A landscape perspective of Holocene organic carbon cycling in coastal SW Greenland lake-catchments

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ABSTRACT

Arctic organic carbon (OC) stores are substantial and have accumulated over millennia as a function of changes in climate and terrestrial vegetation. Arctic lakes are also important components of the regional C-cycle as they are sites of OC production and CO_2 emissions but also store large amounts of OC in their sediments. This sediment OC pool is a mixture derived from terrestrial and aquatic sources, and sediment cores can therefore provide a long-term record of the changing interactions between lakes and their catchments in terms of nutrient and C transfer. Sediment carbon isotope composition (δ^{13}C), C/N ratio and organic C accumulation rates (C AR) of ^{14}C-dated cores covering the last ~10,000 years from six lakes close to Sisimiut (SW Greenland) are used to determine the extent to which OC dynamics reflect climate relative to lake or catchment characteristics. Sediment δ^{13}C ranges from −19 to −32‰ across all lakes, while C/N ratios are <8 to >20 (mean = 12), values that indicate a high proportion of the organic matter is from autochthonous production but with a variable terrestrial component. Temporal trends in δ^{13}C are variable among lakes, with neighbouring lakes showing contrasting profiles, indicative of site-specific OC processing. The response of an individual lake reflects its morphometry (which influences benthic primary production), the catchment:lake ratio, and catchment relief, lakes with steeper catchments sequester more carbon. The multi-site, landscape approach used here highlights the complex response of individual lakes to climate and catchment disturbance, but broad generalisations are possible. Regional Neoglacial cooling (from ~5000 cal yr BP) influenced the lateral transfer of terrestrial OC to lakes, with three lakes showing clear increases in OC accumulation rate. The lakes likely switched from being autotrophic (i.e. net ecosystem production > ecosystem respiration) in the early Holocene to being heterotrophic after 5000 cal yr BP as terrestrial OC transfer increased.

1. Introduction

The Arctic is a major organic carbon (OC) store (~1600 Pg) (Tarnocai et al., 2009), most of it contained in frozen soils. Changes in OC pools are a function of long-term changes in permafrost expansion and retreat and associated vegetation dynamics, although landscape processes (soil development, plant immigration) and regional differences in terrestrial productivity also determine long-term rates of OC sequestration (Oechel and Billings, 1992). Regional precipitation patterns, hydrology and physical relief also influence the movement of dissolved and particulate organic carbon (DOC, POC) across the landscape (Mulholland, 2003).

Soil development and associated nutrient depletion create autogenic terrestrial successional pathways (ontogeny) that impact C-sequestration (Engstrom et al. (2000). Terrestrial (and aquatic) ecosystems in the Arctic are mainly N-limited (Chapin and Bledsoe, 1992). Although Arctic terrestrial C stores have accumulated over millennia, their future today is uncertain, because of pronounced warming at these latitudes, which will increase C mineralization

rates as the active layer deepens and permafrost thaws (Schuur et al., 2015). The fate of this terrestrial C pool is, however, ambiguous, because although some C may be oxidised as soils become warmer, there is also considerable lateral transfer into streams, lakes and the Arctic Ocean (McClelland et al., 2016). It is widely assumed that most of the terrestrial C transferred to aquatic systems will be photo-chemically or microbially-processed and degassed as CO_2 (Cory et al., 2014). While the microbial lability of contrasting C sources varies (McCallister and del Giorgio, 2012; Vonk et al., 2013), there is also debate about the extent to which terrestrial C is more refractory than fresh algal-derived DOM (Guillemette et al., 2013). Input of terrestrial dissolved organic matter (DOM) to lakes has a number of process implications for lake functioning/lake metabolism (the auto-heterotrophic balance), supporting a microbial loop, as well as stimulating primary production, because of the nutrients (N, P) that are transferred together with the C. Terrestrial C input can also restrict aquatic production, because of its influence on in-lake light climate (Karlsson et al., 2009; Seekell et al., 2015).

The input of terrestrial C to lakes is a function of vegetation composition and terrestrial production, as well as the efficiency of the hydrological pathways/processes that move POC/DOC laterally (Mulholland, 2003). These pathways evolve as landscapes age, with climate change and as soils develop (Engstrom, 1987). Input of terrestrial C into lakes drives heterotrophy (Kling et al., 1991), and the implication is that the autotrophic/heterotrophic balance in lakes will change as the landscape ages due to soil and vegetation development (especially migration) (McGowan et al., 2016). These catchment processes also alter nutrient supply and hydrology and, in turn, aquatic ecosystem function (Fritz and Anderson, 2013).

Lakes produce and store OC, as well as accumulating terrestrial C. The organic matter that accumulates in lake sediments thus is a mixture of auto- and allochthonous sources, and partitioning them is difficult (Meyers and Ishiwatari, 1993). Aquatic C is rapidly mineralized at the sediment-water interface, but organic matter (OM) is buried even in oligotrophic lakes where total OC stocks can be > 30 kg C m^{-2} (Anderson et al., 2009; Kortelainen et al., 2004). Primary production in Arctic lakes is strongly nutrient limited and is often dominated by the benthos, where nutrient availability is greater (Vadeboncoeur et al., 2003).

Rates of primary production in Arctic terrestrial and aquatic ecosystems are assumed to be increasing today as a function of climate change (longer growing seasons) and other global environmental change processes (altered soil nutrient and microbial dynamics), but rates and pathways of hydrologic transfer are also changing due to increased precipitation and deepening active layers (Kling et al., 2000). These landscape processes controlling OC loss and accrual in both aquatic and terrestrial ecosystems operate at seasonal to centennial (millennial) timescales, and so understanding long-term ecosystem dynamics has to be derived from both process studies and palaeoecological approaches (Fritz and

Anderson, 2013; Oechel and Billings, 1992). Long-term (10^3 yr) perspectives derived from sediment records can provide a temporal perspective of how C-dynamics changed as climate and vegetation patterns evolved in the early Holocene, changes that have some relevance for vegetation-C balance in a warming world. In particular, geochemical and organic $\delta^{13}C$ can complement contemporary studies (Leng and Marshall, 2004). However, much of the work to date that has focussed on long-term changes in isotope records has been derived from forested landscapes, which limit their applicability to tundra ecosystems.

Catchment characteristics play a major role in determining how a lake will respond to disturbance and climate forcing (Anderson, 2014). Relief and hydrological linkages between lakes can influence particulate and nutrient fluxes, while lake depth/volume will affect thermal stratification and internal geochemical cycling. Here we use a geochemical-palaeoecological approach at multiple sites in a small area to evaluate the role of within lake and catchment processes relative to climate variability in influencing OC dynamics. Specifically, we analysed $\delta^{13}C$, C/N and C fluxes (coupled with $\delta^{13}C$ and C/N from modern plants and soils) from six lakes to evaluate changing OC dynamics in the coastal environment of SW Greenland during the Holocene at the regional landscape scale. In the study area, there is a gradient of lake size and retention time and contrasting vegetation cover and soil stability (Table 1). We determined how OC processing changed following deglaciation and long-term development (and dieback) of terrestrial vegetation, as well as the effect of Holocene climate change (i.e. Hypsithermal and Neoglacial cooling (Briner et al., 2016)) on C dynamics (lateral transfer and burial). The study shows the importance of considering local landscape factors when inferring climate from arctic lake sediments.

2. Methods and study sites

2.1. Study sites

The study sites are located in coastal southern-west Greenland along a NW-SE transect near the town of Sisimiut (Fig. 1), just south of the Arctic Circle. The region has a low Arctic climate (mean annual air temperature [MAT] is −2.1 °C) with discontinuous permafrost. Much of the annual precipitation (~600 mm) (Mernild et al., 2015) falls as snow, and the spring melt period dominates the seasonality of catchment hydrology. Four lakes are located within 10 km of each other, whereas SS49 is situated on a broad peninsula some 50 km to the south-east of the other lakes (Fig. 1), although the maritime influence is still pronounced here (Anderson et al., 2001). The deglaciation history of this area is well constrained, and lake age decreases eastwards (Bennike and Bjorck, 2002).

