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Assessment of branched GDGTs as temperature proxies in sedimentary records from several small lakes in southwestern Greenland

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ABSTRACT

This study of five small (<3.0 ha) lakes in southwestern Greenland examines the veracity of branched glycerol dialkyl glycerol tetraethers (br GDGTs) as a temperature proxy in lacustrine systems. The proximity (<5 km) of the lakes suggests that their temperature history, and thus their br GDGT records, should be similar. Distributions of br GDGTs in (i) surface sediments from all five lakes, (ii) ^{14}C-dated sediment cores from two lakes (Upper and Lower EVV Lakes) and (iii) soil samples from the area surrounding the lakes were examined. The temporal records of br GDGT-based temperature for the two cores exhibited both similarities and major discrepancies. The differences between the paleotemperature records for the two lakes suggest that br GDGTs are not solely soil-derived, reflecting air temperature, but also indicate an additional br GDGT contribution from another source. Among the broader suite of lake sediments, there was a strong correlation (R^2 0.987) between br GDGT-based surface sediment temperatures and measured summer bottom water temperatures for the four lakes with hypoxic/anoxic bottom waters, including Upper EVV Lake. The correlation suggests production of br GDGTs by anaerobic bacteria within the bottom water and/or sediment–water interface, reflecting environmental temperature for the individual lakes and augmenting the uniform, soil-derived signal. Hence, assessment of br GDGTs in Greenland lake sediments provides evidence for their origin from anaerobic autochthonous bacteria and indicates that interpretation of lacustrine br GDGT-based paleotemperature records requires contextual knowledge of individual lake systems and potential source(s) of sedimentary br GDGTs.

1. Introduction

Molecular paleotemperature proxies (e.g. Brassell et al., 1986; Schouten et al., 2002; Rampen et al., 2012) have proven their utility in the assessment and interpretation of ancient climates and in the quantification of rate of climate change. The data provided by these proxies play a vital role in helping constrain and validate climate models, including those employed in predicting the magnitude of future anthropogenically induced climate change (IPCC, 2013). Proxies based on the distributions of glycerol dialkyl glycerol tetraethers (GDGTs) have acquired a significant role as tools for paleoclimate studies spanning a wide range of environments and time intervals (reviewed by Schouten et al., 2013) since the initial proposal (Schouten et al., 2002). The relationship between GDGTs and temperature has been demonstrated in investigations of both marine (e.g. Schouten et al., 2002; Kim et al., 2008; Zhou et al., 2014) and lacustrine sediments (e.g. Powers et al., 2004; Blaga et al., 2009; Günther et al., 2014), soils (e.g. Weijers et al., 2006b) and peats (e.g. Weijers et al., 2011), loess (e.g. Peterse et al., 2011; Huguet et al., 2012), stalagmites (e.g. Yang et al., 2011; Blyth and Schouten, 2013) and hot springs (e.g. Schouten et al., 2007). The utility of GDGT-based proxies for paleotemperature reconstruction in both marine and lacustrine environments (Schouten et al., 2013) provides a rare opportunity to extend and expand the range and scope of molecular paleotemperature records across oceanic and continental realms.

GDGTs occur as two structural types with either isoprenoid (iso GDGT) or branched (br GDGT; also non-isoprenoid) lipids forming their core alkyl groups (Sinninghe Damsté et al., 2000; Schouten et al., 2013) with individual constituents differing in terms of: (i) number and position of cyclopentyl rings (both iso and br

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GDGTs), (ii) presence of a cyclohexyl ring (in crenarchaeol, an iso GDGT) and (iii) number and position of Me substituents (br GDGTs; specifically methylation at C-6 rather than C-5; De Jonge et al., 2014). Iso GDGTs are membrane lipids produced by the major phylogenetic lineages within the Archaea, including Thaumarchaeota, Crenarchaeota and Euryarchaeota, with the exception of halophilic Archaea (reviewed by Schouten et al., 2007, 2013). By contrast, the *sn*1,2 stereochemistry of br GDGTs suggests that they represent bacterial membrane lipids (Weijers et al., 2006a) although the specific bacteria that biosynthesize them, thought to be Acidobacteria (Weijers et al., 2009; Sinninghe Damsté et al., 2011), have to be confirmed via isolation.

Empirical evidence based on numerous analyses of sediment and soil samples has shown that the relative br GDGT distributions for a given sample are temperature dependent (e.g. Weijers et al., 2007a). Br GDGT distributions vary in the extent of methylation and cyclization, which is defined as the MBT (methylation index of branched tetraethers) and CBT (cyclisation ratio of branched tetraethers) indices, respectively (Weijers et al., 2007a). Both indices were initially defined for soils (Weijers et al., 2007a). Assessment of environmental influences on these br GDGT indices reveals that soil pH rather than temperature represents the primary influence on the CBT index, whereas MBT correlates with annual mean air temperature (MAT) and, to a lesser extent, correlates negatively with pH (Weijers et al., 2007a). The CBT index can therefore help correct for the influence of pH and facilitates the use of the combined MBT/CBT index as a measure of temperature (Weijers et al., 2007a). Independent evidence suggests that other environmental parameters like nutrient concentration and water depth exert only a minor influence on the br GDGT indices (Loomis et al., 2014), although a correlation between br GDGTs and alkalinity has been observed for a series of lakes in Minnesota and Iowa (Schoon et al., 2013). Variation in the sedimentary distributions of br GDGTs has been applied in assessment of paleoclimate for both marine (e.g. Weijers et al., 2007b; Schouten et al., 2008) and lacustrine systems (e.g. Bechtel et al., 2010; Tyler et al., 2010; Shanahan et al., 2013). Development of br GDGT-based proxies was initially based on the key assumption that these compounds were derived solely from soil/peat (Weijers et al., 2007a). However, subsequent studies suggest that autochthonous production may occur in lakes (Sinninghe Damsté et al., 2009; Tierney and Russell, 2009; Bechtel et al., 2010; Tierney et al., 2012; Schoon et al., 2013; Naeher et al., 2014; Buckles et al., 2014) and rivers (Zell et al., 2013a,b; De Jonge et al., 2014), an interpretation supported by differences in br GDGT distributions between soils and associated marine surface sediments (Peterse et al., 2009; Weijers et al., 2014). In addition, it appears that br GDGT-reconstructed temperature values consistently exhibit an unexplained "cold bias" when a soil calibration is used (Tierney and Russell, 2009; Tyler et al., 2010; Zink et al., 2010; Tierney et al., 2010b; Pearson et al., 2011). The veracity of sedimentary br GDGTs as temperature proxies is therefore predicated on evidence reflecting both the source(s) of these compounds and any potential "cold bias", which prompts investigation of sedimentary records that can help resolve these issues.

