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# Short-term variability in the sedimentary BIT index of Lake Challa, East Africa over the past 2200 years: validating the precipitation proxy

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## Abstract

The branched vs. isoprenoid index of tetraethers (BIT index) in Lake Challa sediments has been applied as a monsoon precipitation proxy on the assumption that the primary source of branched tetraether lipids (brGDGTs) was soil washed in from the lake's catchment. However, water column production has since been identified as the primary source of brGDGTs in Lake Challa, meaning that there is no longer a clear mechanism linking BIT index variation and precipitation. Here we investigate BIT index variation and GDGT concentrations at a decadal resolution over the past 2200 years, in combination with GDGT data from profundal surface sediments and 45 months of sediment-trap deployment.

The 2200 year record reveals high-frequency variability in GDGT concentrations, and therefore the BIT index. Also surface sediments collected in January 2010 show a distinct shift in GDGT composition relative to those collected in August 2007. Increased bulk flux of settling particles with high Ti / Al ratios during March–April 2008 reflect an event of high detrital input to Lake Challa, concurrent with intense precipitation at the onset of the principal rain season that year. Although brGDGT distributions in the settling material are initially unaffected, this soil erosion event is succeeded by a large diatom bloom in July–August 2008 and a concurrent increase in GDGT-0 fluxes. Near-zero crenarchaeol fluxes indicate that no thaumarchaeotal bloom developed during the subsequent austral summer season; instead a peak in brGDGT fluxes is observed in December 2008. We suggest that increased nutrient availability, derived from eroded soil washed into the lake, stimulated both diatom productivity and the GDGT-0 producing archaea which help decompose dead diatoms passing through the suboxic zone of the water column. This disadvantaged the Thaumarchaeota that normally prosper during the following austral summer. Instead, a bloom of supposedly heterotrophic brGDGT-producing bacteria occurred.

Episodic recurrence of such high soil-erosion events, integrated over multi-decadal and longer timescales and possibly enhanced by other mechanisms generating low

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BIT index values in dry years, can explain the positive relationship between the sedimentary BIT index and monsoon precipitation at Lake Challa. However, application elsewhere requires ascertaining the local situation of lacustrine brGDGT production and of variables affecting the productivity of Thaumarchaeota.

## 1 Introduction

Geographically widespread isoprenoid and branched glycerol dialkyl glycerol tetraether (iso/brGDGT) membrane lipids have allowed the development of several new molecular proxies used in palaeoenvironmental reconstruction (e.g. Schouten et al., 2002, 2013; Hopmans et al., 2004; Weijers et al., 2007a). Although isoGDGTs can be found in soil (Leininger et al., 2006) and peat (Weijers et al., 2004, 2006), they are generally most abundant in marine and freshwater environments (Sinninghe Damsté et al., 2002; Blaga et al., 2009). IsoGDGT-producing Thaumarchaeota (Brochier-Armanet et al., 2008; Spang et al., 2010; earlier referred to as Crenarchaeota, e.g. Wuchter et al., 2006) are now known to occur in most medium to large lakes (Blaga et al., 2009). Since brGDGTs were originally thought to be produced solely in soil and peat (e.g. Hopmans et al., 2004; Weijers et al., 2007a; Schouten et al., 2013), the Branched vs. Isoprenoid Tetraether (BIT) index was developed as a proxy for soil organic matter input in marine sediments (Hopmans et al., 2004; Weijers et al., 2009). BIT expresses the abundance of brGDGTs relative to crenarchaeol, the characteristic membrane lipid of pelagic Thaumarchaeota (Sinninghe Damsté et al., 2002; Pitcher et al., 2011). Subsequently, the BIT index was extended to lake sediments (e.g. Verschuren et al., 2009; Wang et al., 2013). However, this application has become complicated by recent indications of brGDGT production within lakes (e.g., Tierney and Russell, 2009; Tierney et al., 2010; Loomis et al., 2011). Also in Lake Challa, in situ production has been identified in the water column (Sinninghe Damsté et al., 2009; Buckles et al., 2014a) and suggested, but not confirmed, in profundal surface sediments (Buckles et al., 2014a).

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Rainfall variability in equatorial East Africa is governed by biannual passage of the Intertropical Convergence Zone (ITCZ), with the intensity of northeasterly and southeasterly monsoons strongly linked to precessional insolation forcing at long time scales (Verschuren et al., 2009), and to El Niño Southern Oscillation (ENSO) dynamics at inter-annual time scales (Wolff et al., 2011). Verschuren et al. (2009) presented a 25 000 year BIT index record for Lake Challa near Mt. Kilimanjaro, which corresponded well with both known climatic events for the region and the succession of local lake highstands and lowstands evidenced in high-resolution seismic-reflection data. The BIT index was thus interpreted to reflect changes in the amount of soil-derived brGDGTs, associated with variation in the rate of soil erosion that was assumed proportional to rainfall intensity. The recent evidence for overwhelming in situ production of brGDGTs within Lake Challa (Buckles et al., 2014a) implies that the BIT index may not respond (or at least not directly) to a variable influx of soil organic matter, but is rather controlled by the in-lake production of crenarchaeol by Thaumarchaeota (Sinninghe Damsté et al., 2012a). Strong dependence of the BIT index in Lake Challa sediments on crenarchaeol abundance, rather than brGDGT abundance, was also evident in an almost 3 year monthly time series of settling particles (Buckles et al., 2014a). The precise mechanism(s) by which the BIT index responds to changes in precipitation has thus remained elusive. Further investigating the issue, we here present a 2200 year record of GDGT distributions in the Lake Challa sediment record with decadal resolution, with the aim to bridge the resolution (and thus information) gap between our time series of sediment-trap data and the 25 000 year climate-proxy record. To this end, we also analyse GDGT distributions in a chronosequence of recent profundal surface sediments.