Vegetation differences among the lakes reflect the altitude of the study catchments; higher catchments are characterized by thin soils and fell-field vegetation dominated by moss-rich scrubs with

Table 1
Morphometric characteristics of the study lakes.

Lake	Latitude and Longitude	Altitude (m)	Conductivity (μS cm^{-1})	DOC[a] mg l^{-1}	Max depth (m)	Lake surface area (ha)	Catchment area (ha). Values in parentheses include sub-catchments	Area/catchment area ratio	Relief and slope
Sisi12	66.58, 53.43	200	27	nk#	7.7	1.7	111.7	66	Steep sided
Sisi14	66.58, 53.44	181	33	nk#	7.3	2.7	51.6	19	Steep sided
Sisi15 (AT5)	66.57, 53.40	130	45	2.2	3.2	7.5	182 (443)	24 (59)	Intermediate relief
AT4	66.58, 53.30	200	44	1.86	18	9	70	0.9	Steep sided
AT1	66.58, 53.24	475	30	1.24	15	8	49.7	6	Low relief
SS49	66.77, 52.67	320	27	4.8	5	20.7	242	11	Low relief

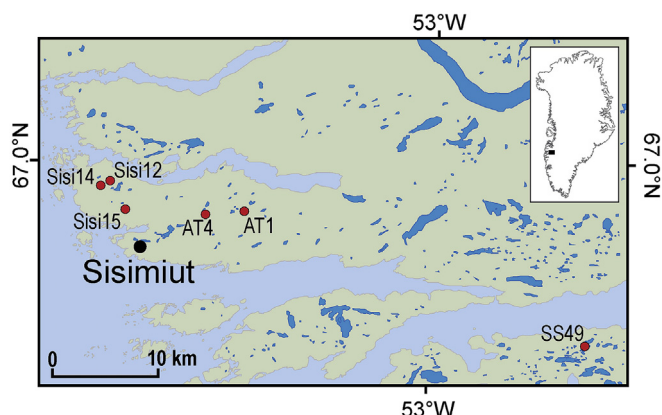[a] DOC: dissolved organic carbon #nk = not known.

Fig. 1. Location of the study sites in southern west Greenland.

snowbed communities (including *Salix herbacea*). The lower catchments have more abundant *Salix glauca*, *Betula nana*, *Ledum palustrae* and *Empetrum nigrum*; the soils are also slightly thicker and more organic rich than those at higher altitude. The vegetation history of coastal SW Greenland is different to that inland, reflecting contrasting plant immigration histories, as well as climate (Wagner and Bennike, 2012). Woody plants, such as *Empetrum nigrum*, arrived around 10.3 cal yr BP at the coast and *Vaccinium uliginosum* a little later, around 10 cal yr BP. *Salix herbacaea*, which is common in snowbed communities at the coast today, arrived in the Sisimiut area ~9.4 cal yr BP but is not abundant in macrofossil records until after 5000 cal yr BP (Bennike and Wagner, 2012).

The lakes are scour basins, small and relatively shallow (Table 1). One lake, AT4 stratifies thermally, because of its depth (~18 m). The

lakes are small (<10 ha), with the exception of SS49 (20.7 ha), and cover a short altitudinal gradient (Table 1). Sisi12 and 14 are the smallest of the lakes, and, together with Sisi15 and AT4, have catchments with considerable relief. In contrast, AT1 and SS49, which are located at >300 m, have catchments with lower relief and more limited vegetation cover. The lakes cover a gradient of catchment-lake ratio, with the associated variability in lake water retention times. The two smallest lakes, Sisi12 and Sisi14, are separated by a small pond (Wagner and Bennike, 2012) and are also likely rapidly flushed. AT4 and SS49 have longer retention times, AT4 because of its greater volume (maximum depth is ~18 m) and SS49 because of its size, low relief and smaller catchment:lake ratio (Table 1). Of the study sites, only Sisi15 is located below the regional marine limit. Additional details of the study sites can be found elsewhere (Anderson et al., 2012; Reuss et al., 2013; Wagner and Bennike, 2012). The thin soils, unproductive terrestrial vegetation, low MAT, coupled with crystalline (gneiss) bedrock, mean that the lakes are dilute and oligotrophic (water conductivity is $< 100 \, \mu S \, cm^{-1}$). DOC concentrations are $<5 \, mg \, l^{-1}$ (Table 1), with more colour as a result of a greater catchment/terrestrial source compared to inland lakes (Anderson and Stedmon, 2007).

2.2. Methods

There are few contemporary plant and soil $\delta^{13}C$ and C/N data from west Greenland. Here we analysed modern plants and soil from around Sisimiut, together with lake particulate organic matter. Sediment core data from Sisi12 presented in Leng et al. (2012) also are summarised here, together with new sediment core data from five lakes. The sediment cores were taken from the deepest part of each lake, and coring and subsample details can be found elsewhere (Perren et al., 2012; Wagner and Bennike, 2012).
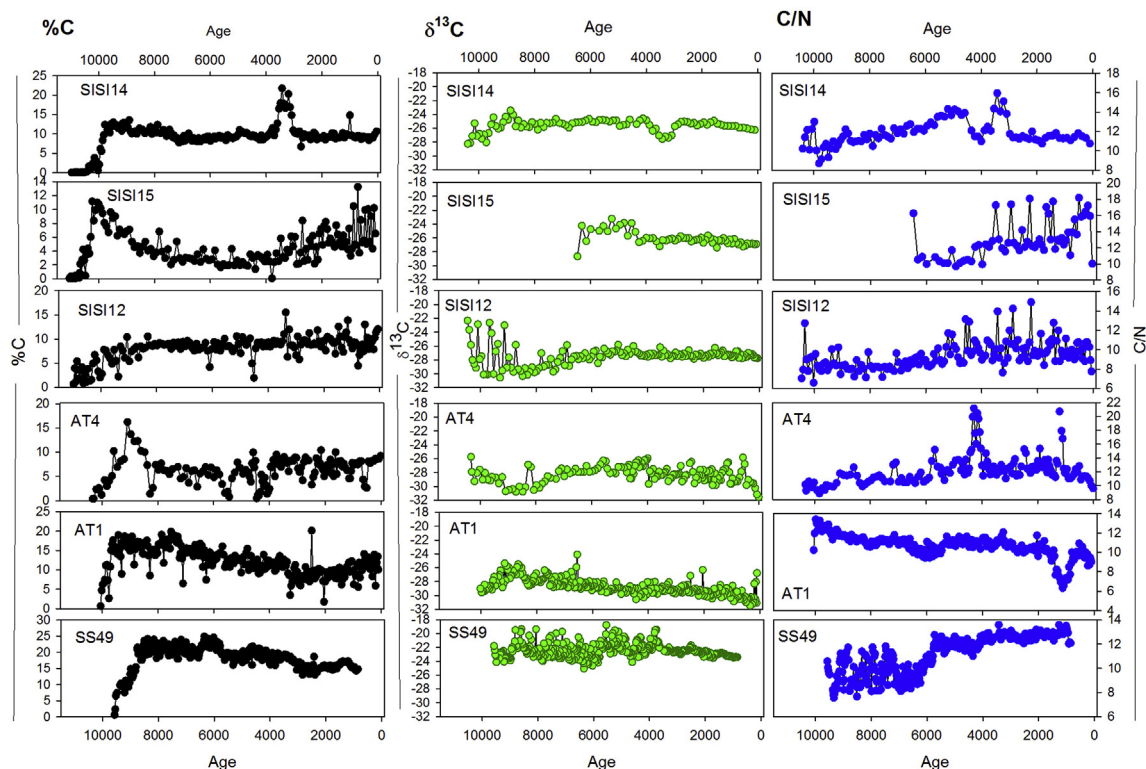

Fig. 2. Carbon content (left column), $\delta^{13}C$ (centre) and C/N (right column) versus sediment age for the study lakes; the Sisi15 $\delta^{13}C$ data are incomplete due to insufficient sample for analysis.

