Rebuttal (response in italic font)

Firstly, we would like to thank the referees very much for their time and constructive comments, which will certainly help to improve the manuscript. We are happy to read that both reviewers consider our work to be ‘a highly valuable contribution’ and that the effort of collecting a long term data series is acknowledged: ‘the data are extremely informative’ (reviewer 1) and ‘data derived from such a longterm monitoring program are very rare but remain necessary to understand the sedimentation processes in lake systems’ (reviewer 2). The referees consider our manuscript a ‘highly worthy contribution’ (reviewer 1), ‘ultimately worthy of publication’ (reviewer 2). However, some important concerns are raised as well, which we would like to address below. Please note that are presented in italics and our responses tot he comments of the reviewer comments are presented in italics text.

Reviewer 1

- Summary: Application of the BIT index as a paleo-precipitation proxy at Lake Challa, East Africa has sparked a highly interesting and complex debate over the past several years. The proxy seems to say something important about hydroclimate on long timescales, but how it works remains a mystery. In this manuscript, Buckles et al. present a new analysis of branched GDGTs in sediment trap and sediment core data in order to clarify how the proxy works. The dataset and the analysis are a highly valuable contribution, and the paper is generally well-written (although the ‘Results’ section needs to be revised for a more general, non-organic geochemistry audience; see my comments below). However, my main issue is that the conclusions are strongly biased towards supporting the BIT index as a robust and consistent paleo-precipitation proxy in this setting, when the authors’ own data show that it is, in fact, not. I strongly recommend that the authors remove the highly speculative conclusions toward the end of the paper (more information below) and focus, instead, on what their dataset does in fact show. The data are extremely informative and they add more pieces to the puzzle, but they do not come close to ‘proving’ that the BIT index “is a reliable precipitation proxy, at least in the Lake Challa system and on (multi-)decadal and longer timescales” (Conclusion, pg 1199). The authors should provide a more honest discussion of what we still need to learn in order to understand how the BIT index reflects hydroclimate at Lake Challa.

We understand that our conclusions are bold, although we believe them to be fully substantiated by our data. To accommodate concerns expressed by the reviewer, we will extend discussion of the 2,200 year record to include further interactions of the BIT index record with known climate events (see section 4.5). The primary conclusion is now: “We conclude that the BIT index of Lake Challa sediments reflects the amount of monsoon precipitation indirectly, as is also the case with varve thickness (Wolff et al. 2011) and many other climate proxies extracted from lake sediments. Prior to application elsewhere, we strongly recommend ascertaining the local situation of lacustrine brGDGT production and of variables affecting the productivity of Thaumarchaeota.” Wolff et al. (2011) observed that varve thickness in Lake Challa sediments is primarily controlled by the thickness of the diatom layer deposited during austral winter, which is determined by the strength of seasonal deep mixing and therefore depends on wind stress during the austral winter. Inter-annual *variation in local wind stress is inversely related to variability in precipitation associated with the ENSO cycle. We propose that the BIT index is primarily controlled by variation in the annual Thaumarchaeota bloom during austral summer, which is suppressed when excess nutrient input associated with occasional rainfall-driven soil-erosion events result in these Thaumarchaeota being outcompeted by nitrifying bacteria. The BIT index therefore can be considered to integrate the frequency of soil erosion events over time, which in the extremely seasonal and semi-arid tropical environment of Lake Challa is most likely proportional to total annual monsoon precipitation. However, since the mean frequency of such erosion events appears low at the (inter-) annual time scale, the positive relationship between BIT and rainfall will not manifest itself unless integrated over multi-decadal and longer time scales.*

- A note on the writing style: The topic of this paper (assessing the validity of the BIT index as a paleo-precipitation proxy in Lake Challa) is highly relevant to the broader paleoclimate community. However, the Results section (and parts of the Discussion prior to section 4.4) is written in such a way that it would be much better suited to an organic geochemistry journal. The authors should revise the paper to be more accessible to a non-organic geochemistry audience. For example, rather than focus on the technical details of every measurement they performed, the authors should give the results alongside a discussion of why these measurements were made in the first place. What is the importance of looking at core lipids and intact polar lipids? Why would someone do this? What new information does this provide? Why are there multiple indices for brGDGTs, and why bother comparing BIT to, e.g., MBT? These may be obvious to an organic geochemist, but it is totally unfamiliar territory to most Climate of the Past readers.

We agree with the reviewer that our paper presents an opportunity to explain the analytical approaches of state-of-the-art organic geochemistry to the Climate of the Past readership, and we have included some further explanation in the text to render it more accessible to non-geochemists.

- Detailed comments: Generally, I find the arguments presented in this section to be rather biased towards "support[ing] use of the BIT index as hydroclimatic proxy in this system (Verschuren et al., 2009)." It should not be assumed a-priori that the interpretation presented in Verschuren et al., 2009 is correct and should be supported, somehow, by the modern data.

The present study does not intend to 'support' the Verschuren et al. (2009) interpretation of longterm BIT index variation in the 25,000-year Lake Challa record. That interpretation is robust, on account of 1) it showing clear signatures of generally recognised widespread climate-change anomalies (such as Younger Dryas drought) at exactly the right time; 2) to the extent allowed by its lower (multi-century scale) resolution, it also shows clear signatures of the known regional climate anomalies, again at the right time; 3) it is in agreement with a second and fully independent hydroclimatic proxy, namely the low-resolution lake-level reconstruction based on seismic-reflection data (Moernaut et al., 2010); and 4) its relatively unique characteristics can be explained by reference to climate dynamics specific to the region of equatorial Africa beyond Atlantic Ocean influence. Therefore, that the Challa BIT index reflects long-term variability in that region's monsoon rainfall is no less certain than *for any other, traditional or more novel, hydroclimate proxy. The purpose of the present study is to find a mechanistic explanation for why the BIT index in Lake Challa sediments reflects monsoon rainfall, given the recent finding that branched GDGTs (constituents of the BIT index) found in Lake Challa sediments primarily originate from production in the water column itself, rather than from the surrounding soils.*

In fact, the data presented here show that BIT is an unreliable and inconsistent hydroclimate proxy in this system. It does seem to respond to ecological changes in the lacustrine system following the early- 2008 erosion event, via suppression of Crenarchaeol production. However, this rainfall event was less intense (if Challa and Taveta are comparable) than a heavy rainfall event in early 2007, but that early 2007 event was not detected by BIT. Therefore, BIT does not in fact respond in a consistent manner to extreme rainfall events. If, in fact, the BIT index does respond to rainfall events that follow severe drought (as the authors postulate for the early-2008 event), then it is an indicator of erosion extremes, not precipitation extremes or seasonal (monsoonal) precipitation.

We do not argue that the BIT index is an one-on-one indicator of precipitation extremes or of seasonal precipitation amount. If it were, it could be used to trace rainfall variation at (sub-) annual to interannual time scales, which we repeatedly state it does not. As suggested by the reviewer, it is indeed an indicator of 'extreme' erosion events, which in this semi-arid tropical region have a threshold relationship with rainfall extremes. We have adjusted the text to better clarify this (lines 462-464). Additionally, please note that our measured precipitation record originates from local hydrological monitoring and therefore was available only as summed precipitation per month. As the creek in the NW corner of the lake is only activated in periods of intense precipitation, it might represent the difference between all the month's rain falling in one day or spread over the full month.

The proxy of course could be detecting high-amplitude variability. It is very possible that in the 25,000 year record, the BIT index is not recording regular monsoonal rainfall but rather extreme flooding events that also follow extreme droughts.