This study of periglacial lakes in western Greenland focuses on br GDGTs in sediments from small lakes and associated nearby soils collected from a valley close to the terminus of the Russell Glacier in southwestern Greenland near (ca. 16.5 km) Kangerlussuaq (Fig. 1). Our objectives encompassed (i) assessment of the veracity of br GDGTs as a temperature proxy in this environment and (ii) production of a br GDGT-based paleotemperature record from lacustrine sediments close to the terminus of Russell Glacier. The first aim, focused on evaluation of possible environmental factors that might influence lacustrine br GDGT records through examination of their distributions in sediments from multiple lakes with differing water chemistry, yet located in close proximity within a single valley, and therefore directly comparable in terms of temperature history. Inclusion of br GDGT analysis of surrounding soils as an integral component of the study also provided an opportunity to compare and contrast likely contributions of br GDGTs from allochthonous sources with the possibility of additional, autochthonous contributions derived from each lake. The second aim addressed the critical need for climate records for Greenland that could provide evidence of past change and rate of change as a historical background to the rapid temperature rise now occurring in the Arctic (IPCC, 2013). Specifically, determination of paleotemperature values for lakes close (<7 km) to Russell Glacier should complement alkenone paleotemperature records from lakes close to the coast near Kangerlussuaq (D'Andrea et al., 2011) and evidence of past climates for other regions of the Arctic obtained using either alkenones in lacustrine sediments from West Spitsbergen, Svalbard (D'Andrea et al., 2012) or based on br GDGT distributions in lake sediments from Baffin Island, Canada (Shanahan et al., 2013).

2. Material and methods

2.1. Study site

The lakes are within a narrow valley (67°05′N, 50°20′W) extending southwest from the terminus of the Russell Glacier to the Søndre Strømfjord near Kangerlussuaq (67°00′N, 50°40′W) in southwestern Greenland. Five of the small lakes in the valley, informally known as Upper Epidote Vein Valley (EVV) Lake, Lower EVV Lake, Teardrop Lake, Potentilla Lake and South Twin Lake (Fig. 1) represent the primary focus of the investigation, complemented by analysis of surrounding soils. The study region has thick, perennial, continuous permafrost < 500 m thick (Jorgensen and Andreasen, 2007) with a seasonally active layer that extends < 50 cm below the ground surface (Cadieux et al., unpublished results). The region is characterized by a Low Arctic continental climate. As a result, there is minimal thoroughgoing drainage and no current surface connection between any of the lakes. The lakes' close proximity suggests that the climate of each, both now and in the past, is likely similar, and perhaps broadly comparable with that of Kangerlussuaq, where current mean July temperature averages ca. 10.5 °C (Weidick et al., 1992; Forman et al., 2007). The lakes range in depth (4–8 m) and surface area (0.2–3.0 ha). They are all dimictic and holomictic, developing thermal stratification both in summer under open water conditions and in winter under ca. 2 m thick ice cover. A separate study focused on the aqueous chemistry in the lakes demonstrated marked variation in O_2 dynamics, ionic concentration and dissolved organic carbon (OC) from lake-to-lake under open water conditions in 2013 (Cadieux et al., unpublished results). In particular, measurements taken in June and July 2013 revealed that the hypolimnetic water in Upper EVV, Potentilla and South Twin lakes was anoxic, whereas that in Teardrop Lake was hypoxic, and Lower EVV Lake remained well saturated with O_2 during this period (Cadieux et al., unpublished results). Additionally, lake bottom water pH ranged from 6.8 to 9.0 during this same period (Cadieux et al., unpublished results).

2.2. Samples

Sediment cores from Upper EVV (38 cm; 19 samples) and Lower EVV (24 cm; 12 samples) lakes were collected using a push corer in April 2012 when the lakes were covered by 2 m of ice. Each core was sub-sectioned into samples spanning 2 cm intervals at Kangerlussuaq prior to shipment and the samples were stored frozen on arrival at Bloomington. Similar procedures were employed in April 2013 for collection, shipment and storage of individual

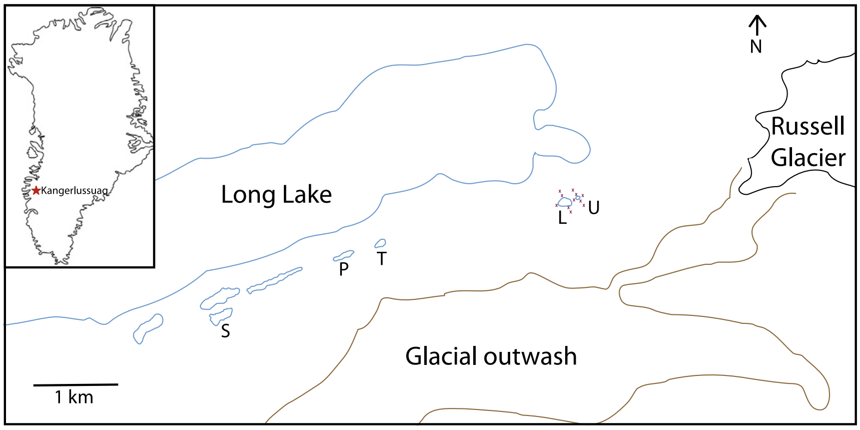

Fig. 1. Location of study area close to the terminus of Russell Glacier East of Kangerlussuaq (67°00′N, 50°40′W). Sediments were recovered from five lakes (U, Upper EVV Lake; L, Lower EVV Lake; T, Teardrop Lake; P, Potentilla Lake; S, South Twin Lake), with soil samples (purple X) from several sites surrounding U and L within the valley.

surface sediments (0–2 cm; 1 sample per lake) from Teardrop, Potentilla and South Twin lakes.