Macrofossil data from Sisi12, 14 and 15 which were originally presented in Wagner and Bennike (2012), are used for comparative analyses in this study. POC samples were collected in 2011 and 2012 from a number of coastal lakes (Whiteford et al., 2016). Water samples were taken with a van Dorn sampler at 1-m water depth and filtered through GFF filters and kept frozen until being freeze dried. The plants and soils were air dried after collection. For the $\delta^{13}C$ analyses, plants were rinsed in 5% HCl to remove any calcite and freezer milled to a fine powder. The soils were treated in a similar fashion to the lake sediments (see below).

The soils and lake sediment samples were treated with 5% HCl to remove calcite. The sediment was measured for %TOC and %TN concentrations (from which the C/N ratio was calculated) using a Carlo Erba 1500 elemental analyser, calibrated through an internal acetanilide standard. $^{13}C/^{12}C$ analyses were performed on the same instrument using an on-line VG Triple Trap and Optima dual-inlet isotope ratio mass spectrometer (IRMS). $\delta^{13}C$ values were calculated relative to the VPDB scale using a within-run laboratory standard (cellulose, Sigma Chemical prod. no. C-6413) calibrated against NBS-19 and NBS-22. Replicate analysis of sample material gave a precision of $\pm <0.1$‰ (1 SD). For lake POC, OC and TN concentrations and $\delta^{13}C$ values were measured on a Thermo 1112 Flash elemental analyser coupled in continuous flow to a Thermo Delta V + IRMS. The %OC and %TN values were calibrated an internal acetanilide standard, while $\delta^{13}C$ values were calculated relative to the VPDB scale using IAEA-C6, IAEA-C8, and IAEA-600 as within-run standards. Replicate analysis of standard material gave a precision of ± 0.3‰.

Dry mass accumulation rates and C content were used to calculate C accumulation rates (C AR: g C m^{-2} yr^{-1}). Where C content was not measured directly, it was estimated from the organic content (determined by %LOI) using a correction factor/ standard relationship (0.468) (Anderson et al., 2009). All cores have well constrained ^{14}C chronologies, full details of which are available elsewhere (Law et al., 2015; Perren et al., 2012; Wagner and Bennike, 2012), and cover ~11,000 cal yr. The chronology for used here differs slightly from that of Perren et al. (2012) as it includes the uppermost 10 cm unconsolidated sediment (taken with a freeze

core: see Reuss et al. (2013)) which was overlooked in the Perren study. $\delta^{13}C$ was not determined for the early Holocene sediments at Sisi15 (sample size was too small for analysis). It was not possible to calculate the C AR at SS49 as no bulk density was available. Statistical analyses (linear regression of trends in selected variables over time; Pearson correlation) and Loess smoothing (span 0.3) were undertaken in Sigmaplot (v.12). To standardise trends in C accumulation rates and β-carotene profiles (from AT1 and AT4; see Law et al., 2015) Z-scores were calculated.

3. Results

3.1. Contemporary plant and soil C isotopes

The modern terrestrial plants have a fairly consistent $\delta^{13}C$ (−27 to −29‰), but C/N ranges from ~20 to >100 (Fig. 3a). The aquatic plants (macrophytes such as *Potamogeton* sp. and aquatic mosses) have reduced C/N variability, but $\delta^{13}C$ is variable, and some samples have significantly higher values than the other materials (up to −15‰). Coastal soils are more uniform with respect to both C/N and $\delta^{13}C$. The seston (particulate OC) from the lakes appears to be predominantly autochthonous (C/N range: 8–12), and $\delta^{13}C$ ranges from −25 to −32‰. The bi-plot of both modern and sediment core data (Fig. 3c) helps distinguish the sources of organic matter that are contributing to the lake sediment samples, although degradation process during sedimentation are not accounted for. Sediment OC in SS49, for example, appear to be strongly influenced by algae and aquatic macrophytes, while sediment OC in lakes AT4 and Sisi15 appeared to have the strongest terrestrial influence.

3.2. Sediment cores

The lakes closest to Sisimiut (Sisi12,14, 15 and AT1, AT4) were formed ~10,000 to 10,500 cal yr BP by the retreat of the Greenland Ice Sheet eastwards, while SS49, located some 35 km to the southeast, is a little younger and has a basal date close to 10,000 cal yr BP (Perren et al., 2012). All cores have low C (or OM) content immediately following deglaciation (Fig. 2) and values increase steadily
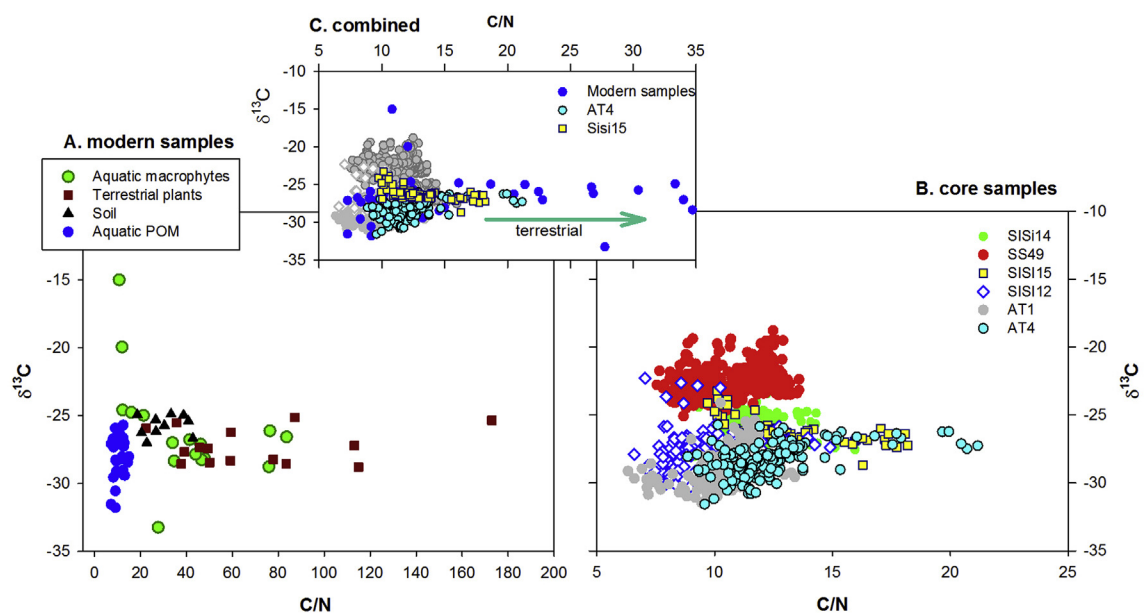

Fig. 3. Fig. 3A–C. A: Scatter plot of C/N versus $\delta^{13}C$ for contemporary soil and vegetation samples from around Sisimiut together with epilimnetic POC samples from selected lakes; B: C/N versus $\delta^{13}C$ for the six lakes; C: combined plot of both modern and core samples to show the terrestrial influence at AT4 and Sisi15 (cores highlighted in colour; all other core samples are grey).

to peak between 9000 and 6000–7000 cal yr BP, with the exception of Sisi12 where values are constant (~8%) from ~9000 until 3800 cal yr BP, followed by a series of peaks >12%. The increase in % C is matched by increasing N content. Maximum C values occur at SS49 and AT1 (15–20%) but then decline steadily up-core. At Sisi14 peak values are lower (~12%) and, from 7000 cal yr BP, constant at 9%, apart from a broad peak >20% around 3800 cal yr BP. At AT4 late Holocene values fluctuate considerably due to numerous minerogenic inwash invents (see Anderson et al. (2012) for details).