That is exactly our proposition. However, it is important to note that over the past 25,000 years, the Lake Challa area has always experienced a semi-arid tropical climate with high propensity for both extreme drought episodes and extreme precipitation events. We can thus surmise that, integrated over time, the impact of extreme erosion events on the aquatic ecosystem of Lake Challa is proportional to longer-term trends in total rainfall.

The BIT signals get smoothed out and even shifted in time relative to the varve record, as is shown in Figure 3G.

We do not know which figure the reviewer is referring to, since the former Figure 3G (Figure 7 in the revised version) shows only a comparison between two sets of BIT index data. He/she may refer to the comparison between BIT index and varvethickness in Figure 7 (now Figure 8), but if so we do not agree with his/her assessment that BIT is shifted in time relative to varve thickness. These are two (largely) independent hydroclimatic proxies, with a similarly complex but different relationship with climate as the ultimate driver of a major part of their *variation through time. This allows the existence of proxy-specific and time-scale dependent biases, and hence there is no reason why long-term trends in these two proxy records should look exactly the same, or why there would be a systematic phase shift between them. We have extended the discussion on the comparison of the varve and BIT records in section 4.5.*

Finally, the authors' conclusions that the BIT index may be reliable on decadal timescales even if it is not reliable on interannual timescales seems highly over-speculative. Conveniently, we do not have decades' worth of modern data to disprove this. But in fact, even in this multi-year dataset, the authors have only one single event on which this interpretation is based.

The importance of rare events in long-term system dynamics is a common element in most geological and ecological processes. As mentioned earlier in our response, we propose the application of the BIT index in Lake Challa solely on multi-decadal or longer timescales since we believe this allows for a low frequency of soil erosion events. In our multi-year dataset, the marked perturbation of the microbiological community in the suboxic water column (as measured in settling particles) for almost two years of a nearly four year dataset is clear and the soil erosion event is the only identifiable cause, despite the extensive ongoing monitoring of the lake, its water column and analysis of sediments that extend the 'status quo' GDGT composition to cover more than 6 years of deposition. The relatively low frequency but long-lasting effect of this event directly supports the conclusion that inter-annual timescales would be too biased by the low frequency of these events, but multi-decadal timescales would integrate these signals. Our trust in the reliability of this rainfall proxy is enhanced by the congruence of our resulting 2,200-year rainfall reconstruction with established (though still relatively scarce) knowledge of the wider region's hydroclimatic history, including three historically documented episodes of prolonged drought over the past 200 years (see section 4.5). As the reviewer will agree, very few lake-based hydroclimatic proxies from any location in the world can claim this level of success. We, therefore, respectfully believe that our results represent a significant step forward in this field.

These findings are important for continuing to develop the interpretation of the Lake Challa BIT record. The authors should recognize that the interpretation of this proxy in this setting will continue to develop through time with new data, and it may even be revised quite thoroughly. Such is the purpose of collecting modern data to inform a proxy.

Our multi-year maintenance of a multi-parameter monthly monitoring programme on a small and remote African crater lake, at considerable time, logistic and financial investment, ought to be sufficient demonstration that we are aware of this and willing to meet the challenge.

The BIT index at Lake Challa does seem to say something about precipitation and erosion on long timescales. However, at this stage it is unclear what this proxy is telling us, and why it seems to work. This paper is an important first step, and additional modern observations will continue to clarify and develop the interpretation of this proxy and its application in other settings. However, I feel that a more laudable approach would be for the authors to explain what they have found and to honestly assess what is still not well-understood. They do not need to 'solve' the BIT proxy in this paper in order for the 25,000 year record to still be useful. In fact I feel the over-speculation weakens the overall findings of the paper, which in themselves are very interesting and a highly worthy contribution to the paleoclimate community.

We thank the reviewer for this appreciation of our work, and are sorry if we gave the erroneous impression that our primary aim is to 'solve' the BIT proxy. Our own publication record (Sinninghe Damsté et al., 2009; Sinninghe Damsté et al., 2012; Buckles et al., 2014a) should make clear that we have dissected the issue from all possible angles and we ourselves have published data casting initial doubt on the reliability of BIT index as rainfall proxy in this system. However, in combination with this previous research on the BIT index and its constituent compounds in Lake Challa (Sinninghe Damsté et al., 2009; Sinninghe Damsté et al., 2012; Buckles et al., 2014a) we believe that we now have a relatively comprehensive idea of the mechanisms that control its variation in Lake Challa. Although quantifying the relative influence of these mechanisms is beyond the scope of this paper and potentially impossible without multi-decadal field data, it does not diminish the congruence of our resulting 2,200-year rainfall reconstruction with established knowledge of the wider region's hydroclimatic history (see section 4.5).

- Line-by-line comments:

Pg 1180: Make it clear that Crenarchaeol = GDGT V

We have carefully defined the GDGT nomenclature (Lines 156-157) and altered Figure A1 (now Appendix) to indicate this. The legend tot he appendix gives a clear explanation tot he used nomenclature. We have also carefully revised the tekst to make this fully consistent.

Lines 5-10: Please include a figure with the age model. In the supplementary material please provide the ^{14}C AMS dates and their 1- and 2-sigma uncertainties.

Both the ^{14}C- and varve-based age models have been published previously (Blaauw et al., 2011; Wolff et al., 2011) and we refer to these previous publications, where details of these age models and associated discussion are readily available. We expanded section section 2.2. to provide more details on the age model.

Pg 1185, Equation 3: Define DC?

DC means degree of cyclisation; this is now clarified in the revised manuscript (Line 182).

Pg 1186, Line 5: Why these 'general guidelines?' How were these cut-offs chosen? 0.5 is very low to be considered "strongly correlated", especially since these are r-values and not r^2.

Dancey and Reidy (2004) recommend the following characterisations for the interpretation of correlations and Pearson's r:

Correlation coefficient	***Strength of correlation***
1	*Perfect*
0.7-0.9	*Strong*
0.4-0.6	*Moderate*

0.1-0.3	*Weak*
0	*Zero*

Our cut-offs were chosen based on the above (now mentioned in the text, Lines 202-210), with slightly lower boundary conditions enforced to reflect the assumption of a large number of confounding factors in the data (although our boundary conditions are still well within the accepted range). These factors include (but are not limited to):

- A relatively large number of GDGT measurements with zero or near-zero values;

- Potential small time offsets between the varve record and the geochemical record;

- The measurement of %Corg at two–centimetre intervals, while all other parameters are per centimetre or less;

- Those affecting the abundance and distribution of GDGTs such as different bacterial producers of branched GDGTs, limited knowledge of the ecology of GDGT producers in Lake Challa and their response to pH/temperature, changes in production/depth of production and allochthonous influxes over time, etc.; and

- The relatively low number of data points available (208).

Additionally, a slight reclassification of the strength of the correlation coefficients would have no impact on our discussion of the data or our conclusions. Each coefficient is interpreted by comparison with the rest of the dataset and their classification is by its nature arbitrary. This is now mentioned in the manuscript.

Pg 1186, What is SD? standard deviation? Please define Pg 1187: Does r=0.67 for [brGDGT] with both crenarchaeol and its regioisomer? Please confirm

SD is standard deviation and r=0.67 for both crenarchaeol and its regioisomer (see Table S2). This is clarified in the revised manuscript.

Pg 1189 line 20: Do the gravity core samples have any actual age control points at the top? Otherwise, it is a strong assumption to say you know their timing down to the month.