Individual soil samples (11) were collected from the region surrounding Upper EVV Lake and Lower EVV Lake in July 2013. Samples were taken from the top 10 cm of soil below the base of living vegetation at sites ranging from 3 to 78 m from the shorelines (Fig. 1). All samples were stored frozen prior to analysis.

2.3. Radiocarbon dating of cores

Subsamples of five sections from the Upper EVV Lake core and four from the Lower EVV Lake core were selected for ^{14}C dating to enable development of age models for sedimentation rate. Each subsample represented a composite of a specific 2 cm interval chosen from the respective cores. Preparation of CO_2 for analysis was performed according to standard methods in routine use (e.g. Unkel et al., 2014). Radiocarbon dating of the total OC (TOC) was performed at the National Ocean Sciences Accelerator Mass Spectrometry Facility (NOSAMS) following established procedures (Karlen et al., 1964; Olsson, 1970; Stuiver and Polach, 1977; Stuiver, 1980).

2.4. Bulk geochemical analysis

TOC content of lake and soil samples (1–13 mg) was measured using an elemental analyzer (EA) after treatment with 1 N HCl to remove inorganic carbon according to standard laboratory methods in routine use (e.g. Finkelstein et al., 2005).

2.5. Lipid analysis

Samples (1.5–6.0 g) were freeze-dried and extracted ultrasonically according to established procedures (Hopmans et al., 2000). Each total lipid extract (TLE) was separated into 3 fractions using Al_2O_3 column chromatography (modified from D'Anjou et al., 2013), eluting successively with hexane/CH_2Cl_2 (9:1, v/v – F1, apolar), hexane/CH_2Cl_2 (1:1, v/v – F2, ketones) and CH_2Cl_2/MeOH (1:1, v/v – F3, polar).

The F3 fraction, containing GDGTs, was dried under a stream of N_2. A small amount of hexane/isopropanol (99:1, v/v) was added and the solution filtered using a 0.45 μm Teflon syringe filter prior to analysis using high performance liquid chromatography–mass spectrometry (HPLC–MS) to identify and quantify GDGTs (Weijers et al., 2007a; D'Anjou et al., 2013). HPLC–MS analysis was performed at the University of Massachusetts Biogeochemistry Laboratory.

2.6. Calculation of GDGT-based proxies

Temperature values were calculated using the global calibration based on the fractional abundances of specific br GDGTs (Pearson et al., 2011) designated Ib (cyclopentyltrimethyl GDGT), II (pentamethyl GDGT) and III (hexamethyl GDGT) as shown in Eq. 1.

$$\begin{aligned}\mathrm{Temp}\ (^{\circ}\mathrm{C}) = 20.9 + (98.1 \times \mathrm{GDGT-Ib}) - (12.0 \times \mathrm{GDGT-II})\\ - (20.5 \times \mathrm{GDGT-III})\end{aligned} \tag{1}$$

This calibration for summer temperature (Pearson et al., 2011) includes numerous high latitude samples, unlike other regional br GDGT temperature calibrations specifically for lakes (Zink et al., 2010; Tierney et al., 2010b; Sun et al., 2011) and may therefore provide an appropriate calibration for the temperature range known for Greenland lakes.

MBT (Eq. 2) and the CBT (Eq. 3) were also calculated from the relative proportions of specific br GDGTs (Weijers et al., 2007a). These indices are described by Eqs. 2 and 3, respectively, expressed in terms of standard designations for br GDGTs (Weijers et al., 2007a). The two indices, when combined, provide an assessment of MAT based on empirical data (Eq. 4; Weijers et al., 2007a), although this soil-based calibration has proven problematic in its application to lacustrine sediments. Hence, the Pearson et al. (2011) calibration, as noted above, was deemed more applicable for high latitude lacustrine sediments.

$$\mathrm{MBT} = (\mathrm{I+Ib+Ic})/[(\mathrm{I+Ib+Ic}) + (\mathrm{II+IIb+IIc}) + (\mathrm{III+IIIb+IIIc})] \tag{2}$$

$$\mathrm{CBT} = -\log[(\mathrm{Ib+IIb})/(\mathrm{I+II})] \tag{3}$$

$$\mathrm{MBT} = 0.122 + (0.187 \times \mathrm{CBT}) + (0.020 \times \mathrm{MAT}) \tag{4}$$

3. Results

3.1. GDGT distributions

A full suite of br GDGTs was present in all samples, whereas individual iso GDGTs were rarely detected. Consequently, we focused on the sources of br GDGTs in our periglacial lakes of southwestern Greenland and the ability of their sediments to provide records of past climate.

3.2. Soil samples

3.2.1. Br GDGT distributions and concentrations

The distributions in the 11 soil samples from the area surrounding Lower EVV and Upper EVV lakes yielded a wide range of MBT (0.08–0.37) and CBT (0.49–1.67) values. The median MBT value was 0.14 (mean 0.15) and CBT value was 0.84 (mean 0.91; Fig. 2). The CBT-based soil pH values (Weijers et al., 2007a) correlated with the measured pH values (R^2 0.752).