At AT1, the C/N ratio declines steadily throughout the Holocene, apart from a period of low values (~6) around 1000 cal yr BP. At Sisi14 C/N increases in the early Holocene (10–14) before dropping to 11 around 4300 cal yr BP, with a peak of 15 at 3800 cal yr BP. At AT4 and Sisi12 C/N values are variable (10–15) after 6000 cal yr BP, with peak values of 20 around 4000 cal yr BP at AT4. C/N is variable (8–12) during the early Holocene (9000–7400 cal yr BP) at SS49 but is relatively constant (~12) after 5800 cal yr BP.

Prior to 8000 cal yr BP, $\delta^{13}C$ is variable at all sites; trends exemplified by Sisi12 (−30 to −22‰) and Sisi14 (Fig. 2). In sediments younger than 8000 cal yr BP, with exception of SS49, most $\delta^{13}C$ values fall within the range −26 to −30‰. Although highly variable before 4200 cal yr BP, $\delta^{13}C$ at SS49 is more positive (range −24 to −20‰). At Sisi12, $\delta^{13}C$ is essentially constant (−27.5‰) after 4200 cal yr BP, whereas at AT4 it drops from 6000 cal yr BP but is variable (switching by up −4‰ between samples). There is, however, a significant negative trend over this time period ($\delta^{13}C = 0.0004_{AGE} − 29.173$; $r^2 = 0.27$; Fig. 2) at this site. Between sample variability is lower at AT1, but here there also is a significant negative trend from ~9000 cal yr BP ($\delta^{13}C = 0.0003_{AGE} − 30.247$; $r^2 = 0.53$). At Sisi14 a major negative excursion in $\delta^{13}C$ that lasts ~600 years is followed by a steady decline from ~1500 cal yr BP.

The C burial rates (g C m^{-2} yr^{-1}) are shown in Fig. 4. Burial rates were initially quite high 10–15 g C m^{-2} yr^{-1}) at Sisi14 and Sisi15 and then declined until ~5000 cal yr BP. The steady decline at Sisi14 was interrupted at 5000 and 3200 cal yr BP. Prior to ~6200 cal yr BP at AT1, C AR varied around 6–8 C m^{-2} yr^{-1} but then halved (see Fig. 4). At AT4 and Sisi-12, C AR were relatively constant until ~5000 cal yr BP after which mean rates approximately doubled at both sites: AT4, ~2 vs ~5 g C m^{-2} yr^{-1}) and Sisi12, ~2 vs ~3.5 g C m^{-2} yr^{-1}. The Sisi15 profile, with variable C AR after 4000 cal yr BP (Fig. 5) is, however, comparable to that observed at these sites. Increasing C AR is associated with increasing C/N ratio at most lakes (Fig. 8) and strongly so at Sisi12 and Sisi15 (R^2 = 0.86 and 0.78 respectively).

4. Discussion

The C content of lake sediment reflects the balance between autotrophic in-lake production and the input of terrestrial C. We use C/N and $\delta^{13}C$ of organic matter from lake sediments, and modern soil and plant, to infer changes in catchment vegetation and organic production in the lakes, as well as direct and indirect climate effects (i.e. catchment vegetation changes and soil degradation), and discuss the implications for the auto-heterotrophy debate in limnology.

4.1. Deglaciation and DIC dynamics

The effect of deglaciation on soil-water C dynamics is well known, but palaeolimnological records from the Late Glacial also show the effects of limited soil development and minimal vegetation cover: lake sediments deposited in the earliest stages after lake formation are highly minerogenic (clay rich). Similar records are observed in the present study. Immediately following the retreat of
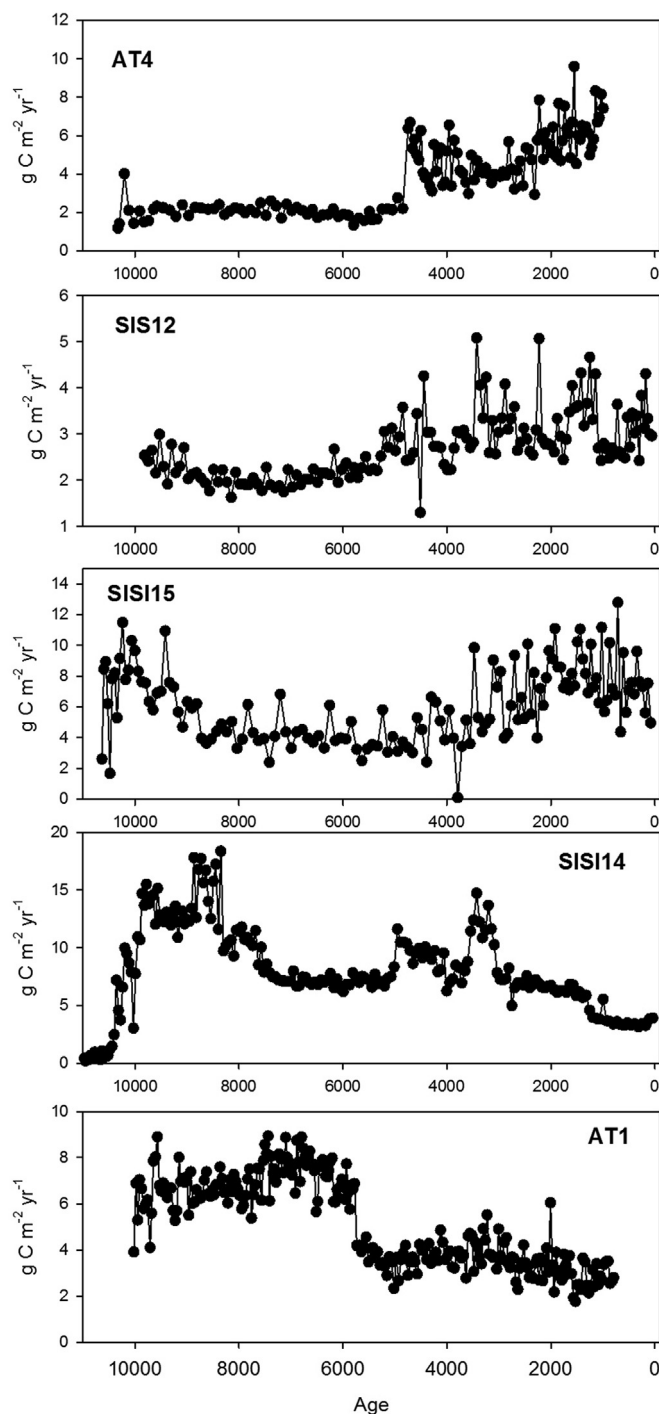

Fig. 4. C burial rates (g C m^{-2} yr^{-1}) plotted against age. Note, these burial rates are not corrected for sediment focussing.

the Greenland Ice Sheet eastwards from the study area, a scattering of glacio-fluvial and morainic debris likely covered the catchments, and vegetation development would have been minimal. Freshly comminuted sediments are rich in reactive inorganic C. Inorganic C (with high $\delta^{13}C$) would readily dissolve and form HCO$_3$ ions, base cations and inorganic nutrients in solution (Boyle et al., 2013). A steady supply of bicarbonate has been shown to occur in many post glacial sites. For example, Hammarlund (1993) showed that organic matter in non-carbonate lakes in southern Sweden has high $\delta^{13}C$
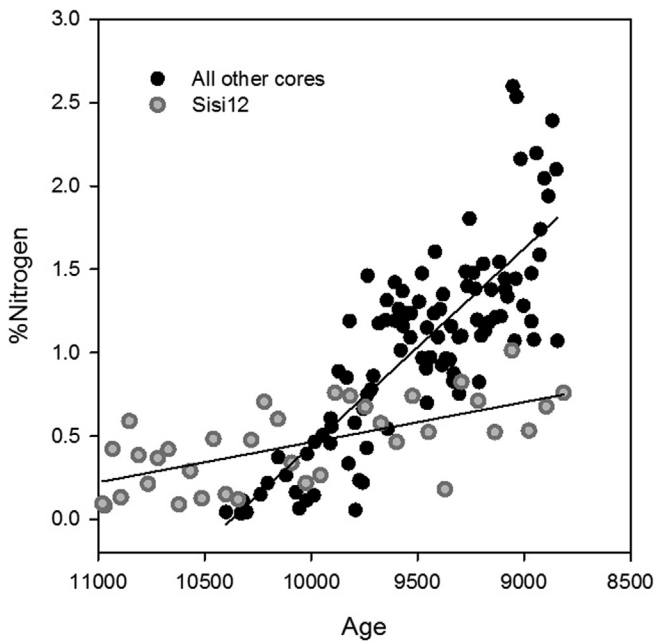
Fig. 5. The early Holocene increase in N content in lakes of the Sisimiut region; the samples from Sisi12 are differentiated because of the substantially lower rate in increase compared to the other lakes (all other cores combined). The fitted linear regressions are statistically different.