Sinninghe Damsté et al. (2009) and Blaauw et al. (2011) state that the uncompacted top centimetre of sediment represents approximately 2 years of deposition. This is based on tie points in the visual fine lamination and magnetic susceptibility profiles of multiple gravity cores collected between 2003 and 2011, and confirmed by 210Pb-dating of a gravity core collected in 1999; see Blaauw et al. (2011) for details. We simply use this data to demonstrate that the surface sediments collected do indeed reflect the composition of the descending particles collected in the sediment trap over that time period, so the assumption is tested and is not egregious. We expanded section section 2.2. to provide more details on the age model.

Figure 6: Confused. I do not see GDGT-0 on this figure. I am only now realizing that GDGT-0 is the same as GDGT-I in the Appendix. This is very confusing. Can you please make this terminology very clear, repeating it throughout the paper so that the reader can follow.

We understand that the nomenclature is somewhat confusing and have clarified this in the revised tekst as stated earlier.

Figure 6: Why no error bars on CH07?

CH07 consists of two samples from 0.0-0.5 cm and 0.5-1.0 cm depth. For our purposes, we integrate these two samples (summed abundance rather than average) and take it to represent one sample of 0.0-1.0 cm depth. As it represents one sample, it does not have error bars; please also see Table S4.

Pg 1193 line 12: Why would it be true that brGDGTs/crenarchaeol are correlated, and hence brGDGT producers are heterotrophic bacteria? This connection is not clear, please explain.

Based on analysis by Buckles et al. (2014), branched GDGTs and crenarchaeol are primarily exported to sediments from the suboxic zone of Lake Challa. The hypothesis is that branched GDGT-producing bacteria are involved in the degradation of organic matter produced by diatom blooms, while Thaumarchaeota are ammonium-oxidising archaea and therefore thrive on the degradation products of the diatom bloom. This would explain the correlation of crenarchaeol with branched GDGTs and is clarified in the revised tekst.

Figure 2a: Because of the missing Challa precipitation data it is difficult to compare the magnitude of the precipitation that resulted in the erosion event with the other precipitation in the records. Please plot Taveta rainfall for the other events as well on Fig 2a, so that the comparison may be made.

We have replaced the precipitation data by monthly precipitation over the 0.5° × 0.5° grid which includes Lake Challa, from the Global Precipitation Climatology Centre data set, version 6 as suggested by referee 2.

Pg 1194, line 19: The onset of the principal rainy season cannot be the only reason for erosion, because the other years' principal rainy seasons did not see similar erosion events.

This is a chronological description of events. Based on accounts of local fishermen the allochthonous influx to the lake was triggered by the onset of the principal rainy season as described, but not all principal rainy seasons will necessarily trigger a soil erosion event.

Pg 1197: "Since stronger austral-winter winds are associated with a weak southeasterly monsoon compromising the main rain season during March–May " Please provide citation for this mechanism.

We refer now to Wolff et al. (2011; Science (Line 477).

Reviewer 2

The aim of this study is to test the BIT index (ratio describing the proportion of branched GDGTs, of soil origin, versus isoprenoids GDGTs, of aquatic origin) as a proxy of precipitation in tropical Africa. Buckles et al. used data from a sediment trap, soils, and lake sediments combined to climate data to evaluate the BIT index in the Lake Challa area (Kenya/Tanzania). They found that brGDGTs were also produced in the lake water column and that the BIT index in Lake Challa sediments reflected the crenarchaeol abundance, rather than brGDGT abundance thus complicating the original interpretation of this proxy. Here, Buckles et al. proposed that pulses of Thaumarchaeota production during the driest and windiest years mostly control the BIT index in a lake system where allochthonous sedimentation is dwarfed by autochthonous sedimentation. I found their interpretation realistic for the modern/recent lake sediments but it is also possible that the proposed mechanism varied for the older sediments (cf. the 25,000 yr record). For example, the high BIT index (of 1) during the early Holocene may also be related to the increase of brGDGTs derived from soils (this period was significantly wetter compared to the present-day conditions). The data presented in this study are highly valuable, particularly since data derived from such a long-term monitoring program are very rare but remain necessary to understand the sedimentation processes in lake systems. Overall I found this paper interesting to read and ultimately worth for publication at Climate of the Past after substantial adjustments. Like the authors, I think that the BIT index remains a potential good proxy for paleohydrology, although the monitoring data presented here suggests that the behavior of the GDGTs and the exact meaning of this proxy remain still elusive in small lake systems. I disagree with the authors that their study ‘validates’ the use of the BIT index in such environments since the new mechanism they promote (i.e. high in situ brGDGT production combined to a production of crenarchaeol triggered by precipitation in the lake’s catchment) to explain this proxy strongly differ with the initial one (i.e. soil versus aquatic origin of the GDGTs).

Moreover, they do not provide a way to evaluate which mechanisms (soil-derived brGDGTs versus in situ production of brGDGTs) can control the BIT index in the sediments. This would be necessary for an unambiguous interpretation of the sedimentary BIT index. For example, when looking at different time periods, both mechanisms could operate in the same lake system and their impact on the BIT index cannot be considered as identical (until proven). The authors should provide here a more balanced discussion and importantly they should also provide more ways to help future understanding of this proxy. Below are other important points that, in my opinion, need to be fully clarified prior to publication.

We agree that the mechanism controlling BIT variation may vary for the older sediments (cf. the 25,000 year record), but this problem can hardly be considered unique to this particular proxy. In fact, many paleoenvironmental proxies (even some supposedly robust traditional proxies) remain unvalidated even in the modern-day system. Sinninghe Damsté et al. (2012) already discuss the longer BIT-index record in detail and therefore this is out of scope for our paper, although we do refer to this previous research in our discussion. Additionally, Buckles et al. (2014) demonstrate that the signature of modern-day branched GDGT influxes to Lake Challa is not only indistinguishable in the BIT index of settling particles in sediment-trap samples, but also that their distributions do not shift towards those found in soils. This is *despite the lake reportedly 'turning brown' and associated changes in the Ti/Al ratio as described by Wolff (2010). Please also note that even with 40% more rainfall, the Lake Challa climate regime would will be semi-arid with similar seasonal and inter-annual variability of monsoon precipitation. We thus believe that despite significantly wetter conditions, the influx of soil-derived branched GDGTs is not the primary mechanism for the increase in the sedimentary BIT index. The new mechanism we propose differs so markedly from the initial mechanism due to the now widespread knowledge that branched GDGTs can be produced in substantial amounts in lakes (e.g., Tierney and Russell, 2009; Tierney et al., 2010; Loomis et al., 2011), in addition to further research in Lake Challa specifically (Sinninghe Damsté et al., 2012; Buckles et al., 2014a). Before the sediment trap time series was expanded, Sinninghe Damsté et al. (2009) noted that influxes of branched GDGTs to the sediment trap appeared to correspond with the precipitation regime; however, extending this time series and combining it with additional analysis of intact polar lipids ('living' branched GDGTs) showed that branched GDGTs in sediments were primarily derived from the water column (Buckles et al., 2014a). This emphasises the value of long-term monitoring. However, please note that both mechanisms rely on influxes of soil to the lake affecting the sedimentary BIT index, either directly or indirectly.*

Point (1). The modern data: settling particles.

Most of the sediment trap data presented by the authors derived from Sinninghe Damsté et al. (2009) and Buckles et al. (2014). The authors mentioned here that they "report additional results for GDGTs I to IV present in these samples". It is not clear reading this manuscript what is really new and what is derived from the former studies on this site. This should be better defined. The authors should emphasize more their discussion on the new findings.