## 2 Materials and methods

### 2.1 Study site

Lake Challa is a 4.2 km<sup>2</sup> crater lake in equatorial East Africa, situated at 880 m elevation in the foothills of Mt. Kilimanjaro. High crater walls (up to 170 m) confine a small catchment area of 1.38 km<sup>2</sup>, which during periods of exceptional precipitation can enlarge to 1.43 km<sup>2</sup> due to activation of a small creek in the NW corner of the lake (Fig. 1). The water budget of this deep lake (92 m in 2005) is dominated by groundwater, which accounts for ca. 80 % of hydrological inputs and is mostly derived from rainfall on the montane forest zone of Mt. Kilimanjaro (1800 to 2800 m elevation; Hemp, 2006). Passage of the Intertropical Convergence Zone (ITCZ) twice annually results in a short and a long rainy season. “Long rains” occur from March to mid-May, while typically more intense “short rains” stretch from late October to December (e.g. Verschuren et al., 2009; Wolff et al., 2011). Over the course of sediment-trap deployment, a long drought stretched from May 2007 to February 2008 due to failure of the short rains in 2007 (Fig. 2a). Air temperatures at the lake are lowest (20–21 °C; 24 h average) in July–August (austral winter), and the highest (~ 25 °C; 24 h average) in January–February (austral summer; data provided by A. Hemp, University of Bayreuth; cf. Buckles et al., 2014a). The lake surface water is coolest (~ 23 °C) between June and September, promoting seasonal deep mixing that reaches down to 40–60 m depth. During austral summer from February to April, the surface water is ~ 28 °C, and experiences daily stratification followed by wind-driven and convective mixing down to 15–20 m depth (Wolff et al., 2014). The bottom water of Lake Challa is constantly 22.3 °C and permanently anoxic, since it does not mix even on a decadal scale. The finely laminated profundal sediments of Lake Challa (Wolff et al., 2011) contain diatom silica mainly deposited during the months of deep seasonal mixing (Barker et al., 2011), alternating with organic matter and calcite laminae that are deposited during the austral spring and summer to produce alternating dark/light layers.

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ilent 1100 series HPLC connected to a Hewlett-Packard 1100 MSD SL mass spectrometer in selected ion monitoring (SIM) mode, using the method described by Schouten et al. (2007). A standard mixture of crenarchaeol: C<sub>46</sub> GDGT was used to check, and to account for, differences in ionisation efficiencies.

5 GDGT distributions in the samples were quantified using the following indices:

$$\text{BIT index} = \frac{[\text{VI}] + [\text{VII}] + [\text{VIII}]}{[\text{VI}] + [\text{VII}] + [\text{VIII}] + [\text{V}]} \quad (1)$$

$$\text{MBT} = \frac{[\text{VI}] + [\text{VIb}] + [\text{VIc}]}{[\text{VI}] + [\text{VIb}] + [\text{VIc}] + [\text{VII}] + [\text{VIIb}] + [\text{VIIf}] + [\text{VIII}] + [\text{VIIIb}] + [\text{VIIIc}]} \quad (2)$$

$$\text{DC} = \frac{[\text{VIb}] + [\text{VIIf}]}{[\text{VI}] + [\text{VIb}] + [\text{VII}] + [\text{VIIf}]} \quad (3)$$

The fractional abundance of each individual GDGT is expressed as:

$$10 \quad f[\text{GDGT } i] = \frac{[\text{GDGT } i]}{[\sum \text{GDGTs}]} \quad (4)$$

where  $f[\text{GDGT } i]$  = fractional abundance of an individual GDGT,  $[\text{GDGT } i]$  = concentration of the individual GDGT, based on surface area relative to the C<sub>46</sub> standard, and  $[\sum \text{GDGTs}]$  = the summed concentration of all measured GDGTs (I to VIIIc, see Appendix).

15 The proportion of IPL compared to CL GDGTs is expressed using % IPL, defined as:

$$\% \text{IPL} = \left( \frac{[\text{IPL}]}{[\text{IPL}] + [\text{CL}]} \right) \times 100 \quad (5)$$

where  $[\text{IPL}]$  = intact polar lipid concentration and  $[\text{CL}]$  = core lipid concentration. IPLs represent living, GDGT-producing bacteria/archaea (e.g. Lipp and Hinrichs, 2009; Pitcher et al., 2011b; Schubotz et al., 2009; Lengger et al., 2012; Schouten et al., 2012; Gibson et al., 2013).

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## 2.5 Statistical analysis

Pearson product-moment correlation coefficients were calculated on un-smoothed time-series data of the core sequence, using a two-tailed test of significance in IBM SPSS Statistics 21, with bootstrapping at the 95% confidence interval and missing values excluded pairwise. Linear varve thicknesses (Wolff et al., 2011) were averaged over the depth intervals of GDGT analysis. Following general guidelines, the strength of (positive/negative) correlation was considered weak if less than 0.3, moderate from 0.3 to 0.5, and strong from upwards of 0.5.

## 3 Results

### 3.1 The 2200 year BIT-index record

The percentage total organic carbon (%  $C_{org}$ ) in the composite sediment sequence (Table S1 in the Supplement) varies from 4.4 to 12.5%, with the lowest values generally grouping between 1200 and 1830 AD (Fig. 3a). The concentrations of GDGTs quoted here are expressed relative to  $C_{org}$  content, unless otherwise stated. The concentration of GDGT-0 (GDGT-I; Appendix) varies widely (97 to 921  $\mu\text{g g}^{-1} C_{org}$  and SD of 150  $\mu\text{g g}^{-1} C_{org}$ ; Table S1) and at an average of 273  $\mu\text{g g}^{-1} C_{org}$  is generally high. A baseline concentration of 200–400  $\mu\text{g g}^{-1} C_{org}$  is interrupted by relatively long-term pulses of > 500  $\mu\text{g g}^{-1} C_{org}$  (Fig. 3b), the longest of which stretch from around AD 100 to 200, AD 300 to 500 and AD 1200 to 1400.

Crenarchaeol fluctuates by two orders of magnitude between 9 and 412  $\mu\text{g g}^{-1} C_{org}$  (ca. 1210 and 1810 AD respectively; SD of 64  $\mu\text{g g}^{-1} C_{org}$  Table S1). Periods of high crenarchaeol (> 150  $\mu\text{g g}^{-1} C_{org}$ ) occur from around AD 400 to 450, AD 1250 to 1300 and AD 1700 to 1850 (Fig. 3c). The proportion of the crenarchaeol regioisomer with respect to crenarchaeol ( $\text{GDGTV}' / (\text{V} + \text{V}')$ ) is relatively low and constant at around

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2.5 to 3% throughout the analysed core sequence (with a maximum of 4.0% around 730 AD; Fig. 3d), confirming that the majority of recovered crenarchaeol originates from aquatic, rather than soil, Thaumarchaeota (cf. Sinninghe Damsté et al., 2012a, b).