The samples yielded br GDGT concentration values ranging from 0.03–1.45 μg/g dry sediment extracted and 1.38–26.53 μg/g TOC (Fig. 5).

3.3. Sediment cores

3.3.1. Age models

Visual inspection of the cores at the times of collection from both Lower EVV Lake and Upper EVV Lake revealed gradational changes in color and grain size, with no obvious sedimentological hiatuses, which suggested that the records were relatively continuous. Radiocarbon dating provided supporting evidence for continuous sedimentation throughout the time intervals represented by the cores, corresponding to an average sedimentation rate for Lower EVV Lake and Upper EVV Lake of 11 cm/kyr and 17 cm/kyr, respectively. Linear interpolation between the datum points enabled estimation of the approximate age of all other core samples. Thus, each 2 cm sampling interval from Lower EVV Lake and Upper EVV Lake sediments reflected sediment accumulation over an average of 190 ± 25 yr and 120 ± 25 yr, respectively (Table 1, Fig. 3) and the entire cores from the two lakes represented time intervals of 2380 ± 25 yr and 2580 ± 25 yr relative to 1950, respectively.

3.3.2. Br GDGT distributions and resulting paleotemperature records

For Lower Lake, the down core MBT and CBT ranges were 0.13–0.18 and 0.06–0.74, respectively (Fig. 4). For all Lower Lake samples, br GDGT III was the most abundant, followed by II then I, with minimal input from br GDGTs Ib, Ic, IIb, IIc, IIIb and IIIc (Fig. S1). For Upper Lake, the down core MBT and CBT ranges were 0.12–0.24 and 1.06–1.34, respectively (Fig. 4). In the Upper Lake core, br GDGT II and III alternated as the most abundant, followed by br GDGT I. As for Lower Lake, br GDGTs Ib, Ic, IIb, IIc, IIIb and IIIc were present in minimal abundance.

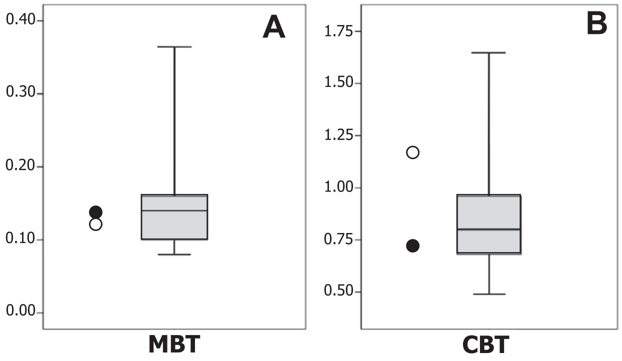

Fig. 2. MBT (A) and CBT (B) values for 11 soil samples surrounding Upper EVV Lake and Lower EVV Lake derived from br GDGT distributions defined by Eqs. 2 and 3, respectively. The data are shown as an overall range with boxes delineating the 25 and 75 percentiles and the horizontal line marking the median. Surface sediment values from Upper Lake (open circles) and Lower Lake (closed circles) are shown for comparison.

Table 1
Radiocarbon results.[a]

	Accession number	Yr BP relative to 1950	Age error (yr)
Lower EVV Lake			
0–2 cm	OS-106278	135	25
8–10 cm	OS-106279	685	25
14–16 cm	OS-106280	1220	30
22–24 cm	OS-106611	2380	25
Upper EVV Lake			
0–2 cm	OS-106438	285	25
8–10 cm	OS-106277	805	30
18–20 cm	OS-106439	1280	25
26–28 cm	OS-106440	2080	25
36–38 cm	OS-106441	2580	20
Teardrop Lake			
0–2 cm	OS-106276	315	25
Potentilla Lake			
0–2 cm	OS-106275	260	25
South Twin Lake			
0–2 cm	OS-106274	150	25

[a] All radiocarbon measurements were performed at the National Ocean Sciences AMS (NOSAMS) facility.

The Lower EVV Lake sediment record (2380 ± 25 yr) yielded temperature values ranging from 8.8–13.0 °C, with a mean of 9.9 °C based on the global summer temperature calibration of Pearson et al. (2011). The overall average error based on results for duplicate samples was ± 0.37 °C. The down core trends based on the Pearson et al. (2011) calibration closely resembled those from other temperature calibrations using MBT/CBT indices (i.e. Weijers et al., 2007a; Tierney et al., 2010b; Sun et al., 2011; Peterse et al., 2012), except that the other calibrations shift absolute temperature by as much as ca. 15 °C. The down core temperature record for Lower EVV Lake showed no long term progressive trend but simply minor variation about the mean value, including a cooling of ca. 1 °C at 690 ± 190 yr BP and a marked warming event of ca. 3 °C at 1220 ± 190 yr BP (Fig. 4A). Values for the Upper EVV Lake sediment sequence (2580 ± 25 yr), again calculated using the Pearson et al. (2011) summer calibration, ranged from 7.2–12.5 °C, with a mean of 10.2 °C (Fig. 4B). Inter-sample stratigraphic variation in paleotemperature for this lake was greater than that for Lower EVV Lake. However, despite differences in sampling resolution, the temperature records for the two lakes exhibited several similarities in terms of alternating cooler and warmer episodes from ca. 2400 to ca. 1000 yr BP. By contrast, the Upper EVV Lake record exhibited a marked long term progressive cooling trend of > 3 °C that began ca. 1000 yr BP and extended to the youngest core sample, which was absent from the Lower EVV Lake record (Fig. 4).

The actual surface water temperature values in June/July for the two lakes were similar and ca. 3 °C warmer than mean down core br GDGT derived values for both lakes, which, in turn, were comparable to July air temperature from Kangerlussuaq (Fig. 4). By contrast, the bottom water values in June/July for the two lakes showed a marked difference, with Upper EVV ca. 2 °C colder than the br GDGT values for the surface sediments.