In the results section, we specify which results have been presented elsewhere and collated within this manuscript, and which are new. The results focus on describing the new data: GDGT concentrations and GDGT-based indices from the 2,200-year sediment record and isoprenoid GDGTs from surface sediments and settling particles (excluding crenarchaeol and its regioisomer).

I wonder if the GDGTs data (and their indexes) from the sediment trap are contemporaneous with the weather events presented for comparison (temperature, precipitation: : :). Here are some open questions that should be discussed more in detail in this manuscript: - What is the estimated residence time of the GDGTs in both the Lake Challa water column and its watershed?

This is discussed in detail by Buckles et al. (2014). We refer to this research where relevant.

- What is the velocity of settling particles within the water column of the Lake Challa? Does this velocity remain constant during a seasonal cycle? Looking at the Wolff et al. (2014) data, it seems that there is a systematic lag of 2-4 months between the deposition of Ti and the preceding main peak of precipitation. Does that also apply to the GDGTs?

As shown in our Figure 2, the peak in Ti/Al ratio is concurrent with the peak in precipitation. The velocity of sinking particles would depend on their size; GDGTs from the upper water column are likely to be exported to sediments relatively rapidly (Buckles et al., 2014). However, the majority of GDGTs exported to sediments are produced in the vicinity of the sediment trap and thus their time to reach the sediment trap would likely be even shorter than that of soil-derived materials.

Point (2). The paleo-record: comparison of the BIT index and varve thickness during the last 2000 yr. The authors spent a large part of their manuscript to discuss the modern data, while a smaller part of it is devoted to the discussion of the paleo-record. The balance between the two parts could be improved.

We have substantially extended the discussion of the 2,200-year record to include further comparisons of the BIT index record with regional climate records (see section 4.5). However, our focus is on the discussion of modern data and on the validation of the proxy rather than interpreting regional climate dynamics as expressed by the BIT index record, which cannot be done without a full review of regional climate records with their respective merits and defects. The reviewer will agree that this requires a different kind of study.

Figure 7 shows the direct comparison of the BIT index and varve thickness during the last 2000 yr. The authors used 5-point and 7-point running average for the BIT index and varve thickness data, respectively, which were sampled with a different resolution. Instead, I would advise them to resample the varve thickness data using the exact sampling resolution as for the BIT index and to show a time series of varve thickness (with the mean and standard deviation for each sample interval) that is directly comparable with the BIT index data. Then, the same running averages could also be overlaid above the two records. Correlation plots with significance level would be also valuable. Does the correlation vary in time? The authors suggested 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.” To validate their statement, they should calculate the correlation (and demonstrate that it is significant or not) between the BIT index and the varve data at different timescale (using for example a moving average or a band-pass filter): according to their statement, correlation should increase towards lower frequency variability of the BIT index. Buckles et al. compared their BIT index data with the total varve thickness. However, Wolff et al. (2011, 2014) demonstrated that “the total varve thickness is controlled by the thickness of the light layer: : : varve thickness mainly reflects the quantity of diatom frustules deposited during the dry season and in particular during April to September”. Wolff et al. (2014) noted that “varve thickness can be used as a proxy to reconstruct paleo wind variations during the dry season.” Thus the total varve thickness is not a direct proxy of precipitation. Instead, the small and organic dark varve layers in the sediments record monsoon precipitation (unfortunately biased by additional in situ lake precipitation products): “The darker layers represent the two rainy seasons (November to December and March to May) and the brief intervening dry season with amorphous organic matter derived from phytoplankton and calcite precipitation: : :” (Wolff et al. 2014). The authors should then also compare the BIT index data with the dark varve thickness data and provide a correlation plot as for the total varve thickness data. It would be important to know which varve thickness data (light or dark) provide the best correlation with the BIT index through time.

Wolff et al. (2011) proposed that varve thickness in Lake Challa sediments is primarily controlled by the layer of diatoms deposited during the austral winter, which is determined by the strength of seasonal deep mixing and therefore depends on wind stress during the austral winter. Wolff et al. (2011) also show that "El Niño (La Niña) events are associated with wetter (drier) conditions in East Africa and decreased (increased) surface wind speeds." The interpretation of the variation in annual varve thickness data in fact reflects ENSO variability, which is on a shorter timescale to the decadal resolution of our BIT index record. As such, they are not directly comparable by the method described by the reviewer as dark varve layers are deposited during the two rainy seasons but are not a proxy for monsoon precipitation. However, we have further explored correlations between the BIT index and varve thickness on various timescales, such as during the past 200 years, in our revised manuscript (see section 4.5).

Additionally, the reviewer suggests that the varve thickness data be resampled to the depth resolution of the BIT index for comparison. However, the depth axis is not independent of varve thickness, i.e. a different number of varves (and parts of varves) must be summed to achieve 1 cm resolution.

Other points:

- Although I am totally confident with the GDGTs data produced by the authors and the calculated BIT index for the different substrates presented in this study, I am not very confident with the precipitation data they used for comparison. Buckles et al. (2014) first showed the precipitation data and indicated 10 that it derived from a governmental agricultural station "immediately north of Lake Challa". In the current paper Buckles et al. showed the same data but discriminated the data from "Challa" and "Taveta". The time series looks weird, it is not seasonal but erratic, showing for example only one significant month of precipitation for 2007 with ca. 650 mm (extreme precipitation amount for March), and almost not a single drop of water during the rest of this year. What is even stranger is that other authors who also worked on the same material from the same sediment trap used other precipitation data for their comparisons: the record of Voi located 100 km to the East (Wolff et al., 2014). The Voi station is also not ideal since it is located too far away from the study site and may not have recorded some major events at Challa. Since the precipitation record from Voi show less gaps of precipitation (i.e. prolonged period of no precipitation) and since the mean annual precipitation is significantly lower in Voi compared to Taveta according to Sinninghe Damsté et al. (2009; Fig. 3f), the data presented here seem obviously incomplete/biased. According to the data of Voi (Wolff et al., 2014), there is no "long drought that stretched from May 2007 to February 2008 due to failure of the short rains in 2007" as Buckles et al. asserted. If the precipitation data derived from local ground-based stations have issues, the authors may check the remote sensing product for alternative solutions. For example, the data from the Tropical Rainfall Measuring Mission (TRMM) are easily available, and provide 3 hourly rainfall amounts with a spatial resolution of 5x5 km^2 from 1997 to 2015. I am not arguing that a more robust precipitation record would provide a complete mechanism for interpreting the complex BIT signal but this will certainly help the data interpretations.

This analysis is fair and the comment helpful. We have replaced the precipitation data by monthly precipitation over the 0.5° × 0.5° grid which includes Lake Challa, from the Global Precipitation Climatology Centre data set, version 6

- The nomenclatures of the GDGTs are not straightforward and will confuse many of the readers who are non-specialists. Referring to GDGT-0 for GDGT-I or to GDGT-1 for GDGT-II does not facilitate the understanding of complex ratios of molecules. It would be wise to state that this dual nomenclature exists at the beginning of this manuscript and to stick to one or the other all over the manuscript.

As stated before we have corrected this (Line 50-51) and Appendix.

- I also found highly relevant and valuable the comments provided by the first reviewer.

Agreed.