immediately after deglaciation, but enriched $\delta^{13}C$ in organic matter does not occur in lakes where carbonates precipitate. This implies that when carbonate is precipitated, the ^{13}C (from the bicarbonate ion) is consumed and is not incorporated into aquatic plants via photosynthetic pathways. The highly variable $\delta^{13}C$ profile at Sisi12 immediately following deglaciation was attributed to dissolution of silicate minerals by CO_2 rich groundwater by Leng et al. (2012). Dissolution of silicate minerals by CO_2 causes the bicarbonate to have high $\delta^{13}C$, similar to the $\delta^{13}C$ of glaciolacustrine clays exposed inland around Kangerlussaq today ($\delta^{13}C$ of $+3.2‰$; Leng unpublished data). Similarly high $\delta^{13}C$ occurs in AT4 (Fig. 2), whereas in

Sisi14 and AT1, $\delta^{13}C$ increases steadily in conjunction with increasing N and C content.

4.2. Carbon accumulation rates

Bulk C accumulation rates do not differentiate between autotrophic and terrestrial C. C/N and $\delta^{13}C$ can, theoretically, be used to indicate sources of OC (Meyers and Ishiwatari, 1993). The C burial rates observed in this study range from ~2 to >18 g C m^{-2} yr^{-1}. The above average C AR observed in the early Holocene at AT1, Sisi14 and Sisi15 are probably indicative of autotrophic production (peak rates are >15 g C m^{-2} yr^{-1} at Sisi-14). In contrast, at AT4, Sisi12 and Sisi15 maximal rates are associated with catchment instability and erosion during the late Holocene (Anderson et al., 2012; Leng et al., 2012). Anderson et al. (2009) estimated mean Holocene rates for the Kangerlussuaq region as ~5 g C m^{-2} yr^{-1} (these rates were not corrected for sediment focussing). The mean ^{210}Pb-derived C burial rate (corrected for focussing) for coastal lakes over the last 150 years is 1.5 g C m^{-2} yr^{-1} (Anderson unpublished).

The timing and extent of peak C AR values in the early Holocene vary, in contrast to the near synchronous increases in C AR associated with Neoglacial cooling at AT4, Sisi12 and Sisi15; there is limited change at AT1 in this period (Fig. 4). Sisi14 exhibits pulsed increases at this time but then stabilizes. Although it is probable that early Holocene primary production was higher than that after 5000 cal yr BP (due to Neoglacial cooling) (see discussion of β-carotene in the Synthesis section), limited emphasis should be placed on the differences in C burial rates among lakes, primarily due to sediment focussing. Sediment focussing (enhanced burial of sediment in the deeper parts of a lake) is a major constraint on quantitative comparisons among lakes (Engstrom and Rose, 2013) and requires that whole-basin estimates are made, for example, as has been made for boreal lakes (Ferland et al., 2012; Kortelainen et al., 2004). Another caveat when estimating C burial rates for the early Holocene especially (i.e. >10,000 cal yr BP), is the difficulty in dating the basal minerogenic sediments as they transition into more organic sediments; this means that the older parts of the chronologies are not well constrained (Wagner and Bennike, 2012). Therefore, early C AR rates (i.e. >10,000 cal yr BP) should be treated with caution. Moreover, element accumulation rates from lake
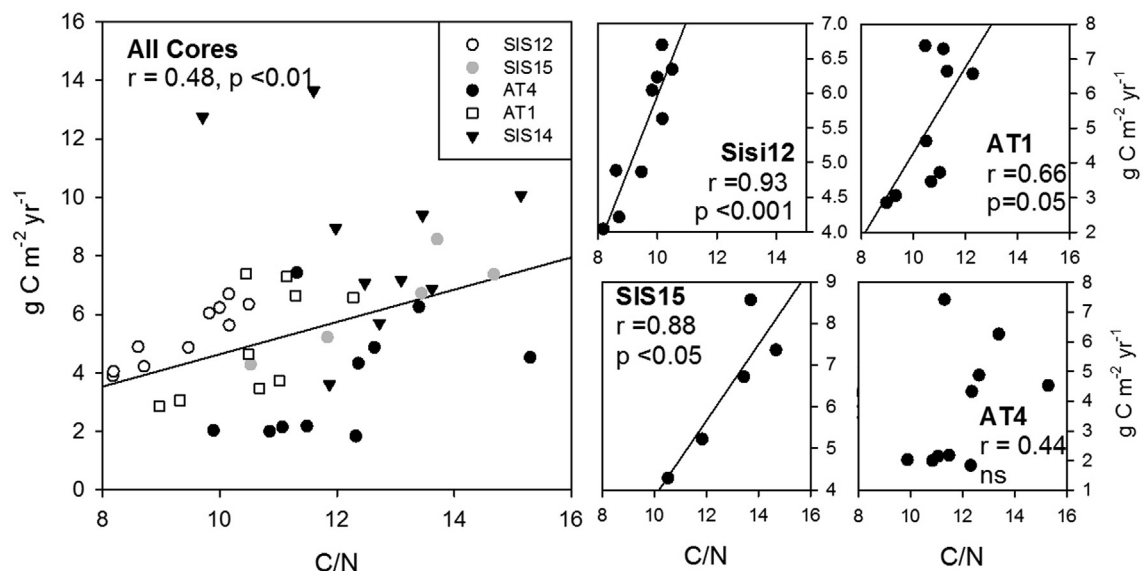