3.3.3. Br GDGT concentration

The concentration of br GDGTs in Lower EVV Lake and Upper EVV Lake ranged from 0.10–0.95 μg/g and 0.16–2.77 μg/g normalized to extracted dry sediment, respectively, and from 1.69–27.94 μg/g and 0.58–66.26 μg/g normalized to TOC, respectively. The latter value (μg br GDGT/g TOC) in the Lower EVV Lake sediments was broadly comparable with those of the modern soils from the vicinity of the lakes ($t = 1.0148$, $df = 21$, $p = 0.3217$). By contrast, the Upper EVV Lake sedimentary record included several samples with br GDGT concentration far higher than for the soils, most notably for sediments younger than ca. 1000 yr BP (Fig. 5). Additionally, the Upper Lake sediments overall were less similar

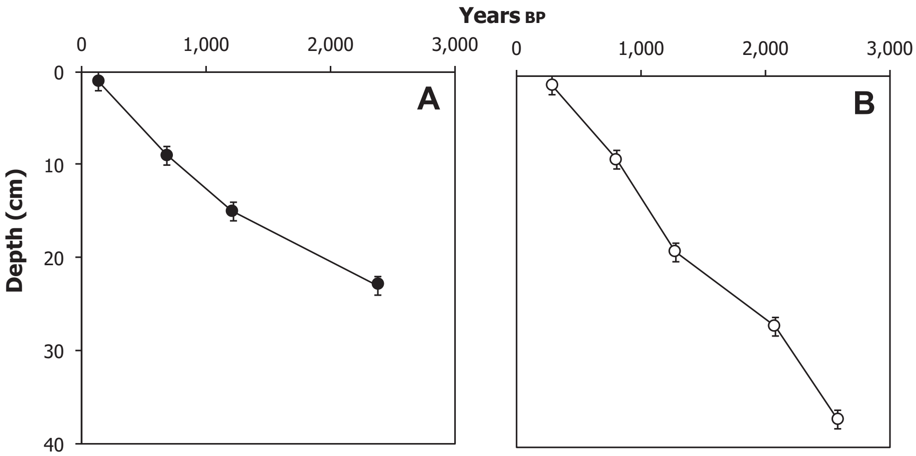

Fig. 3. Radiocarbon dates for individual sediment samples from Lower EVV Lake (A) and Upper EVV Lake (B) showing the proposed chronology for the records. All ^{14}C dates have an error of ±20–30 yr. Vertical error bars represent the 2 cm depth range that each ^{14}C-dated sample spans.

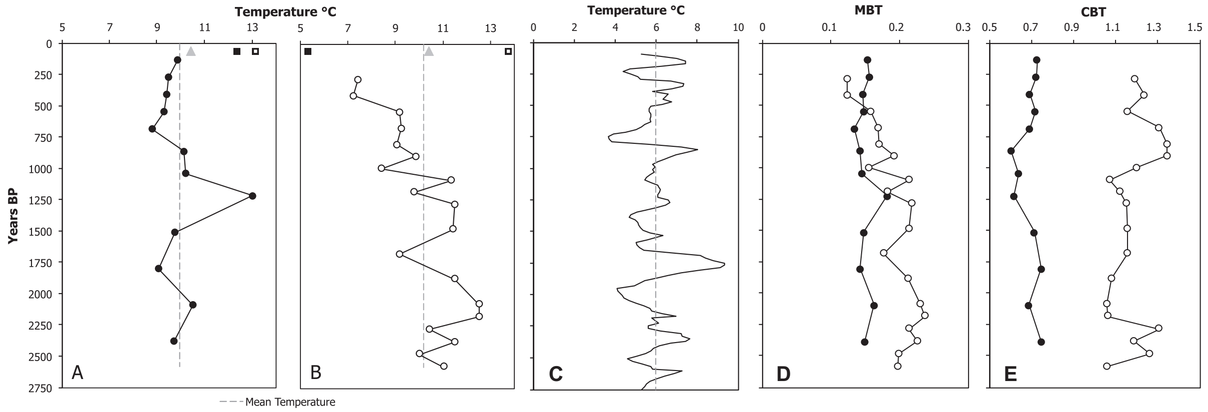

Fig. 4. Paleotemperature records from Lower EVV Lake (A) and Upper EVV Lake (B) derived from br GDGT distributions using the Pearson et al. (2011) calibration. Gray dashed lines represent mean paleotemperature calculated for the entire record of each core. Open and closed squares represent mean June/July water temperature for surface and bottom water, respectively. Gray triangles represent mean July air temperature from Kangerlussuaq. Alkenone-based (C) Kanger Stack temperature (°C) from D'Andrea et al. (2011). Gray dashed line represents mean paleotemperature calculated from 2750 yr BP to present. Down core MBT (D) and CBT (E) values from Upper Lake (open circles) and Lower Lake (closed circles).

to the soils ($t = 1.8268$, $df = 26$, $p = 0.0792$) than the Lower Lake sediments were to the soils.

3.4. Surface sediments

3.4.1. Br GDGT distributions and resulting paleotemperature records

Br GDGT distributions in surface sediments (0–2 cm) from the lakes yielded disparate temperature values (7.4–15.4 °C) based on the Pearson et al. (2011) calibration (RMSE 2.0 °C), and a range of MBT (0.12–0.21) and CBT (0.54–1.19) values (Table 2). In all lakes except South Twin, br GDGT I was the most abundant br GDGT in the surface sediments. In all 5 lakes, Ic, IIc, and IIIc were the least abundant (Fig. S1).