Interannual and (multi-)decadal variability in the sedimentary BIT index of Lake Challa, East Africa over the past 2,200 years: Assessment of the precipitation proxy

Laura K. Buckles[a], Dirk Verschuren[b], Johan W. H. Weijers[a]*, Christine Cocquyt[c], Maarten Blaauw[d] and Jaap S. Sinninghe Damsté[a,d]†

[a] University of Utrecht, Faculty of Geosciences, P.O. Box 80.021, 3508 TA Utrecht, The Netherlands

[b] Limnology Unit, Department of Biology, Ghent University, K. L. Ledeganckstraat 35, B-9000 Gent, Belgium

[c] Botanic Garden Meise, Nieuwelaan 38, B-1860 Meise, Belgium

[d] School of Geography, Archaeology and Palaeoecology, Queen's University Belfast, Elmwood Avenue, Belfast BT7 1NN, UK

[e] NIOZ Royal Netherlands Institute for Sea Research, Department of Marine Organic Biogeochemistry, P.O. Box 59, 1790 AB Den Burg, Texel, The Netherlands

* Present address: Shell Global Solutions International B.V., Kessler Park 1, 2288 GS Rijswijk, The Netherlands

†To whom correspondence should be addressed. Jaap S. Sinninghe Damsté, NIOZ Royal Netherlands Institute for Sea Research, Department of Marine Organic Biogeochemistry, P.O. Box 59, 1790 AB Den Burg, Texel, The Netherlands. E-mail: jaap.damste@nioz.nl; phone number: +31 222-369550; and fax number: +31 222-319674.

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ABSTRACT

The branched vs. isoprenoid index of tetraethers (BIT index) is based on the relative abundance of branched tetraether lipids (brGDGTs) and the isoprenoidal GDGT crenarchaeol. In Lake Challa sediments the BIT index has been applied as a proxy for local monsoon precipitation on the assumption that the primary source of brGDGTs is soil washed in from the lake's catchment. Since then, microbial production within the water column has been identified as the primary source of brGDGTs in Lake Challa sediments, meaning that either an alternative mechanism links BIT index variation with rainfall or that the proxy's application must be reconsidered. Here we investigate GDGT concentrations and BIT index variation in Lake Challa sediments at a decadal resolution over the past 2,200 years, in combination with GDGT data from 45 monthly sediment-trap samples and a chronosequence of profundal surface sediments.

Our 2,200-year geochemical record reveals high-frequency variability in GDGT concentrations, and therefore in the BIT index, superimposed on distinct fluctuations at the multi-decadal to century time scale. Additionally, surface sediments collected in January 2010 show a distinct shift in GDGT composition relative to sediments collected in August 2007. Increased bulk fluxes of settling particles with high Ti/Al ratios during March-April 2008 reflect an event of unusually 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 massive dry-season diatom bloom in July-September 2008 and a concurrent increase in the flux of GDGT-0. Simultaneous near-absence of crenarchaeol indicates that no Thaumarchaeota bloom developed at that time; instead a peak in brGDGT fluxes is observed in December 2008. We suggest that increased nutrient availability, derived from the eroded soil washed into the lake, stimulated productivity of both diatoms and the GDGT-0 producing archaea, which probably grow on decomposing dead diatoms passing through the sub-oxic zone of the water column. This disadvantaged the Thaumarchaeota that in more typical years prosper during the austral summer. Instead, a bloom developed of supposedly heterotrophic brGDGT-producing bacteria.

Decade-scale BIT index fluctuations in Lake Challa sediments exactly match the timing of three known episodes of prolonged regional drought within the past 250 years. Additionally, the principal trends of inferred regional rainfall variability over the past two millennia are consistent with the hydroclimatic history of equatorial East Africa as has been documented thus far from other (but less well dated) lake records. We therefore propose that variation in GDGT production originating from the episodic recurrence of strong soil-erosion events, when integrated over (multi-)decadal and longer time scales, generates a stable positive relationship between the sedimentary BIT index and monsoon rainfall at Lake Challa. However, application of this paleoprecipitation proxy at other sites requires ascertaining the local processes which affect the productivity of crenarchaeol by Thaumarchaeota and brGDGTs.

INTRODUCTION

Geographically widespread isoprenoid and branched glycerol dialkyl glycerol tetraether membrane lipids (iso/brGDGT; see Appendix for their structures) have allowed the development of several new molecular proxies used in palaeoenvironmental reconstruction (e.g. Schouten et al., 2002; Hopmans et al., 2004; Weijers et al., 2007; Schouten et al., 2013). Although isoGDGTs can be found in soil (Leininger et al., 2006) and peat (Weijers et al., 2004; Weijers et al., 2006), they are generally most abundant in marine and freshwater environments (Sinninghe Damsté et al., 2002; Blaga et al., 2009). Mesophilic isoGDGT-producing Crenarchaeota (e.g. Wuchter et al., 2004) now called Thaumarchaeota (Brochier-Armanet et al., 2008; Spang et al., 2010) 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., 2007; 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., 2009b). BIT expresses the abundance of brGDGTs relative to the isoGDGT crenarchaeol (V; see for structures and nomenclature the Appendix), the characteristic membrane lipid of pelagic Thaumarchaeota (Sinninghe Damsté et al., 2002; Pitcher et al., 2011b). 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., 2014), and suggested, but not confirmed, in profundal surface sediments (Buckles et al., 2014).

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 both with known climatic events for the region and with 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., 2014) 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., 2014). 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 2,200-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.

1. MATERIALS AND METHODS

2.1. Study site

Lake Challa is a 4.2 km^2 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^2, which during periods of exceptional precipitation can enlarge to 1.43 km^2 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 (Payne, 1970) and is mostly derived from rainfall on the montane forest zone of Mt. Kilimanjaro (1800 to 2800 m elevation; Hemp, 2006). Passage of the 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). Mean daily air temperatures at the lake are lowest (20-21^{o}C; 24 h average) in July-August (austral winter), and the highest (25-27^{o}C; 24 h average) in January-February (austral summer; data from 2006-2009 provided by A. Hemp, University of Bayreuth; cf. Buckles et al., 2014). The lake surface water is coolest (~23^{o}C) between June and September, promoting seasonal deep mixing that reaches down to 40-60 m depth. During austral summer the surface water can reach 28^{o}C in late afternoon, and experiences shallow daytime 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 ^{o}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 cool and windy winter months of deep seasonal mixing (Barker et al., 2011), alternating with organic matter and calcite laminae deposited during the austral spring and summer to produce alternating dark/light layers.

2.2 Core collection, sampling and age model

The composite sediment sequence studied here mostly consists of a mini-Kullenberg piston core (CH03-2K; 2.6 m) recovered in 2003 from a mid-lake location (Fig. 1), supplemented at the top by a cross-correlated gravity core (CH05-1G) and a short section of a Uwitec hammer-driven piston core (CH05-3P-I) recovered in 2005 (Verschuren et al., 2009; Wolff et al., 2011). Importantly, core CH05-1G was kept upright upon retrieval, and its intact sediment-water interface was drained of superfluous water by perforating the transparent core tube shortly below that level. It was then allowed, for two days, to evaporate part of its upper interstitial water so as to enable transport without disturbing the fine lamination of recently deposited sediments. The detailed age model for this composite core sequence, which covers the period between ca. 2150 cal yr BP (ca. 200 BCE) and the present (2005 AD) is a smoothed spline through 45 INTCAL09-calibrated AMS ^{14}C ages of bulk organic carbon, each corrected for an evolving old-carbon age offset determined by paired AMS ^{14}C dates on charcoal, and supplemented by six sub-recent age markers cross-correlated from the ^{210}Pb-dated gravity core CH99-1G on the basis of shared high-resolution magnetic-susceptibility profiles (Blaauw et al., 2011). This particular core sequence has also been dated through varve counting (Wolff et al., 2011). The latter chronology is fully consistent with the radiometric (^{210}Pb, ^{14}C) chronology, demonstrating that the sediment has been deposited in a continuously anoxic deep-water environment throughout this period. In this study, we examined 208 integrated sediment intervals from 0 to 213 cm depth, each 1 cm (10 mm) thick and sampled contiguously with the exception of five intervals (23-24, 28-29, 99-100, 100-101 and 153-154 cm) where previous analyses had depleted the available material. Each interval of the resulting time series thus represents 10.4 years, on average.