The concentration of total brGDGTs relative to %C<sub>org</sub> varies by one order of magnitude between 94 and 493 µg g<sup>-1</sup> C<sub>org</sub> (Table S1). BrGDGT concentrations are higher, on average, than those of crenarchaeol (197 vs. 113 µg g<sup>-1</sup> C<sub>org</sub>, respectively; brGDGT SD of 68 µg g<sup>-1</sup> C<sub>org</sub>) but similarly display a baseline (here between 200 and 250 µg g<sup>-1</sup> C<sub>org</sub>; Fig. 3e) interspersed by peaks of which the timing generally corresponds with those reported for crenarchaeol. This trend persists when using absolute concentrations in µg g<sup>-1</sup> dry weight). In fact, brGDGT concentrations correlate strongly with crenarchaeol concentrations and those of its regioisomer ( $r = 0.67$  and  $0.67$ ; Table S2).

The BIT index ranges between 0.42 (101–102 cm; ca. AD 1175) and 0.93 (142–143 cm; ca. AD 730), with an average of  $0.65 \pm 0.09$  (Table S1). Generally higher BIT values are evident from ca. AD 650 to 950 (Fig. 3f), followed first by a period of lower BIT values (ca. AD 1150 to 1270) and then a period of higher BIT values (ca. AD 1550 to 1700). Following a 60 yr period of lower BIT values (AD 1770 to 1830), an overall increase to the present is interrupted by two ~ 30 yr periods of lower BIT values, in the late 19th century and the mid-20th century. The BIT index does not correlate with the concentrations of any brGDGTs, but shows strong negative correlation with the concentrations of crenarchaeol and its regioisomer ( $r = -0.70$  and  $-0.68$ , respectively; Table S2). The BIT index also correlates with measures of brGDGT distribution: moderately positive with MBT ( $r = 0.44$ ) but weakly so with DC ( $r = 0.16$ ; Table S2). MBT values (ranging 0.40 to 0.54) and DC (0.15 to 0.26) themselves do not vary widely (Fig. 4; Table S1).

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## 3.2 Settling particles

Results for bulk sediment flux, % C<sub>org</sub>, crenarchaeol and brGDGTs in the monthly sediment-trap time series have been presented elsewhere (Sinninghe Damsté et al., 2009; Buckles et al., 2014a). Here they are shown (Fig. 2b, d and e) as reference for new data on the CL and IPL fractions of GDGT-0 (Fig. 2c). Fluxes of IPL GDGT-0 in settling particles are generally low (0.3–0.4 µg m<sup>-2</sup> day<sup>-1</sup>) from the start of its measurement in December 2007 until June 2008 (Fig. 2c, Table S3), but subsequently peak at 7.7 µg m<sup>-2</sup> day<sup>-1</sup> in August 2008 at the height of a massive, mixing-season diatom bloom (Fig. 2b) After this maximum, IPL GDGT-0 fluxes vary between 0.0 and 1.7 µg m<sup>-2</sup> day<sup>-1</sup>, with an additional peak of 2.8 µg m<sup>-2</sup> day<sup>-1</sup> in September 2009, again coincident with a (now smaller) seasonal diatom bloom (Barker et al., 2011). CL GDGT-0 fluxes track those of IPL GDGT-0 but are notably lower, ranging from < 0.05 to 2.0 µg m<sup>-2</sup> day<sup>-1</sup> (Table S3). From December 2007 to the end of August 2008, IPL GDGT-0 contribute a flux-weighted average of 77 % to total GDGT-0.

## 3.3 Surface sediments

Sinninghe Damsté et al. (2009) and Buckles et al. (2014a) presented data on % C<sub>org</sub>, crenarchaeol (including its regioisomer), GDGT-0 and brGDGTs in Lake Challa surface sediments collected in, respectively, August 2007 (from gravity core CH07-1G: 0–0.5 and 0.5–1 cm depth, here combined into a single result for 0–1 cm) and January 2010 (seven CH10 gravity core tops, all 0–1 cm depth). Here they are shown again (Figs. 5b, c and 6a, c) as reference for new data on the 2200 yr sediment record and on IPL and CL GDGT-0 (Tables 1 and S4). IPL and CL GDGT-0 concentrations in CH10 surface sediments are, on average, 14.5 and 8.7 µg g<sup>-1</sup> dry wt. (Table 1). The dominant GDGT in these sediments is GDGT-0, with fractions of 0.85 (IPL) and 0.49 (CL) (Fig. 6a; Table 1). Additionally, IPL GDGT-0 represents on average 61 % of total GDGT-0.

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## 4 Discussion

### 4.1 Temporal variability in sedimentary GDGT composition and BIT index

The 2200 year, decadal-resolution organic geochemical record of Lake Challa shows a great deal of variation in GDGT composition (Fig. 3b–e), particularly with respect to the concentrations of brGDGTs and crenarchaeol that underpin the BIT index. Here we quantified the absolute GDGT concentrations, which had not been examined for the 25 000 yr, lower-resolution record (Verschuren et al., 2009; Sinninghe Damsté et al., 2012a), allowing greater insight into the factors potentially affecting the BIT index over time. Our 2200 year record is generated from the upper portion of the same composite core sequence, consequently it generates comparable BIT values (both in absolute values and variability) when averaged over four adjacent 1 cm samples to mimic the measurements on homogenized 4 cm increments of the low-resolution record (Fig. 3g). This demonstrates that the lower-resolution analysis fails to capture strong variation in sedimentary GDGT concentrations (and therefore in the BIT index) on short timescales (Fig. 3b–e). To better understand this high-frequency variability, we first evaluate what can be learned from variability in the present-day system as reflected in the sediment-trap time series and in our chronosequence of surface sediments.

### 4.2 GDGT variability in Lake Challa settling particles and surface sediments

The soft, uncompacted top centimetre of mid-lake profundal surface sediments in Lake Challa represents approximately two years of deposition (Sinninghe Damsté et al., 2009; see also Blaauw et al., 2011). CH07 can, therefore, be treated as broadly representing the period from mid-2005 to August 2007, and CH10 the period from early 2008 to January 2010. By comparison, one centimetre of compacted sediments in our 2200 year record represents about a decade of deposition.