The br GDGT-based temperature for surface sediments from Upper EVV Lake was 2.5 °C cooler than that for Lower EVV Lake, despite the close proximity of the two lakes. The MBT values for the surface sediments from Upper EVV Lake and Lower EVV Lake were broadly comparable with the mean value for soils surrounding the lakes (median MBT 0.14) (Lower Lake: $t = 0.0.1171$, $df = 10$, $p = 0.9091$; Upper Lake: $t = 1.4058$, $df = 10$, $p = 0.1901$), although the overall range for the soils (0.08–0.37; Fig. 2) was more extensive. The CBT values for surface sediments from Upper EVV Lake were significantly higher than those for Lower EVV Lake (Table 2) and this distinction applies to the entire cores (Upper EVV 1.06–1.34; Lower EVV 0.58–0.74) ($t = 17.0413$, $df = 28$, $p < 0.0001$). The median CBT value for the surrounding soils (0.84) lay between the values for the two lakes and the soils exhibited a CBT range (0.49–1.67; Fig. 2) that encompassed all the lake sediment values.

3.4.2. Environmental controls on surface sediment br GDGT distributions

For the four lakes with hypoxic (Teardrop) or anoxic (Upper EVV, Potentilla, and South Twin) bottom water, there was a strong correlation (R^2 0.987, $p < 0.01$) between br GDGT-based temperature values for the surface sediment (0–2 cm) and the average measured bottom water (water < 1 m from the sediment–water interface) values recorded in June and July 2013 (Fig. 6; Cadieux et al., unpublished results). In marked contrast, the values for Lower EVV Lake, which remained saturated with O_2 throughout its entire water column in June and July 2013, plotted far from this trend, suggesting distinctly different controls on the temperature recorded from its br GDGT distributions. Additionally, the correlation between mean summer bottom water temperature, and both MBT and CBT strengthened when Lower EVV Lake (R^2 0.232–0.596; R^2 0.716–0.836, respectively) was excluded.

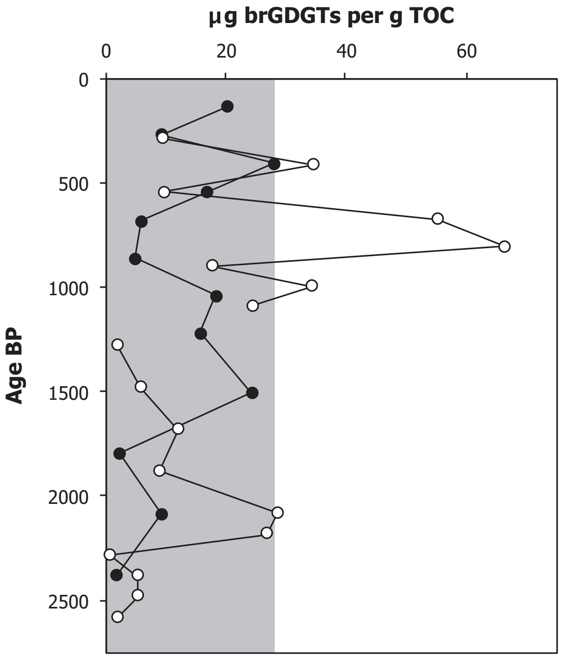

Fig. 5. Br GDGT concentration (µg/g TOC) vs. sediment age for Lower EVV Lake (closed circles) and Upper EVV Lake (open circles) cores and the concentration range of modern soil samples (shaded box).

4. Discussion

4.1. Paleotemperature records

The plethora of br GDGT-based paleotemperature calibrations of proven utility (e.g. Weijers et al., 2007a; Tierney et al., 2010b; Pearson et al., 2011; Sun et al., 2011) combined with the absence of an independent or valid calibration appropriate for the region of study and coupled with the inconsistent absolute temperatures determined using each individual calibration collectively lead to two inferences. First, that the observed trends in temperature are likely authentic and, second, that the temperature values derived from the GDGT proxy based on the Pearson et al., 2011 calibration may not necessarily be accurate as absolute values. A br GDGT temperature record for the Holocene has been obtained for lacustrine sediments from Nanerersarpik in southeastern Greenland (de Wet, 2013). However, the data here represent the first determination of br GDGT-based paleotemperature for southwestern Greenland, which therefore precludes the possibility of direct comparison with other br GDGT records for the region.

The magnitude of the temperature variation in the GDGT records for Lower and Upper EVV Lakes is similar to that reported for another paleotemperature study in western Greenland (D'Andrea et al., 2011), which used the alkenone based U_{37}^{k} temperature proxy (Fig. 4). However, radiocarbon dating shows that the temporal resolution (ca. 190 yr and ca. 120 yr for Lower EVV and Upper EVV lakes, respectively) attained from core sampling at 2 cm intervals is insufficient to discern and define short term climatic events. Hence, discrepancies in the records of specific warming/cooling events from this sampling of Upper and Lower EVV Lake sediment cores are to be expected. Also, similar incongruities can be expected to exist in comparisons with other regional paleotemperature data (D'Andrea et al., 2011), notwithstanding the likelihood of distinct climatic differences arising from the contrasting distances from the ocean and the Greenland ice sheet of the two study areas.

The respective paleotemperature records derived from the sediment cores of Lower EVV and Upper EVV Lakes show some similarities, but also major discrepancies. The br GDGT records of both indicate a warming event from ca. 1500 to 1100 yr BP and from ca. 2100 to 1900 yr BP. However, the comparative magnitude of both events in the two cores is inconsistent, although such discrepancies can partly be attributed to differences in sampling resolution (Fig. 4). There are other inconsistencies between the two cores, most notably a ca. 2 °C offset in the paleotemperature from ca. 400 yr BP to the present. In addition, the longer term cooling trend that appears in the Upper EVV Lake record from ca. 1000 yr BP to the present is not evident in the Lower EVV Lake record (Fig. 4). These significant differences between the paleotemperature records from the two lakes are unexpected. The lakes are

Table 2
Physical characteristics of lakes and br GDGT data for surface sediments.

