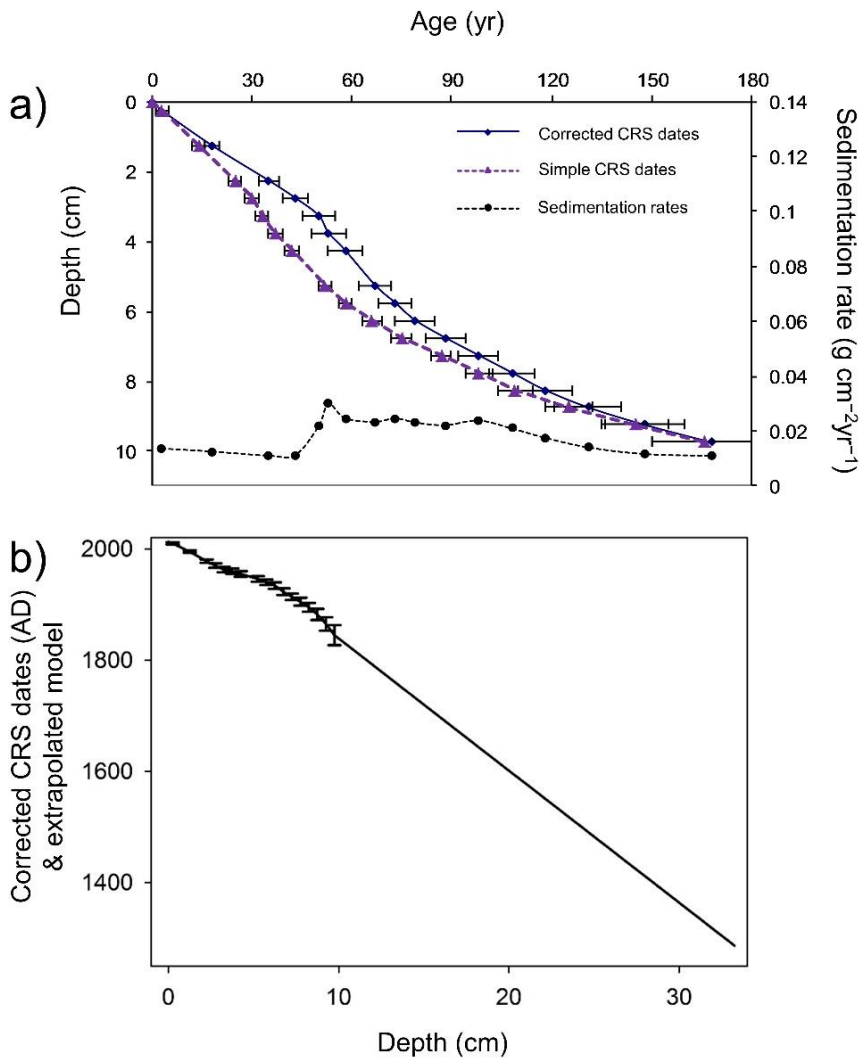


Figure 6: a) Age depth models and sedimentation rates using corrected CRS model for the Disko 2 K1 core. The simple CRS (constant rate of supply) model derived from ^{210}Pb dating only is indicated by purple triangles. The corrected CRS (constant rare of supply) model is derived from a ^{210}Pb base, 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.

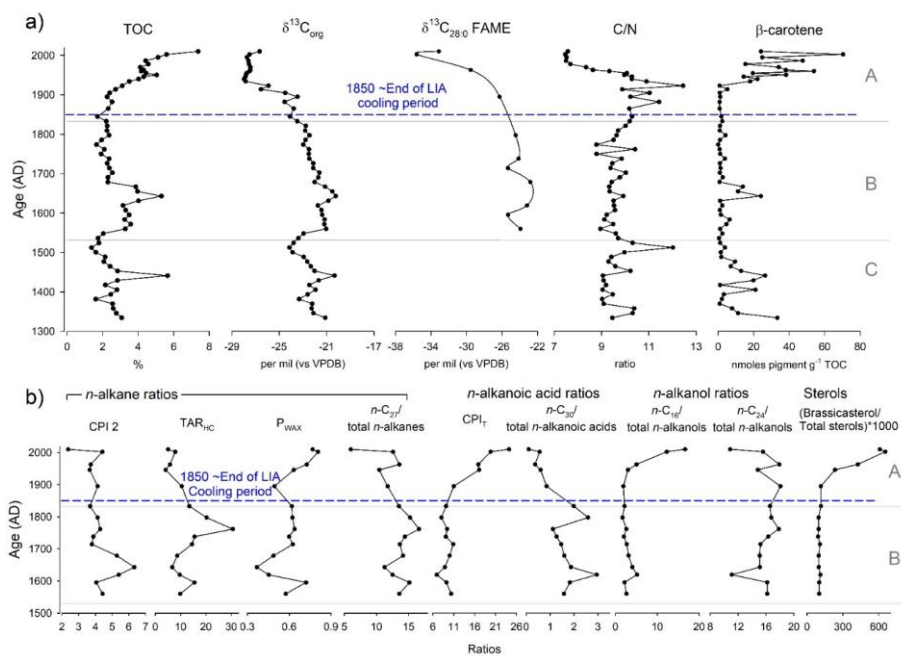


Figure 7: a) Down-core bulk and compound-specific isotopic geochemical parameters from the Disko 2 (K1) short core, presented against modelled age (AD). b) Down-core lipid geochemical ratios and equations including CPI 2, TAR_{HC}, P_{wax}, $n-C_{27}/total\ n-alkanes$, CPI_T, $n-C_{30}/total\ n-alkanoic\ acids$, $n-C_{16}/total\ n-alkanols$, $n-C_{24}/total\ n-alkanols$ and (brassicasterol/total sterols)*1000. For abbreviations see Table 4.