GDGTs in the CH10 surface sediments are dominated by GDGT-0 and brGDGTs, with relatively low proportions of crenarchaeol (Fig. 5a–c). IPL and CL BIT index val-

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the peak of the 2008 diatom bloom, and the position of the sediment trap in the sub-oxic water column, we tentatively infer that the GDGT-0 producers in Lake Challa are likely involved in the degradation of dead, settling diatoms as the austral winter bloom reaches its peak. Alternatively, as a result of the unusually high oxygen demand of the 2008 diatom bloom, the chemocline may have temporarily ascended. This could have resulted in the GDGT-0 producing archaea residing above the sediment trap and their signal captured by our analysis.

### 4.3 Effect of a soil-erosion event on the GDGT-producing community

The occurrence of a short-lived influx of allochthonous material in March–April 2008 provides a potential explanation for the change in the GDGT-producing community of Lake Challa that caused the dramatic shift in GDGT composition between CH07 and CH10 surface sediments.

Material settling in Lake Challa from March to May 2008 displayed a singularly large peak in the molar ratio of titanium to aluminium (Ti/Al; Fig. 2a), a tracer for the origin of detrital mineral sediment components (Weltje and Tjallingii, 2008; Wolff et al., 2014). This coincided with a peak in bulk flux ( $4.0\text{--}2.0\text{ g m}^{-2}\text{ day}^{-1}$ ), very low organic-matter content (3%  $C_{\text{org}}$ ; Fig. 2b), and local reports of the lake “turning brown”, all pointing to enhanced allochthonous input to the lake triggered by the onset of the principal rain season that year (Fig. 2a). The likely source of this material is loose topsoil on and beyond the NW rim of Challa crater, mobilised during particularly intense precipitation and carried to the lake by the usually dry creek which breaches the rim there (Fig. 1). This particular precipitation event occurred after 10 consecutive months of drought (Fig. 2a; Table S3), which must have wilted the already sparse vegetation cover in that area and made significant soil erosion more likely. Notably, brGDGT distributions and abundances in particulate matter settling during these months were not discernibly affected by soil-derived brGDGTs, most probably due to the high background flux of lacustrine brGDGTs and the low organic matter content of the eroded soil (Buckles

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2011). Since stronger austral-winter winds are associated with a weak southeasterly monsoon compromising the main rain season during March–May, dry years promote large diatom blooms that are followed by a greater proliferation of Thaumarchaeota and crenarchaeol production. As a result, dry years may tend to produce low BIT indices.

#### 5 4.5 A high-resolution record of monsoon precipitation?

Wolff et al. (2011) produced a high-resolution record of inter-annual rainfall variation over the past 3000 years based on varve thickness variation in the same composite sediment sequence that we analysed at 1 cm resolution (Fig. 7a). Varve thickness in Lake Challa is mainly determined by the abundance of diatom frustules developing during the mixing season in austral winter; thus thicker varves were tied to stronger and more extended seasonal windy conditions associated with dry (La Niña) years. Although it was noted by Wolff et al. (2011) that this precipitation reconstruction corresponds with the findings of the low-resolution, 25 kyr BIT index record (Verschuren et al., 2009), here we find only a weak (negative) correlation ( $r = -0.14$ ) between the high-resolution BIT index and integrated varve thickness per cm (Table S2; Fig. 7b). Since the varve thickness record has been shown to correspond well with El Niño/La Niña events spanning just 2 to 3 years, it seems likely that the wide variability seen in the BIT index record does not reflect the variability in crenarchaeol fluxes over 2–3 years (as observed in the sediment-trap record) sufficiently to work well as precipitation proxy at interannual timescale. However, on the decadal timescale the BIT index record average out the occurrence of multiple rainfall-driven “events”, such as the large influx of soil organic matter reported here, that affect local microbial community structure and therefore the balance of GDGTs deposited in sediments. In fact, the three most prominent negative excursions in our high-resolution BIT-index record match the timing of the three most prominent episodes of (multi-)decadal drought in equatorial East Africa over the past 250 years (Fig. 7b): the late-18th and earliest 19th century (Verschuren, 1999; Verschuren et al., 2000; Bessems et al., 2008); the late 19th century (Nicholson et al., 2012); and the mid-20th century (Verschuren et al., 1999; Ver-

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schuren, 2004). The BIT record also replicates the generally dry conditions during the 12th and 13th centuries and wetter conditions during the Little Ice Age known from elsewhere in the region (Verschuren, 2001, 2004). We therefore suggest that the BIT index should not be used as a precipitation proxy on the interannual timescale. Rather, one data point per decade seems sensible, with a five-point moving average (Fig. 7b) providing a robust reconstruction of longer-term dry/wet trends.

Finally, we note that our reconstruction of monsoon precipitation using the BIT index of Lake Challa sediments is contingent upon site-specific conditions: the permanent stratification of its lower water column, dominance of in situ produced brGDGTs, strong seasonality in rainfall, drought and wind, and the resulting intermittent fluxes of soil mobilised from a semi-arid landscape. Since these peculiarities make the BIT index an effective precipitation proxy at Lake Challa, its application in other lakes is only recommended when the degree of in situ brGDGT production and factors controlling the productivity of Thaumarchaeota have been identified.

## 5 Conclusions

Loose soil material transported to Lake Challa by intense precipitation between March and May 2008 stimulated aquatic productivity and set in motion a sequence of events which shifted the composition of GDGTs exported to profundal bottom sediments. It included a suppression of the seasonal Thaumarchaeota, and thus reduced the production of crenarchaeol reflected in the BIT index of settling particles and profundal sediments. Similarly, variation in the sedimentary BIT index over the past 2200 years results from fluctuations in crenarchaeol production against a background of high in situ brGDGT production. Integrated over 10 year intervals, the magnitude of this longer-term BIT-index variation is smaller than that observed in the 45 month long time series of settling particles, but similar to that observed between two sets of recent surface sediments collected before and after the episode of Thaumarchaeota suppression.

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Decadally-averaged BIT index values correlate weakly with the annually-resolved precipitation reconstruction for the Lake Challa region based on varve thickness, but capture the three most prominent known episodes of prolonged regional drought of the past 200 years. This suggests that, while rainfall-triggered events of Thaumarchaeota suppression may occur rather infrequently at the inter-annual time scale, their probability of occurrence is reduced during longer episodes of relative drought, and enhanced during longer episodes of higher average rainfall, such that a temporally integrated BIT index record reflects multi-decadal trends in rainfall. Marked decadal maxima and minima in the sedimentary BIT index are further smoothed by integration over longer intervals, such as the ca. 40 year intervals represented by each data point in the 25 000 year BIT index record (Verschuren et al., 2009; Sinninghe Damsté et al., 2012a). We conclude that the BIT index is a reliable precipitation proxy, at least in the Lake Challa system and on (multi-)decadal and longer time scales. However, prior to application elsewhere we recommend ascertaining the local situation of lacustrine brGDGT production and of variables affecting the productivity of Thaumarchaeota.