Fig. 6. C/N (A, upper plots) and C accumulation rate (B, lower plots) plotted against *Cenococcum* scletoria concentration; all data points are means of 1000-yr time bins. The fitted linear regressions lines are shown only where there is a statistically significant correlation between the two variables as indicated.
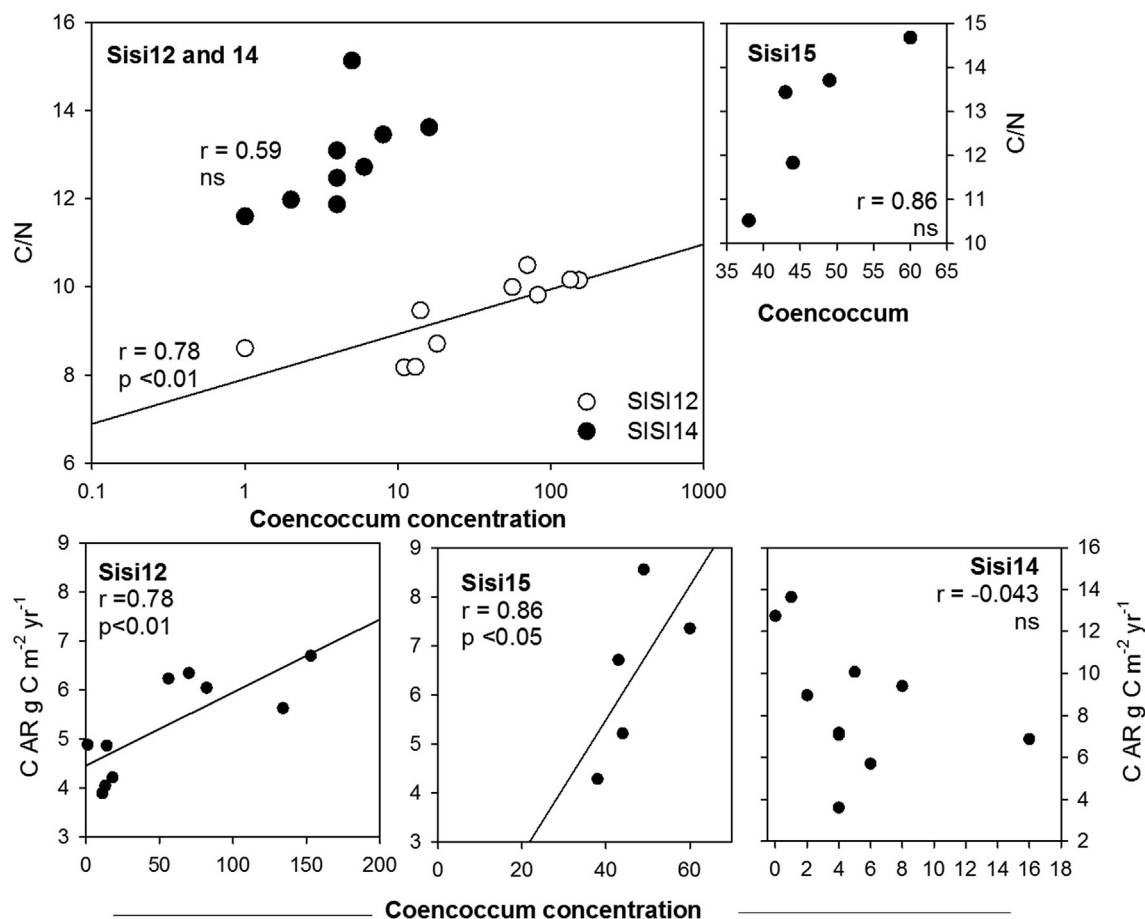
Fig. 7. The C accumulation rate plotted against the C/N ratio for selected cores; as in Fig. 6, data points are summarized as 1000-yr period means. The fitted linear regressions lines are shown only where there is a statistically significant correlation between the two variables; note that C AR increases with increasing C/N ratio at all lakes.

sediments are strongly influenced by the age-depth models chosen and hence changes should not be over interpreted. However, the age-depth models at these sites are considered relatively robust (see Wagner and Bennike, 2012; Law et al., 2015). Smaller lakes (ponds) tend to have higher primary production (per unit volume) but also probably have stronger focussing effects in the early Holocene, hence the high rates observed in Sisi14. Although Sisi14 and Sisi12 are situated close together (see Wagner and Bennike, 2012), the different C AR rates and profiles at these sites in the late Holocene are probably best explained by the difference in catchment sizes (Table 1) and the presence of three inflow streams at Sisi12. To fully evaluate catchment and lake C dynamics throughout the Holocene it is necessary to do proper whole basin estimates (*sensu* Ferland et al., 2012) and date terrestrial soil profiles.

4.3. Organic carbon sources: aquatic and terrestrial primary production

The OC content of lake sediments is a mixture of aquatic and terrestrial sources (Meyers and Ishiwatari, 1993) and separating these sources is often done using bulk composition indicators, such as C/N and δ^{13}C (Fig. 3). Modern samples (Fig. 3a) show the range of plant and coastal soils in relation to the lake samples on a C/N v δ^{13}C cross plot. The δ^{13}C values of all the core sediments range from −19 to −31‰, which is smaller than the range in plants and soil. Olsen et al. (2013) observed a similar range at two inland lakes in SW Greenland. Some of the lakes, i.e. SS49 and AT1, have contrasting C/

N and δ^{13}C values. The other sites (AT4, Sisi12, Sisi14, Sisi15) have a smaller range in δ^{13}C, mainly between −24 and −28‰. POM samples have low δ^{13}C (−25 to −28‰) and C/N ratios (ca. 5−15), values consistent with sources from recent pelagic primary production (Fig. 3a) (Kling et al., 1992; Tank et al., 2011; Zigah et al., 2011). Importantly, coastal lakes in SW Greenland are well connected hydrologically to their catchments (Osburn et al., 2017), due to the level of precipitation and generally steeper relief, resulting in greater runoff from the catchments. Yet POC appears to be predominantly autochthonous (Fig. 3a) and, as such, hydrologic connectivity is probably important for nutrient transfer (Curtis et al., 2018; Osburn et al., 2017). In oligotrophic lakes where growth rates are low, ^{12}C can be used selectively resulting in lower δ^{13}C. Uptake of respiratory CO_2 by primary producers in lakes often can result in POC values less than −28‰ (McCallister and del Giorgio, 2008; Zigah et al., 2011) while photooxidation of OM can influence the δ^{13}C value. The mean δ^{13}C of epilimnetic DOC in the coastal lakes is −27‰ (Osburn unpublished), similar to that of POC (Fig. 3a). However, the DOC concentration in coastal lakes is low (<5 mg l^{-1}) (Anderson and Stedmon, 2007) (Table 1), and so DOC flocculation is unlikely to contribute substantially to the organic load in the sediment.

The sediment deposited prior to 5800 cal yr BP at SS49 has C/N ratios <10 (mean 9.7) (as does the sediment at AT1 around 1000 cal yr BP), which is considered indicative of an algal source. Physically, SS49 lake sediment is a gelatinous algal gytta. The majority of samples from the other lakes, however, have C/N that fall in
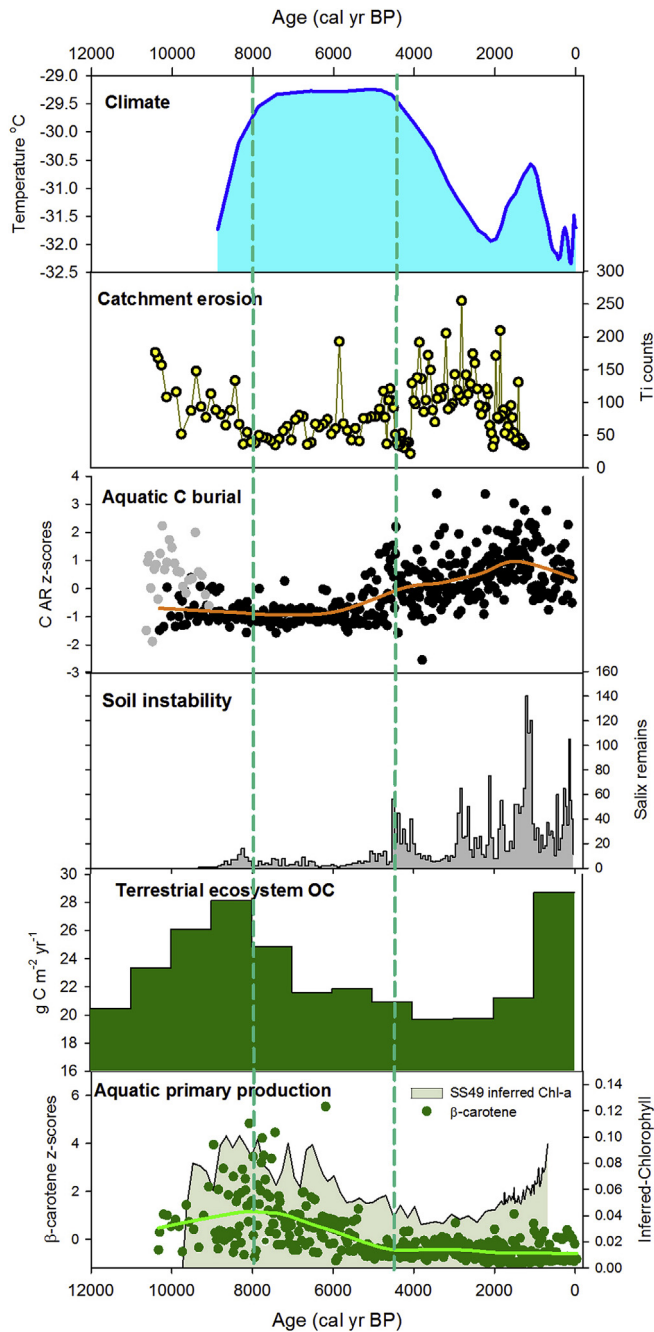
Fig. 8. A synthesis of local, regional and broader scale changes in climate and environmental conditions and their relation to carbon dynamics. Panel titles are indicative of the environmental inferences and conditions, as well as primary production as referred to in the main text. Data sources and specific proxies are listed below. Loess smoothers are fitted as described in the Methods; the samples identified by grey symbols in the Aquatic C burial plot were not used in the calculation of the Loess smoother. The green vertical dashed lines are for guide only and represent the approximate period of peak temperatures in the early Holocene and the onset of Neoglacial cooling (see Briner et al., 2016). **Climate**: temperature inferred directly from the GRIP ice core (Dahl-Jensen et al., 1998); **Catchment erosion**: Ti counts from AT4 (redrawn from data in Anderson et al., 2012); **Aquatic C burial**, data from Fig. 4 in this paper; Loess smoother is the orange line; **Soil instability**: concentration of *Salix herbacea* remains from Sisi12 (Wagner and Bennike, 2012); **Terrestrial ecosystem organic carbon**: Holocene carbon accumulation of northern peatlands (Yu et al., 2014); **Aquatic primary production**: β-carotene concentration [as z-scores with Loess smoother in green) from lakes At1 and AT4 (data from Law et al., 2015) and inferred chlorphyll a from SS49 (data from Perren et al., 2012). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