2.3 Organic carbon analysis

Percent organic carbon (%C$_{org}$) data are based on determination of percent organic matter (%OM) at contiguous 1-cm intervals, obtained by the loss-on-ignition (LOI) method (Dean, 1974) and using a linear regression against %C_{org} values obtained on a subset of the same intervals. These %C_{org} values were determined through combustion of acidified sediment samples on a Fisons NA1500 NCS elemental analyser (EA) using the Dumas method (courtesy of Birgit Plessen, GFZ-Potsdam). GDGT concentrations reported in this paper are relative to the sample's C_{org} content, unless otherwise stated.

2.4 Diatom analysis

Diatom productivity was quantified as the flux of diatom frustules settling in a 58 cm^2 sediment trap suspended at 35 m water depth, sampled on a near-monthly basis from 18/11/2006 to 31/08/2010 (i.e., continuously for 45 months in total). For the first 21 months, diatom analysis was performed on 1/8 of the sediment-trap material retained on a GFF filter and preserved frozen until use. This residue was brought back in suspension with distilled water; the filter was rinsed and checked under the microscope for any remaining diatoms. For the remaining 24 months plus one overlapping month (August 2008), diatom analysis was performed on unfiltered but freeze-dried subsamples of the collected sediment-trap material, also brought back in suspension with distilled water. In both cases the suspension containing diatoms was then diluted to a known volume and studied quantitatively at 400x magnification. The 21 samples from December 2006 to August 2008 were pipetted onto a microscope slide and analyzed under an Olympus BX50 microscope with differential-interference contrast. The remaining 24 samples were analyzed under an inverted Olympus CX41 microscope using sedimentation chambers of 10 ml (Uthermöhl, 1931). Total diatom counts were converted to the number of frustules settling per m^2 per day. We note that total diatom abundance (and numerical flux) are not linearly proportional to total diatom biomass (and production) at any one time, because the latter also depends on the average cell volume of the species which dominate the community at that time. However, these two sets of variables are broadly proportional to each other at the order-of-magnitude scale of variability observed between successive months and seasons in Lake Challa.

2.5 GDGT analysis

Freeze-dried sediments (1-2 g) were extracted with a dichloromethane (DCM)/methanol solvent mixture (9:1, v/v) using a Dionex™ accelerated solvent extraction (ASE) instrument at high temperature (100°C) and pressure (1000 psi). Each extract was rotary evaporated to near-dryness and separated by column chromatography using Al_2O_3 stationary phase, with the first (apolar) fraction eluted by hexane: DCM (9:1, v:v) and the second (polar) fraction by DCM: methanol (1:1, v:v). 0.1 µg of C_{46} GDGT standard (cf. Huguet et al., 2006) was added to the polar fraction. The apolar fraction was archived.

Analysis of the sediment-trap material and recently deposited surface sediments has been described elsewhere (Buckles et al., 2014). Here we report additional results for GDGTs I to IV (see Appendix for structures) also present in these samples. Sinking particulate matter was sampled at a central location on a near-monthly basis from 18/11/2007 to 31/08/2010, and surface sediments were sampled at seven mid-lake locations in January 2010 (Fig. 1). These samples were processed for GDGT analysis in a slightly different way than core samples (Buckles et al., 2014). In short, the sediment-trap material and surface sediments were extracted using a modified Bligh-Dyer method, yielding both intact polar lipid (IPL) and core lipid (CL) GDGTs. IPL GDGTs were separated from CL GDGTs using column chromatography with an activated silica gel stationary phase, using hexane: ethyl acetate 1:1 (v/v) and methanol to elute CL and IPL GDGTs, respectively. IPL GDGTs were subsequently subjected to acid hydrolysis to remove the functional head groups and analysed as CL GDGTs.

Each fraction was dissolved in hexane:isopropanol 99:1 (v:v) and passed through PTFE 0.45 μm filters prior to high-performance liquid chromatography/atmospheric pressure chemical ionisation - mass spectrometry (HPLC/APCI-MS). This used an Agilent 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_{46} GDGT was used to check, and to account for, differences in ionisation efficiencies.

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

$$\text{BIT index} = \frac{[\text{VIa}]+[\text{VIIa}]+[\text{VIIIa}]}{[\text{VIa}]+[\text{VIIa}]+[\text{VIIIa}]+[\text{V}]} \quad (1)$$

$$\text{MBT} = \frac{[\text{VIa}]+[\text{VIb}]+[\text{VIc}]}{[\text{VIa}]+[\text{VIb}]+[\text{VIc}]+[\text{VIIa}]+[\text{VIIb}]+[\text{VIIc}]+[\text{VIIIa}]+[\text{VIIIb}]+[\text{VIIIc}]} \quad (2)$$

$$\text{DC} = \frac{[\text{VIb}]+[\text{VIIb}]}{[\text{VIa}]+[\text{VIb}]+[\text{VIIa}]+[\text{VIIb}]} \quad (3)$$

The fractional abundance of each individual GDGT is expressed as:

$$\text{f}[\text{GDGTi}] = \frac{[\text{GDGTi}]}{[\Sigma\text{GDGTs}]} \quad (4)$$

Where roman numerals refer to GDGTs in the Appendix, f[GDGTi] = fractional abundance of an individual GDGT, [GDGTi] = concentration of the individual GDGT, based on surface area relative to the C_{46} standard; [ΣGDGTs] = the summed concentration of all measured GDGTs (I to VIIIc); MBT = the methylation index of branched tetraethers; and DC = the degree of cyclisation.

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

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

Where [IPL] = intact polar lipid concentration and [CL] = core lipid concentration. IPLs represent living, GDGT-producing bacteria/archaea (e.g. Lipp and Hinrichs, 2009; Pitcher et al., 2011a; Schubotz et al., 2009; Lengger et al., 2012).

The measurements of the BIT index were performed in duplicate for all samples; the differences between the two measurements were on average 0.02. The concentrations of crenarchaeol and the summed acylic brGDGTs (i.e. VIa+VIIa+VIIIa) were also determined in duplicate.