**The Supplement related to this article is available online at doi:10.5194/cpd-11-1177-2015-supplement.**

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**Table 1.** Mean ( $\pm\sigma$ ) GDGT distributions and indices in CH10 and CH07 surface sediments. CH07 sediment data are collated from Sinninghe Damsté et al. (2009) and CH10 sediment data are collated from Buckles et al. (2014a).

Depth interval (cm)	No. of cores	Water depth (m)		GDGT-I ( $\mu\text{g g}^{-1}$ dry wt.)	GDGT-V ( $\mu\text{g g}^{-1}$ dry wt.)	$\Sigma$ [brGDGTs] ( $\mu\text{g g}^{-1}$ dry wt.)	BIT	MBT	DC	
CH10 0–1	7	68–92	IPL	14.5 ( $\pm$ 5.3)	0.3 ( $\pm$ 0.5)	2.2 ( $\pm$ 1.1)	0.90 ( $\pm$ 0.10)	0.34 ( $\pm$ 0.06)	0.15 ( $\pm$ 0.01)	
			CL	8.7 ( $\pm$ 3.2)	1.2 ( $\pm$ 1.5)	8.5 ( $\pm$ 2.0)	0.87 ( $\pm$ 0.13)	0.36 <sup>2</sup> ( $\pm$ 0.06)	0.17 ( $\pm$ 0.02)	
CH07 0–1	1	94	CL	5.1	7.4	6.5	0.50	n.m. <sup>2</sup>	n.m.	
Depth interval (cm)	No. of cores	Water depth (m)		$f$ [GDGT-I] <sup>3</sup>	$f$ [GDGT-II]	$f$ [GDGT-III]	$f$ [GDGT-IV]	$f$ [GDGT-V]	$f$ [GDGT-V']	$f$ [ $\Sigma$ brGDGTs]
CH10 0–1	7	68–92	IPL	0.85 ( $\pm$ 0.10)	0.00 ( $\pm$ 0.00)	0.00 ( $\pm$ 0.00)	0.00 ( $\pm$ 0.00)	0.02 ( $\pm$ 0.02)	0.00 ( $\pm$ 0.00)	0.12 ( $\pm$ 0.07)
			CL	0.49 ( $\pm$ 0.13)	0.01 ( $\pm$ 0.01)	0.01 ( $\pm$ 0.01)	0.01 ( $\pm$ 0.01)	0.07 ( $\pm$ 0.08)	0.00 ( $\pm$ 0.00)	0.42 ( $\pm$ 0.04)
CH07 0–1	1	94	CL	0.25	0.02	0.06	0.02	0.32	0.01	0.32

<sup>1</sup> summed GDGTs VI, VII and VIII.

<sup>2</sup> n.m. = not measured.

<sup>3</sup>  $f$ [GDGT] represents fractional abundances of total GDGTs.

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**Table 2.** Fluxes and indices of GDGTs in settling particles throughout the estimated deposition periods of CH10 and CH07 surface sediments. GDGT data from 18 November 2006–1 December 2007 is collated from Sinninghe Damsté et al. (2009) and from 31 December 2007–31 August 2010 is collated from Buckles et al. (2014a).

Settling particles	Deployment date	Collection date	C <sub>org</sub> (%)	Bulk flux (g m <sup>-2</sup> day <sup>-1</sup> )	GDGT-I (μg m <sup>-2</sup> day <sup>-1</sup> )		GDGT-V (μg m <sup>-2</sup> day <sup>-1</sup> )		Σ[brGDGTs] <sup>1</sup> (μg m <sup>-2</sup> day <sup>-1</sup> )		BIT		MBT		DC	
					IPL	CL	IPL	CL	IPL	CL	IPL	CL	IPL	CL	IPL	CL
2008–2010 (CH10)	30 Jan 2008	30 Jan 2010	12.5	1.3	1.3	0.3	0.1	0.2	0.4	1.6	0.86	0.86	0.29	0.28	0.17	0.20
2006–2007 (CH07)	18 Nov 2006	24 Aug 2007	18.1	1.5	n.m. <sup>2</sup>	0.4	n.m.	2.3	n.m.	2.3	n.m.	0.50	n.m.	0.25	n.m.	0.18

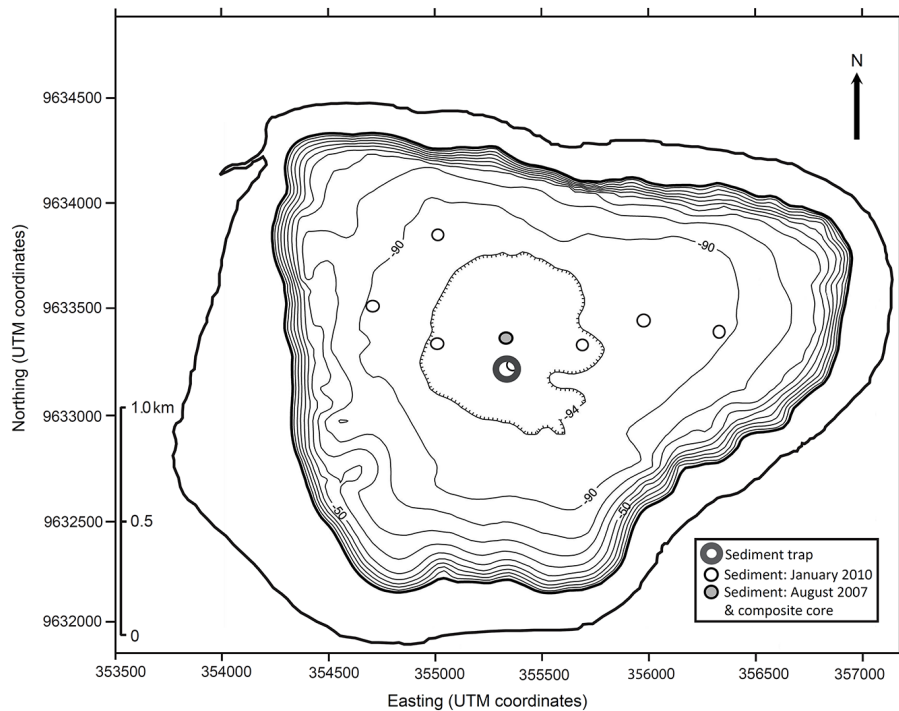
<sup>1</sup> summed GDGTs VI(b, c), VII(b, c) & VIII(b, c).