the range 10–15 (Fig. 4), which suggests a predominantly autochthonous source but with some terrestrial inputs (Meyers and Ishiwatari, 1993; Olsen et al., 2013). At Sisi12 after ~5000 cal yr BP, at AT4 around ~4200 and 1400 cal yr BP, and at Sisi14 around 3500 cal yr BP, C/N values approach 20 indicating an increasing terrestrial input (see Fig. 3c). The presence of *Coencoccum* sclerotia in the lake sediments, which is associated with soil instability and erosion, is positively correlated with C/N ratios, suggesting increasing terrestrial C inputs after ~4600 cal yr BP (Fig. 6).

At SS49, prior to 5800 cal yr BP, C/N is low, indicative of autochthonous C, and the $\delta^{13}C$ is relatively high compared to the other lakes in this study (Fig. 3b). While higher $\delta^{13}C$ values can be indicative of a predominantly bicarbonate-derived DIC source, enhanced benthic algal productivity can also lead to higher $\delta^{13}C$. Benthic algae are also known to have higher $\delta^{13}C$ due to boundary layer effects on C diffusion and fractionation (France, 1995). These data strongly suggest that autotrophy dominated at SS49 due to its small catchment:lake ratio and relatively long retention time.

While aquatic macrophytes contribute to both the autochthonous DOC and POC pools in lakes, it is unlikely that this is the dominant source in the coastal Greenlandic lakes, because of their oligotrophic nature. Macrophytes are present but in low abundance (Anderson unpublished). Similarly, planktonic primary production is low today (Whiteford et al., 2016) and, as with many oligtrophic lakes, algal benthic production probably dominates (Vadeboncoeur et al., 2003).

4.4. Catchment development and $\delta^{13}C$

After an early phase of catchment instability and relatively rapid geochemical changes ($\delta^{13}C$, C/N, Ti) following deglaciation (Figs. 2, 5 and 8), including a rapid increase in C content at four sites, the lakes and their catchments started to stabilize (see also Anderson et al., 2012). Nitrogen content increased rapidly (and at a similar rate) during the first 2000 years at most sites, with the exception of Sisi12, where the rate of N gain was much slower (Fig. 5). The importance of N availability in Arctic ecosystems is well known, and its accrual at the landscape scale and impact on aquatic ecosystems has been shown by the Glacier Bay chronosequence (Engstrom and Fritz, 2006; Engstrom et al., 2000). In Alaska, *Alnus* plays an important role in N-fixation (Shaftel et al., 2012), but fixation by terrestrial cynanobacterial mats, *Dryas* and cyanobacterial/lichen complexes is also important throughout the Arctic (Chapin and Bledsoe, 1992). In lakes, fixation by cyanobacteria is also important; *Nostoc* is widespread in SW Greenland lakes today. Nitrogen fixation by benthic cyanobacteria at SS4, a shallow lake, may also account for the high sediment N content (2.2%; 5800–9000 cal yr BP; data not shown), which is nearly twice that observed in the other cores (see Reuss et al. (2013) and Curtis et al. (2018) for a discussion of the N-pool in these coastal lakes).

There is a trend towards lower $\delta^{13}C$ in all lakes as they mature, especially during the last 2000 years. This trend is particularly pronounced at AT1 (Fig. 2) where there was a steady decline in $\delta^{13}C$ (~0.3‰/1000 yr) from ~8000 cal yr BP, showing a similar trend to that of diatom assemblage change, which is interpreted as a climate-independent process associated with landscape development (i.e. ontogeny) (Law et al., 2015). A similar decline (0.4‰/1000 yr) occurred at AT4 from 7000 cal yr BP, although at this site there was considerable short-term variability ($\delta^{13}C$ values range from −30 to −26‰), which likely reflects catchment instability superimposed on the long-term declining trend (see below). At SS49, $\delta^{13}C$ also decreases from ~5000 cal yr BP but irregularly (see Fig. 2). As the period after 8000 cal yr BP includes both Hypsithermal maximum temperatures and the pronounced cooling of the Neoglacial (Kaufman et al., 2004; Wagner and Bennike, 2012).

While these climate trends are observed across the region (Briner et al., 2016), the decline in δ13C is best considered a reflection of catchment responses to indirect climate forcing as well as onto-genetic processes rather than a direct response to climate (Fritz and Anderson, 2013).

At AT4 the low δ^{13}C (−30 to −31‰) between 9200 and 7500 cal yr BP is coincident with maximum lake production as indicated by low C/N and high β-carotene concentrations, a carot-enoid indicative of total algal production (Law et al., 2015) (Fig. 8). Diatom assemblages are dominated by benthic *Fragilaria* species, suggesting that in-lake production was primarily benthic. Both C and N peak at this time (~9000 cal yr BP) (Figs. 2 and 5), indicating that in-lake nutrient limitation was offset by the increasing N pool, if only temporarily. In contrast, at AT1 there is a long period of higher than average β-carotene from 8500 to 5400 cal yr BP and higher δ^{13}C (−26‰), highlighting the specific nature of the δ^{13}C profiles at these sites. Peak β-carotene values occurred from 8400 to 7400 cal yr BP at AT1 (Law et al., 2015) (see Fig. 8), coincident with high sediment N content and C AR >6 g C m^{-2} yr^{-1}.

Prior to ~4000 cal yr BP at SS49, δ^{13}C varies from −20 to −25‰ (Fig. 2), a period of high-inferred chlorophyll values (Fig. 8), inter-preted as high primary production (Perren et al., 2012). At this time the lake was probably autotrophic, with CO_2 drawn down due to high benthic production and limited external DOC load. Between 7000 and 9000 cal yr BP, there are, however, a number of distinct positive oscillations in δ^{13}C (close to −20‰) accompanied by large oscillations in the abundance of a small planktonic diatom (*Cyclo-tella stelligera* agg; Perren et al., 2012). The variable δ^{13}C at this time may represent differential fractionation between benthic non-siliceous algae and planktonic diatoms or variable inputs of HCO$_3$ derived from morainic debris (see below). δ^{13}C at SS49 stabilizes after 4300 cal yr BP but also declines steadily, as it does at Sisi14 from ~2000 cal yr BP (Fig. 2). While these declines may also be associated with declining lake production (i.e. oligotrophication) as observed at AT1 (Law et al., 2015), there was pronounced regional cooling in the Sisimiut area from 2000 yr cal BP. Reduced produc-tivity can lead to lower δ^{13}C or might suggest a vegetation change (the C/N in SS49 increases after 4300 cal yr BP) and alternative C inputs.