2.6 Statistical analysis

Pearson product-moment correlation coefficients were calculated on un-smoothed time series of the geochemical data at 1-cm interval, using a two-tailed test of significance in IBM SPSS Statistics 21, with bootstrapping at the 95% confidence interval and missing values excluded pair-wise. Calculating mean varve thickness at fixed 1-cm intervals of core depth is complicated, because it requires averaging over a variable number of varves (including partial varves at the start and end of each interval). In addition, the exact boundaries of individual varves can only be discerned microscopically in thin-sectioned sediment, which has inevitably sustained some deformation during its embedding in epoxy. We therefore calculated the correlation between BIT index values and a 9-point running average of annual varve thickness, for varve years most closely matching the mid-depth radiometric age of successive 1-cm BIT index intervals. Due to gaps in the varve-thickness record, and widening of those gaps in the 9-point average time series, this correlation is limited to 159 data pairs. Cut-off values for designation of correlation strengths were based on guidelines by Dancey and Reidy (2004), however with slightly lower boundary conditions allowed to take into account confounding factors such as a relatively large number of GDGT measurements with zero or near-zero values; small but potentially significant time offsets between the calendar-dated varve record and the radiometrically-dated geochemical record; the relatively low number of data points in the geochemical time series (208); and ecological factors such as changes in GDGT production (or mean depth of production) and in the influxes of allochthonous materials over time. 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 2,200-year BIT index record

The percent total organic carbon (%C_{org}) in the composite sediment sequence (Table S1) varies from 4.4 to 12.5%, with the lowest values generally grouping between 1200 and 1800 AD (Fig. 3A). The concentration of GDGT-0 (GDGT-I; Appendix) varies widely (97 to 921 µg g^{-1} C_{org} and standard deviation of 150 µg g^{-1} C_{org}; Table S1), and at an average of 273 µg g^{-1} C_{org} it is generally high. A baseline concentration of 200-400 µg g^{-1} C_{org} is interrupted by relatively long-term pulses of >500 µg g^{-1} C_{org} (Fig. 3B), the longest of which stretch from around 100 to 200 AD, 300 to 500 AD and 1200 to 1400 AD.

The crenarchaeol concentration fluctuates by two orders of magnitude between 7 µg g^{-1} C_{org} at ca. 740 AD and 398 µg g^{-1} C_{org} at ca. 1800 AD (standard deviation of 64 µg g^{-1} C_{org}; Table S1). Periods of high crenarchaeol (>150 µg g^{-1} C_{org}) occur from around 600 to 650, 1250 to 1300, 1520 to 1570 and 1750 to 1820 AD (Fig. 3C). The proportion of the crenarchaeol regioisomer with respect to crenarchaeol (GDGT V'/(V+V')) is relatively low and constant at around 2.5 to 3% throughout the analysed core sequence (peaking at 4.0% at ca. 740 AD; Fig. 3D), confirming that the majority of recovered crenarchaeol originates from aquatic, rather than soil, Thaumarchaeota (cf. Sinninghe Damsté et al., 2012a; 2012b).

The summed concentration of all brGDGTs (relative to %C_{org}) varies by one order of magnitude between 95 and 557 µg g^{-1} C_{org} (standard deviation of 68 µg g^{-1} C_{org}; Table S1). On average, the total brGDGT concentration is higher than that of crenarchaeol (197 vs. 113 µg g^{-1} C_{org}) but similarly displays a baseline (here between 200 and 250 µg g^{-1} C_{org}; 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^{-1} 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. 1205 AD) and 0.93 (142-143 cm; ca. 740 AD), with an average of 0.65 ±0.09 (Table S1). Generally higher BIT values are evident from ca. 650 to 950 AD (Fig. 3F), followed first by a period of lower BIT values (ca. 1170 to 1300 AD) and then a period of higher BIT values (ca. 1550 to 1700 AD). Following a 40-yr period of very low BIT values (1780-1820 AD), an overall increase to the present is interrupted by two brief periods of lower BIT values, in the late 19th century and in the 1970s. 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).

3.2 Settling particles

Results for bulk sediment flux, %C_{org}, crenarchaeol and brGDGTs in the monthly sediment-trap time series have been presented elsewhere (Sinninghe Damsté et al., 2009; Buckles et al., 2014). Here they are shown (Figs. 2B, 2D and 2E) 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^{-2} day^{-1}) from the start of its measurement in December 2007 until June 2008 (Fig. 2C, Table S3), but subsequently peak at 7.7 µg m^{-2} day^{-1} in August 2008 during a massive, mixing-season diatom bloom (Fig. 2C). After this maximum, IPL GDGT-0 fluxes vary between 0.0 and 1.7 µg m^{-2} day^{-1}, with an additional peak of 2.8 µg m^{-2} day^{-1} in September 2009. CL GDGT-0 fluxes track those of IPL GDGT-0 but are notably lower, ranging from <0.05 to 2.0 µg m^{-2} day^{-1} (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 (Table S4).

3.3 Surface sediments

Sinninghe Damsté et al. (2009) and Buckles et al. (2014) presented data on %C_{org}, 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 labelled CH07) and January 2010 (seven CH10 gravity core tops, all 0-1 cm depth). Here they are shown again (Figs. 5B, 5C, 6A and 6C) as reference for new data on the 2,200-yr sediment record and on IPL and CL GDGT-0 (Tables 1, S5 and S6). IPL and CL GDGT-0 concentrations in CH10 surface sediments are, on average, 14.5 and 8.7 µg g^{-1} 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.

4. DISCUSSION

4.1 Temporal variability in sedimentary GDGT composition and BIT index

The 2,200-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. To allow greater insight into the factors affecting BIT index variation over time, we here quantify the absolute GDGT concentrations, which had not been examined for the 25,000-year, lower-resolution BIT index record (Verschuren et al., 2009; Sinninghe Damsté et al., 2012a). As our 2,200-year record is generated from the upper portion of the same composite core sequence, it should show BIT values which are comparable, both in absolute values and variability, to those of the 25,000-year record when the measurements are integrated over identical depth intervals. Indeed, averaging the BIT index values of our new decadal-resolution record over four adjacent 1-cm samples is found to closely mimic the BIT index values obtained from contiguous and homogenized 4-cm sampling increments of the low-resolution record (Fig. 7). This exercise demonstrates that the lower-resolution record fails to capture strong variation in sedimentary GDGT concentrations (and therefore in the BIT index) on short timescales, as revealed by the high-resolution analysis (Figs. 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 time series of sediment-trap samples and in our chronosequence of surface sediments.

4.2 GDGT variability in Lake Challa settling particles and surface sediments

As confirmed by ^{210}Pb-dating of core CH99-1G (see Blaauw et al., 2011 for further details), shared tie points in the visible fine lamination and in magnetic-susceptibility profiles of multiple gravity cores collected between 2003 and 2011 show that the very soft (water content >95%) and uncompacted uppermost centimetre of mid-lake profundal surface sediments in Lake Challa represents approximately two years of deposition (Sinninghe Damsté et al., 2009; Blaauw et al., 2011). Consequently our core-top sample CH07 (0-1 cm) can be treated as broadly representing the period from mid-2005 to August 2007, and CH10 (0-1 cm) the period from early 2008 to January 2010. By comparison, one centimetre of compacted sediments in our 2,200-year record represents about a decade of deposition. Note that this is also the case at its very top, because the intact sediment-water interface of core CH05-1G was 'compacted' in the field by draining and evaporation of interstitial water in preparation of transport (cf. section 2.2).