<sup>2</sup> n.m. = not measured.

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**Figure 1.** Map of Lake Challa with bathymetry and sampling sites indicated; scale is in the Universal Transverse Mercator (UTM) global coordinate system. The outermost bold black line denotes the catchment area boundary.

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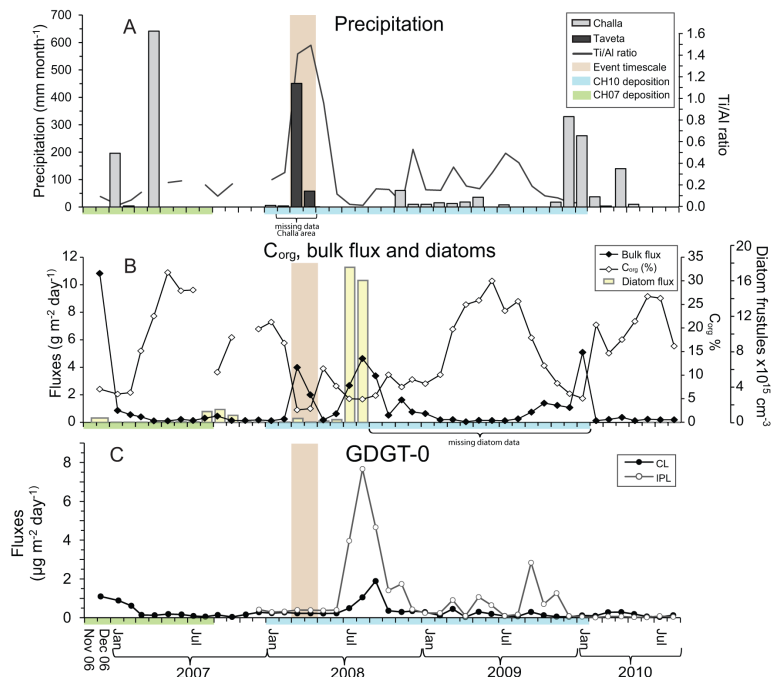
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**Figure 2.** Fluxes and GDGT parameters of approximately monthly sediment-trap samples of settling particles, from 18 November 2006 to 31 August 2010. **(a)** Monthly precipitation at a Kenya government agricultural station immediately north of Lake Challa (cf. Buckles et al., 2014a) and the Ti/Al ratios of settling mineral particles (from Wolff et al., 2014). **(b)** Fluxes of bulk sedimenting particles and diatoms, and bulk percentage organic carbon (%  $C_{org}$ ) content. Fluxes of **(c)** IPL and CL GDGT-0, **(d)** IPL and CL brGDGTs **(e)** IPL and CL crenarchaeol (GDGT-V) and **(f)** IPL and CL BIT index, with dashed horizontal lines representing the CL BIT indices of surface sediment deposited over those time periods. In **(c-f)**, data from 18 November 2006 to 1 December 2007 is by Sinninghe Damsté et al. (2009) and data from the following months until 31 August 2010 are by Buckles et al. (2014a).

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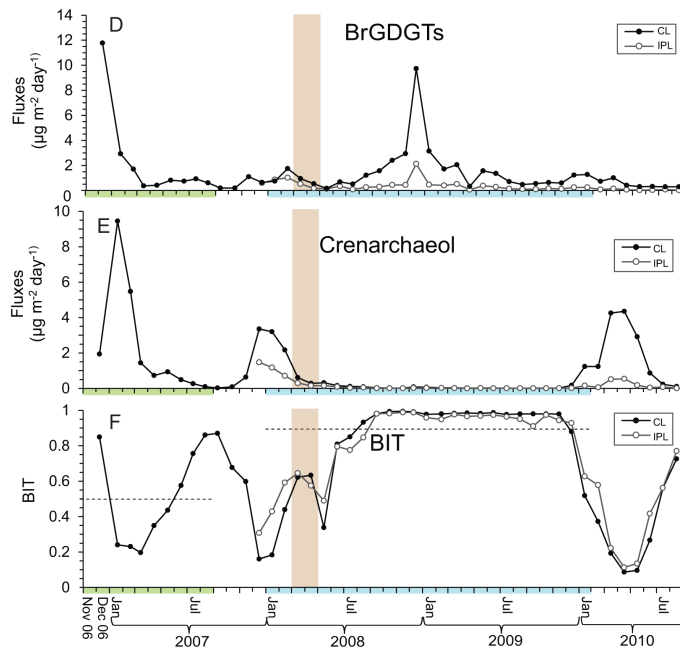


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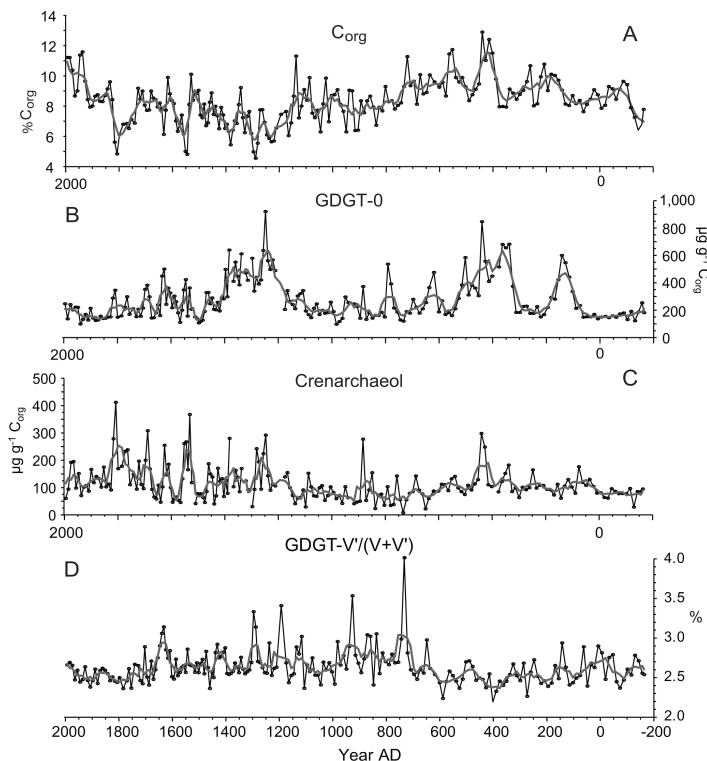
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**Figure 3.** Bulk and GDGT parameters in the 213 cm long Lake Challa sediment sequence against years AD. **(a)** %  $C_{org}$ , **(b)** GDGT-0 abundance, **(c)** crenarchaeol (GDGT-V) abundance, **(d)** the percentage of crenarchaeol regioisomer with respect to crenarchaeol, **(e)** summed brGDGT abundance, and **(f)** BIT index. Points connected by a thin line represent raw data and the thicker black lines denote 5-point running averages. Missing data for GDGT abundances are due to gaps in %  $C_{org}$  measurement. **(g)** Comparison of 4 averaged 1 cm sections of the BIT index from the 213 cm core against integrated 4 cm sections of the same core analysed earlier by Verschuren et al. (2009).