4.5. Catchment instability and soil erosion

At high latitudes, because of the sensitivity of plant commu-nities to their environment, climatic changes (both temperature and precipitation) drive vegetation responses. Increased landscape instability in response to Neoglacial cooling has been inferred both regionally and from sites cross the Davis Strait (Briner et al., 2016). In the Sisimiut lakes there is a switch in C AR around 5000 cal yr BP at 4 sites (Fig. 4). At AT4, Sisi12 and Sisi15 increased C AR from 4900 to 5000 is coincident with increasing C/N ratio (Fig. 7), suggesting that the C is partly of terrestrial original and regional indicators of catchment instability. This increase in C−flux is associated with Neoglacial cooling that resulted in vegetation die-back, increased cryonival activity and soil erosion. There is an extended period (~500 years) of instability at Sisi14, indicated by a spike in C/N and reduction in δ^{13}C around 3500 cal yr BP. These changes are also marked by enhanced burial rates (10−15 g C m^{-2} yr^{-1}), results which taken together represent unambiguous evidence of input of terriginous material. At Sisi12, δ^{13}C is essentially constant from 5200 cal yr BP (ca. −27‰) (Fig. 2), with clear indications of soil erosion from 5300 cal yr BP, as shown by both increasing C AR and *Coenococcum* and *Salix* concentration (Figs. 6 and 7) (see Leng et al., 2012). These changes are coeval with indicators of catchment disturbance at other sites (e.g. Sisi12, 15 and AT4) (Figs. 6 and 7) (see summary in Fig. 8). It is reasonable to assume that these

catchment changes caused a switch in the auto-heterotrophic balance from around ~5000 cal yr BP, i.e. the lakes became net sources of CO_2 due the increasing input of terrestrial C (Prairie, 2008).

The increase of *Salix herbacea* leaf fragments, together with that of *Cenococcum* sclerotia in the sediment macrofossil record (Fig. 8), is indicative of increased soil instability (e.g. due to die back and frost heave) and enhanced transfer of plant remains from the catchment to the lake (Heggen et al., 2010; Wagner and Bennike, 2012). In the Sisimiut region, these vegetation and soil changes are more marked in those catchments with the steepest catch-ments (e.g. Sisi12, 15 and AT4). At AT4 and Sisi12 there are large increases in minerogenic inputs associated with the increased erosion on the steep back-wall surrounding the lake (Anderson et al., 2012; Leng et al., 2012). The regional extent of landscape instability, however, is also marked by increasing C/N ratios, C AR and increased concentrations of *Cenococcum* sclerotia at Sisi15 and Sisi14 (and to a lesser extent at Sisi12) (Fig. 6). *Cenococcum* remains are also correlated with both C/N ratio and C AR (Fig. 7). Wagner and Bennike (2012) observed greater concentration of terrestrial macrofossils in the Sisimiut lake sediments (due to increasing soil instability) compared to those inland (Olsen et al., 2013; Anderson et al., 2012), which highlights the much greater hydrological con-nectivity in the coastal catchments, caused by both the higher precipitation and greater relief. The expansion of permafrost associated with Neoglacial cooling, reduced vegetation and increased cryonival activity (i.e. frost-heave in thin soils) will have increased soil instability and the lateral transfer of terrestrial POC at snow melt.

5. Synthesis

The palaeoecological records from coastal west Greenland broadly reflect the regional climate and environmental changes (Briner et al., 2016) both in terms of timing and intensity. The use of multiple sites within a small geographic area, however, highlights the role of catchment processes in determining the production and fate of carbon at the landscape scale (Anderson, 2014). Despite the differences in C dynamics at the individual lakes, as inferred from δ^{13}C, C/N and C AR (Fig. 2), it is possible to identify regional pat-terns. Specifically, the effects of Neoglacial cooling on C AR and C/N ratios are unambiguous at three sites (Figs. 2 and 8), although catchment relief also affects OM flux through its effect on erosion and hydrological transfer of terrestrial C. At the landscape scale there was retention of primarily autochthonous OC by lakes in the early Holocene due to relatively "high" aquatic primary production (as shown by β-carotene at lakes AT1 and AT4; Fig. 8) (Law et al., 2015). This enhanced aquatic primary production reflects the greater nutrient availability at this time. The high phosphorus content of fresh mineral soils has been observed in a range of en-vironments (Boyle et al., 2013), but primary production would also have required the establishment of an N-pool (Engstrom and Fritz, 2006; Engstrom et al., 2000), which in the Sisimiut catchments increased steadily after deglaciation (Fig. 5). At the same time that there was a probably a build-up of OM in catchment soils and terrestrial vegetation (driven by increasing nutrient availability and a warming climate). This accumulation of terrestrial OM at the landscape scale in the early Holocene is apparent in circumpolar peat records of C burial (Fig. 8) which mirrors the greater aquatic primary production at this time. Interestingly, C accumulation rates are comparable in both lake sediment and peat (<20 g C m^{-2} yr^{-1}).

With Neoglacial cooling from ~4000 cal yr BP there was a reduction in terrestrial peat accumulation and aquatic primary production (compare the peat and −carotene records in Fig. 8) and a switch to more lateral transfer of C at the landscape scale. The

latter is apparent at a number of the lake cores in the study area (see Fig. 8; synthesized as C-burial z-scores in Fig. 8) with an un-ambiguous increase in C burial as stresses on terrestrial plant communities increased. Regional cooling would have destabilized the thin soils as cryonival processes increased causing a dieback in terrestrial vegetation (see Wagner and Bennike (2012) and Heggen et al. (2010) for a discussion of these processes). This instability in terrestrial soils and vegetation is shown by increased accumulation of *Salix herbacea* at Sisi12 (Fig. 8) and *Cenococcum* (Fig. 7) remains in lake sediments across the region (Wagner and Bennike, 2012). Some of this eroded terrestrial OC would have been retained by lakes, as evidenced by the increased C-burial rates (Figs. 4 and 8) but given the rapid hydrological flushing of these catchments, there were probably greater losses (to the ocean). Instability and element transfer in the late-Holocene is also shown by Ti records from AT4 (Fig. 8) (see Anderson et al. (2012) for a discussion of soil erosion in this area).

As is clear from the discussion above, climate change, notably cooling, clearly affected C dynamics. But as well as temperature effects, altered precipitation regimes and catchment relief will have influenced landscape C transfer. A combination of higher precipi-tation in this region is responsible for the distinct increase in C AR and C/N from ~5000 cal yr BP, which is not discernible at inland sites where runoff is very limited (Olsen et al., 2013; Anderson et al., 2012). Snow pack thickness and melt are also more pronounced at the coast (see Curtis et al. (2018) for a discussion of snow melt in this area). C profiles from Arctic lake sediments are interpreted as reflecting regional temperature (see for example Briner et al. (2016) and references there-in) but this landscape scale assessment in-dicates that this is simplistic and highlights the need for some circumspection.

Determining the controls on $\delta^{13}C$ in bulk organic matter is complex (eg. source values overlap: see Fig. 3) and there is not a typical signal in the six lake sediment trends beyond a tendency towards lower $\delta^{13}C$ in the late Holocene. These results highlight the long-term development of, and interplay among terrestrial and aquatic C sources as well as the fluctuating balance between auto-vs heterotrophy at the landscape scale. Care should, therefore, be taken when interpreting C profiles from a single site as records of past climate.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.quascirev.2018.09.006.

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