	Upper EVV	Lower EVV	Teardrop	Potentilla	South twin
Physical characteristics[a]					
Area (ha)	0.2	1.7	1.1	1.6	3.0
Maximum depth (m)	5.5	4.75	5.25	7.75	5.25
Summer dissolved O_2 (%)	0	60.8	8.6	0.6	0.7
Bottom water Temp. (°C)	5.3	12.4	8.3	7.0	12.5
Surface water Temp. (°C)	13.8	13.1	13.9	13.9	15.8
Bottom water pH	6.8	8.7	9.0	7.3	7.9
GDGT data					
GDGT conc. (µg/g TOC)	9.49	20.07	13.71	68.59	3.10
GDGT temp. (°C)[b]	7.4	9.9	11.5	9.3	15.4
MBT[c]	0.12	0.15	0.21	0.15	0.20
CBT[d]	1.19	0.72	0.69	0.88	0.54

[a] Measured in summer 2013 (Cadieux et al., unpublished results).
[b] Based on Pearson et al. (2011) calibration.
[c] Defined in Eq. 2.
[d] Defined in Eq. 3.

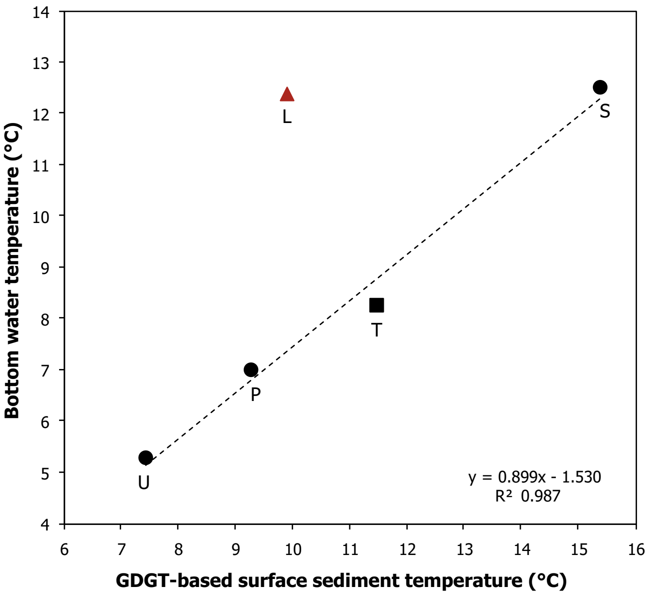

Fig. 6. Br GDGT-based temperature for surface sediments (0–2 cm) for each of the five lakes vs. average bottom water temperature measured in June/July 2013. Lakes with anoxic bottom water (i.e. Upper EVV, U; Potentilla, P and South Twin, S) are represented by black circles, the hypoxic lake (Teardrop, T) is shown by a black square and the fully oxygenated lake (Lower EVV, L) is designated by a red triangle. There is a strong correlation (dashed line) between GDGT-based temperature and measured summer bottom water temperature in the four lakes with little or no O_2 (R^2 0.987; for interpretation of the references to color in this figure legend, the reader is referred to the web version of the article).

< 100 m apart and share a common catchment area, which means that they have experienced a comparable climate throughout their history. Hence, the supply of br GDGTs from local soils, likely via seasonal eolian transport or soil runoff, would be expected to be similar for both lakes, thereby producing broadly comparable records. Discrepancies in paleotemperature could arise as a function of sampling resolution related to differences in sedimentation rate associated with basin morphology (Cadieux et al., unpublished results). However, this cannot account for the significant differences in temperature and other temporal variation in the two records, notably the prominent offset from ca. 1000 yr BP to the present. Thus, it is evident that the surrounding soils cannot be the sole source of br GDGTs in these lacustrine sediments. An additional influence must be invoked to explain differences in the comparative distribution of br GDGTs in two discrete lacustrine records that lie within a common catchment area.

4.2. Multiple br GDGT sources

Br GDGT concentrations relative to TOC in sediments from Lower EVV Lake are comparable to those in the surrounding soil (Fig. 5). In addition, the distributions of br GDGTs in the Lower EVV Lake surface sediment expressed in terms of specific indices (i.e. MBT, CBT) are similar to those of the surrounding soils and the br GDGT-based temperature (9.9 °C) is roughly comparable with that of summer air temperature for the region (ca. 10.5 °C). By contrast, the distributions and down core concentrations of br GDGTs for Upper EVV Lake are markedly different (Fig. 2, Table 2). Specifically, the CBT value (1.19) is distinctly different from the surrounding soils (mean 0.95; median 0.84) and the surface sediment of Lower EVV Lake (0.72). Moreover, the br GDGT concentrations in Upper EVV Lake exceed those of the surrounding soils, specifically from ca. 1000 yr BP to the present (Fig. 5). Collectively the data are consistent with the interpretation that soils serve as the dominant, and possibly sole, contributor of br GDGTs to the Lower EVV Lake sediments, whereas Upper EVV Lake clearly shows evidence for some secondary source of br GDGTs that is absent from or minimal in Lower EVV Lake. This additional supply or production of br GDGTs enhances the concentration of these compounds and influences the relative distributions of br GDGTs in Upper EVV Lake sediments. It is notable that a large increase in br GDGT concentration in the Upper EVV Lake record occurred ca. 1000 yr BP, approximately coincident with the onset of major discrepancies between the two lake records, so it seems that the secondary br GDGT source within Upper EVV Lake became a significant factor at that time. The timing happens to coincide with a rapid cooling event observed in other Arctic records from Greenland (D'Andrea et al., 2011; Fig. 4) and Svalbard (D'Andrea et al., 2012), which may perhaps have triggered changes in environmental and/or hydrological conditions favoring allochthonous production of br GDGTs in Upper EVV Lake. The wide range of br GDGT-based temperature (7.4–15.4 °C) for the surface sediments from all five lakes provides further evidence in support of a secondary source of br GDGTs because the data suggest that br GDGT distributions do not simply record soil-derived air temperature or even the narrow range of lake surface water temperature (13.1–15.8 °C), but attest to some other contributing factor affecting the distributions in specific lakes.