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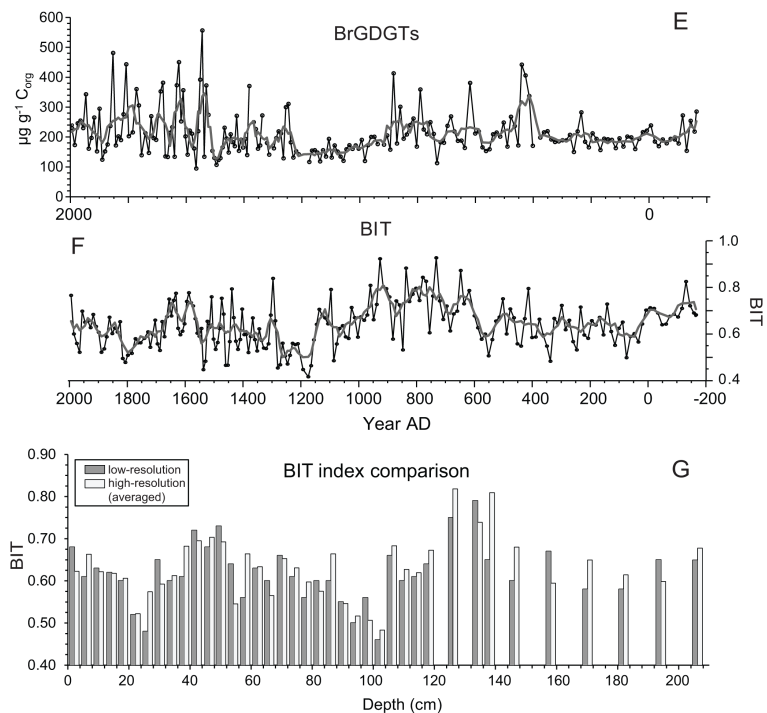
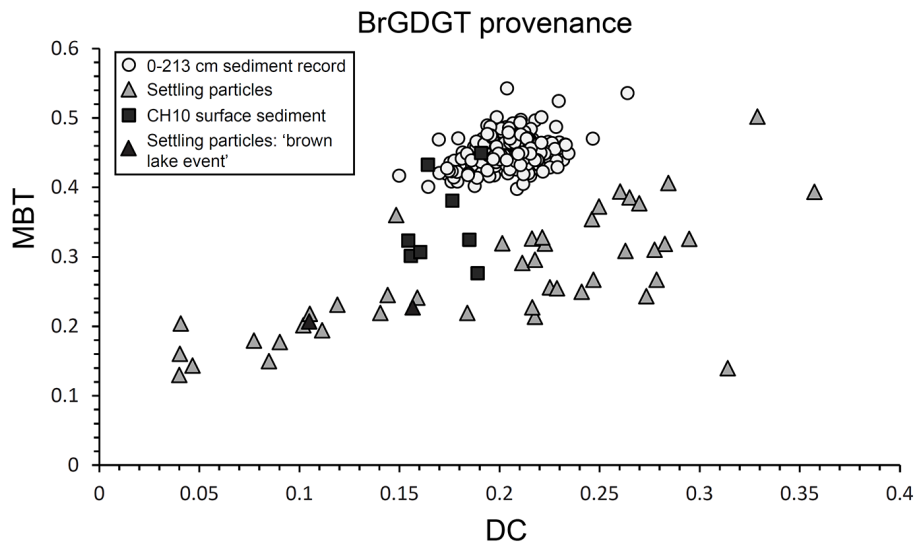


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**Figure 4.** MBT vs. DC plot for the 0–213 cm sediment record (circles), for CH10 surface sediments (squares) from Buckles et al. (2014a), and settling particles (triangles; data from 18 November 2006 to 1 December 2007 are by Sinninghe Damsté et al., 2009 and data from the following months until 31 August 2010 are by Buckles et al., 2014a). Black triangles represent settling particles from March and April 2008, when the lake was reported turning brown.

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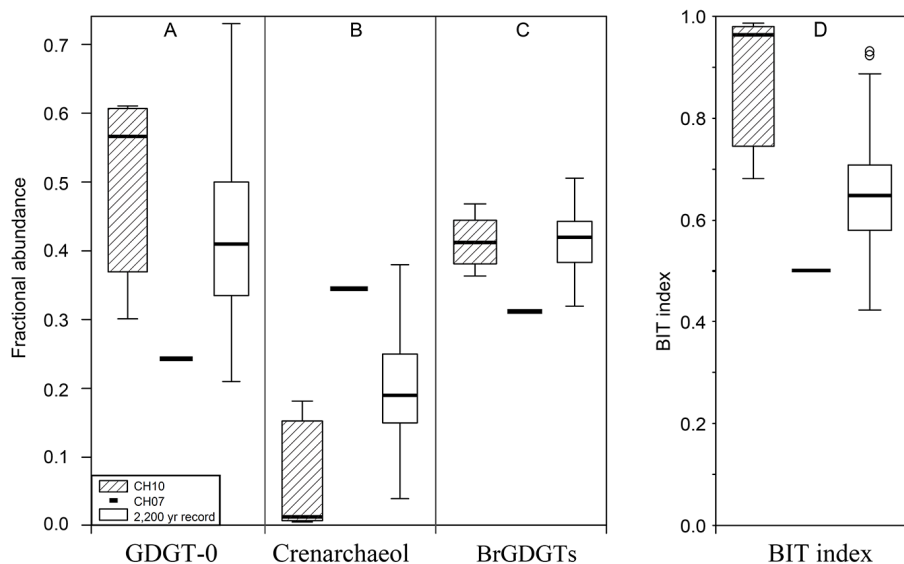
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**Figure 5.** Boxplot of fractional abundances of **(a)** CL GDGT-0, **(b)** CL crenarchaeol, **(c)** CL summed brGDGTs and **(d)** of the BIT index, for respectively surface sediments CH10 ( $n = 7$ , 0–1 cm depth) collated from Buckles et al. (2014a), CH07 collated from Sinninghe Damsté et al. (2009) and the 2200 year sediment record (0–213 cm depth at 1 cm resolution, where 1 cm represents on average 10.4 years of deposition). Note that surface sediments represent 2–3 years of deposition. The box corresponds to the interquartile range and the whiskers extend to 1.5 times the length of the box (unless the full range of data is smaller than this); outliers are defined here as being outside the maximum extent of the whiskers. The black horizontal line inside the box represents the median.

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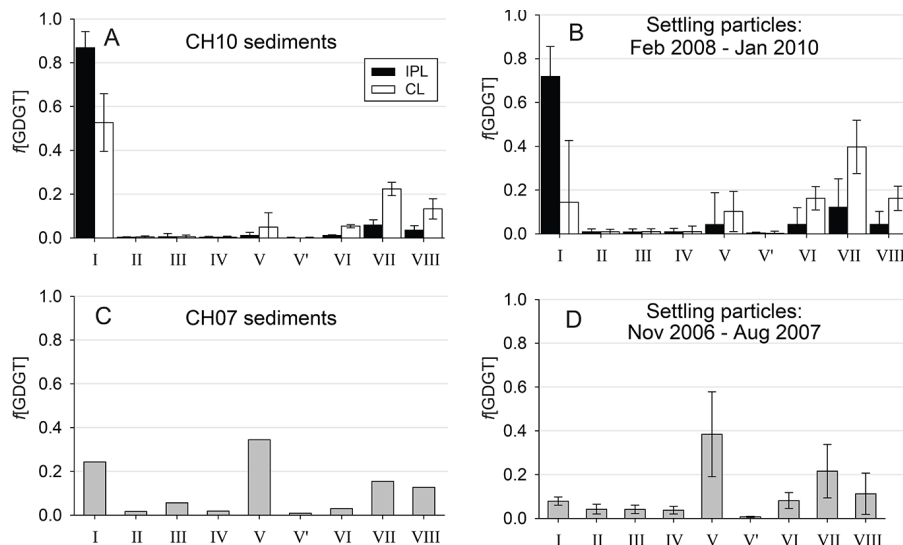
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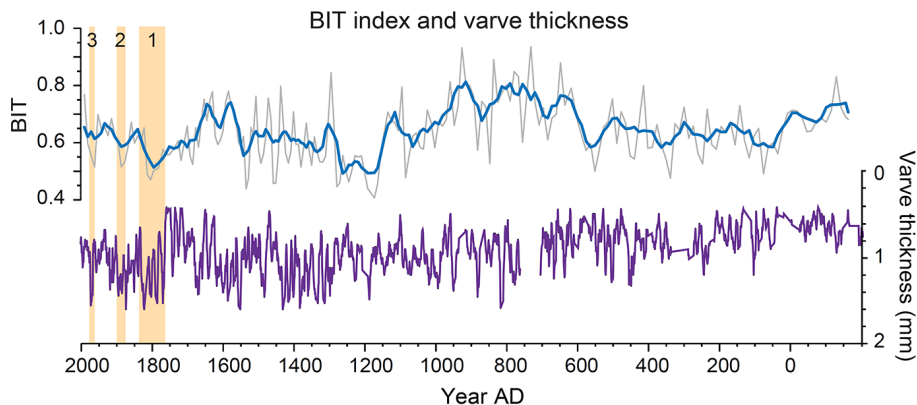
**Figure 6.** IPL and CL GDGT distributions from surface sediments **(a)** CH10 from Buckles et al. (2014a) with **(b)** corresponding weighted average IPL and CL GDGT distributions from summed fluxes of settling particles between 30 January 2008 and 30 January 2010, also from Buckles et al. (2014a). **(c)** CL GDGT distributions from surface sediment CH07 from Sinninghe Damsté et al. (2009) and **(d)** corresponding weighted average CL GDGT distributions from summed fluxes of settling particles between 18 November 2006 and 24 August 2007.

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**Figure 7.** (a) BIT index in the 2200 year sediment record, where grey lines represent raw data and the blue line a 5-point running average. (b) A 7-point running average of varve thickness from Wolff et al. (2011). Orange shaded bars highlight recent periods of drought in East Africa: (1) 1760–1840 (Verschuren et al., 2000; Bessems et al., 2008), (2) 1880–1900 (Nicholson et al., 2012), and (3) spread over drought years 1964–1967, 1972–1973 and 1975–1976 (Verschuren et al., 1999; Verschuren, 2004).

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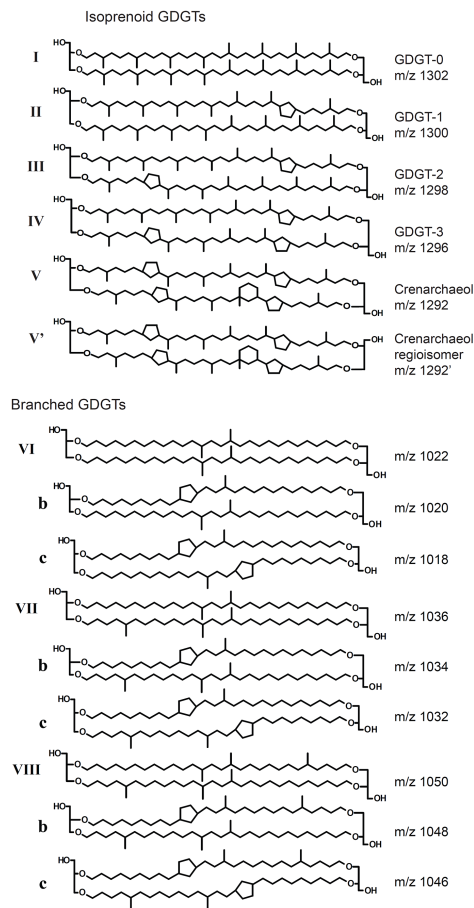
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**Figure A1.** Key to GDGT structures.

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