4.3. Evidence for anaerobic autochthonous production of br GDGTs

The strong correlation (R^2 0.987) between br GDGT-based temperature for the surface sediments (0–2 cm) and average June/July bottom water temperature (Fig. 6) for the four lakes that have either anoxic (Upper EVV, Potentilla, South Twin) or hypoxic (Teardrop) bottom water merits specific consideration. Indeed, it is significant that the correlation of br GDGTs with bottom water temperature is stronger than their relationship to any other measured physical characteristic of the lakes. For example, the correlation coefficients of br GDGT temperature with summer surface water temperature and the distance from the terminus of Russell Glacier are 0.651 and 0.698, respectively, and those for CBT relative to summer bottom water temperature (cf. Zink et al., 2010) and pH are 0.716 and 0.799, respectively. Collectively, these data substantiate arguments that temperature represents the primary control on br GDGT distributions (cf. Loomis et al., 2014). The most plausible explanation for the observed temperature correlation is the presence of (a) substantial autochthonous anaerobic source(s) of br GDGTs within either the bottom water of each lake or its sediment, producing a temperature-dependent portion of sedimentary br GDGTs that records the bottom water temperature of the various lakes. This interpretation furthers the conclusions of recent studies that proposed autochthonous production of br GDGTs in lacustrine environments (Tierney and Russell, 2009; Tierney et al., 2012; Schoon et al., 2013; Loomis et al., 2014; Buckles et al., 2014), notably when dissolved O_2 is low. This autochthonous source of br GDGTs is absent from or minimal in the oxic bottom water of Lower EVV Lake, where the allochthonous br GDGT signal from soils is dominant. By contrast, we conclude that sediments in lakes from the study area with O_2-limited bottom water likely receive a br GDGT contribution from both allochthonous soil-derived sources and in situ autochthonous microbial production, thereby preserving a br GDGT temperature signal that combines records of soil-derived air temperature and summer (i.e. growing season) bottom water temperature. Although in situ production likely occurs primarily in the bottom water, a minor contribution from the upper water column (Buckles et al., 2014) cannot be excluded.

These br GDGT data from lakes in southwestern Greenland have major implications for the interpretation of lacustrine paleotemperature records, suggesting that the distributions in small high latitude lakes with fully oxygenated bottom water dominated by an allochthonous input provide a reliable record of paleotemperature, specifically summer air temperature. However, sedimentary br GDGT distributions in lakes that experience episodes of hypoxia or anoxia likely combine records of soil-derived air temperature and summer bottom water temperature in varying proportion, thereby yielding paleotemperature signals that are difficult to decipher and interpret, especially for small lakes with relatively small watersheds. Yet, variation in br GDGT distributions may primarily reflect the response of soil bacteria to air temperature where lakes are sufficiently deep that the bottom water temperature remains constant year-round. Indeed, such a consistent cold bottom water signal mixing with a changing soil-derived signal could possibly contribute to the "cold bias" observed in some sedimentary records from deeper lakes (Tierney and Russell, 2009; Tyler et al., 2010; Zink et al., 2010; Tierney et al., 2010b; Pearson et al., 2011). Thus, the additional evidence for autochthonous production of br GDGTs in small anoxic/hypoxic Greenland lakes confirms the importance of understanding the environmental characteristics of individual lake systems, and therefore their likely source(s) of br GDGTs, as a prerequisite for assessment and interpretation of br GDGT-based paleotemperature from their sediment records.

5. Conclusions

Assessment of br GDGT distributions in sediments from a series of small lakes in southwestern Greenland, coupled with investigation of these compounds in surrounding soils, suggests that the stratigraphic profile of br GDGTs for Lower EVV Lake, which has a fully oxygenated water column, records trends in local air temperature. Paleotemperature values based on the Pearson et al. (2011) calibration appear reasonable, although the absence of independent proxies for the region precludes their validation as accurate measures of absolute temperature. The concentrations of br GDGTs in these lake sediments are sufficient to permit higher resolution sampling and thereby assessment of short term climate fluctuations that could not be addressed by this investigation at 2 cm sediment intervals. The ability to interpret the paleotemperature record for Upper EVV Lake is compromised by evidence for anaerobic production of br GDGTs in the bottom water and/or sediment–water interface, which supplements and, in some intervals exceeds, the supply of br GDGTs from surrounding soils. Assessment of br GDGT distributions in the surface sediments of three other Greenland lakes with anoxic/hypoxic bottom waters strengthens the evidence for multiple sources of br GDGTs, confirming that interpretation of lacustrine br GDGT-based paleotemperature records requires contextual knowledge for each individual lake system and of likely sources of sedimentary br GDGTs. It is unclear whether the recent recognition of differences in the position of methylation of br GDGTs might help resolve and discriminate their autochthonous and allochthonous sources (De Jonge et al., 2014). Thus, further research is needed to investigate the relationship between environmental temperature and br GDGT distribution/concentration in lakes that experience anoxia, both annually and seasonally, and the possibility of production in subsurface anoxic sediments. Moreover, further evidence of br GDGT production in the bottom water and/or sediment–water interface of other anoxic lakes would facilitate the possibility to account for multiple sources of br GDGTs in future calibrations through incorporation and combination of both air and bottom water temperatures. These considerations undoubtedly complicate the use of br GDGT-based proxies in lacustrine settings but also provide opportunities to resolve and comprehend bacterial production of br GDGTs in different environments, ultimately leading to the ability to recognize and differentiate the various bacterial contributions of br GDGTs within sedimentary records.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.orggeochem.2015.02.005.

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