GDGTs in CH10 surface sediments are dominated by GDGT-0 and brGDGTs, with relatively low proportions of crenarchaeol (Figs. 6A). IPL and CL BIT index values are therefore high at ca. 0.90 (Table 1; Fig. 5D). This differs markedly from the CL GDGT composition of CH07 surface sediment. CH07 has a higher proportion of crenarchaeol than either brGDGTs or GDGT-0 (Figs. 6C), and consequently display a lower BIT index value (0.50; Fig. 5D). This shift in fractional abundances is also reflected in the absolute concentrations (Table 1). The BIT index difference between CH10 and CH07 surface sediments is consistent with BIT index trends in settling particles, which are higher, on average, over the period covered by CH10 than over the period covered by CH07 (Fig. 2F). Whereas 45 months of sediment trapping has yielded BIT index values ranging between 0.09 and 1.00, the absolute difference (0.40) between BIT index values of the temporally more integrated surface sediment samples CH07 and CH10 is comparable to the full range of BIT index variation in the 2,200-year sediment record (Fig. 5D). Our monthly collections of settling particles also yield far greater differences in GDGT distribution (Figs. 2C-F) and brGDGT composition (Fig. 4) than any other sample group. This implies that a still higher-resolution geochemical analysis of a long sediment record would yield even greater temporal variation in GDGT distribution than observed in this study, at least in the case of Lake Challa where seasonal variation in the composition of settling materials is preserved intact as finely laminated sediments with annual rhythm (varves).

Since the brGDGTs and crenarchaeol found in Lake Challa sediments are thought to be primarily produced between 20 and 40 m depth (Buckles et al., 2013; 2014), it is tempting to attribute these rapid changes in the GDGT composition of descending particles and surface sediments to shifts in the GDGT-producing community within the water column. In Lake Challa, crenarchaeol is produced by Thaumarchaeota that have bloomed annually during the austral summer (between November and February) in three out of four monitored years (Fig. 2E). Its production in the suboxic zone between 20 and 45 m depth (Buckles et al., 2013) is where the majority of GDGTs found in surface sediments originate (Buckles et al., 2014). Thus, data from settling particles trapped at 35 m depth can be used to assess the amounts and distribution of GDGTs exported to the sediments. Here, we examine fluxes of settling particles (Sinninghe Damsté et al., 2009; Buckles et al., 2014) integrated over the time period from November 2006 to August 2007 and from February 2008 to January 2010 (Table 2). Encompassing the two years prior to collection of our CH10 surface-sediment samples, the latter period is taken to represent the contribution of GDGTs from the water column to CH10 sediments (0-1 cm depth). The former period encompasses just under a year of deposition prior to collection of CH07 surface sediments and thus does not cover the two years of deposition approximately represented by its 0-1 cm interval; however, GDGT compositions of the 0-0.5 cm and 0.5-1.0 cm intervals of CH07 (analysed separately; Table S4) are comparable.

Comparison of these two types of time-integrated samples (Figs. 7A-D) shows, simultaneously, the strong contrast in the distribution of GDGTs exported to Lake Challa sediments during these two time periods (cf. Fig. 6A-D) and the good overall correspondence between GDGT distributions in settling particles and surface sediments that represent the same period (Fig. 6: A-B versus C-D). GDGT-0 is present in higher proportions in CH10 and CH07 surface sediments than in settling particles (Fig. 6: A-C versus B-D), likely indicating additional production within the bottom sediments and/or in the water column below 35 m depth (cf. Buckles et al., 2014). BrGDGTs (GDGTs VI-VIII) appear to have similar proportions in sediments and settling particles (accounting for the difference in GDGT-0). However, MBT indices of the two sample groups are slightly offset (Fig. 4). This is most likely due to a (small) contribution from sedimentary brGDGT production, as identified previously by Buckles et al. (2014). Besides these minor differences, the GDGT distributions in settling particles during both periods largely replicate the contrast in GDGT distribution between CH10 and CH07. As the former are due to changes in the GDGT-producing community within the upper part of the water column, we can use our monthly GDGT-flux time series to determine the cause(s) of short-term shifts in sedimentary GDGT distribution.

CL crenarchaeol fluxes in settling particles reached three clear peaks, indicating Thaumarchaeota blooms, in January 2007 (9 μg m^{-2} day^{-1}, Fig. 2E; Table S3), December 2007 to January 2008 (3 μg m^{-2} day^{-1}) and March to April 2010 (4 μg m^{-2} day^{-1}). IPL crenarchaeol fluxes (where available) were an order of magnitude lower than, but co-varied with CL fluxes. IPL GDGT-1, -2 and -3 fluxes co-varied with crenarchaeol (Table S3), indicating that they are primarily produced by Thaumarchaeota as previously discussed by Buckles et al. (2014). In contrast, CL brGDGT fluxes peaked at 12 μg m^{-2} day^{-1} between mid-November and December 2006 (Fig. 2D) and at 10 μg m^{-2} day^{-1} in December 2008. Concurrent with maxima in IPL brGDGTs, they are likely due to blooms of brGDGT-producing bacteria in the water column (Buckles et al., 2014). Although both maxima occurred near the end of the short rain season and may thus represent a seasonal bloom, they occurred in only two of four such seasons that we monitored. Since the first peak in brGDGT fluxes occurred during deposition of CH07 and the second during deposition of CH10, this may account for their similar brGDGT concentrations (Table 1; Figs. 5C, 6A, and 6C).

GDGT-0 fluxes were high (CL: ca. 1 μg m^{-2} day^{-1}; IPL not measured) between mid-November and December 2006 but declined to near-zero values by March 2007 (Fig. 2C; Table S3). GDGT-0 fluxes peaked again in August 2008 (2 and 8 μg m^{-2} day^{-1}, respectively, for CL and IPL). During these maxima, crenarchaeol and cyclic isoGDGTs did not co-vary with GDGT-0, confirming a separate source for GDGT-0 in the water column as previously suggested (Sinninghe Damsté et al., 2009; 2012a; Buckles et al., 2013).

Notably, Thaumarchaeota did not bloom during the 2008/2009 austral summer, which most likely accounts for the low crenarchaeol abundance in CH10 surface sediments compared to CH07 (Figs. 2E and 5B). Since Thaumarchaeota are nitrifiers (Könneke et al., 2005; Wuchter et al., 2006), they should in principle have prospered on the ammonium released by biomass degradation resulting from the massive diatom bloom recorded in July-September 2008. In fact, North Sea studies have shown that Thaumarchaeota blooms follow phytoplankton blooms in that setting (Wuchter et al., 2006; Pitcher et al., 2011c). However, in Lake Challa during the 2008/2009 austral summer, Thaumarchaeota appear to have been outcompeted first by GDGT-0 producing archaea, resulting in the high proportion of GDGT-0 in CH10 sediments (Fig. 6A), and perhaps subsequently by brGDGT-producing bacteria occupying a similar niche in the suboxic water column (Buckles et al., 2013; Buckles et al., 2014). Although little is known about the ecology or even identity of brGDGT-producing bacteria (Weijers et al., 2009a; Sinninghe Damsté et al., 2011, 2014), the occurrence of a similar brGDGT peak in December 2006 (Sinninghe Damsté et al., 2009; Fig. 2C), i.e. following the austral winter diatom bloom of 2006 (Wolff et al., 2014), suggests that brGDGT-producing bacteria may also thrive on diatom degradation products. Abundances of total brGDGTs and crenarchaeol in monthly fluxes of settling particles do correlate (r = 0.66), indicating that the brGDGT-producing bacteria and Thaumarchaeota within the water column of Lake Challa require similar environmental and/or ecological conditions. This would fit with compound-specific carbon isotopic analyses on soils (Weijers et al., 2010; Oppermann et al., 2011), which suggest that brGDGT producers are heterotrophic bacteria.

Microbiological analysis of suspended particulate matter (SPM) from Lake Challa collected in February 2010 (Buckles et al., 2013) yielded no evidence of methanogens or other anaerobic archaea in the upper 35 m of the water column, in line with the near-zero fluxes of GDGT-0 in settling particles trapped at this time (Fig. 2C). However, in SPM from anoxic waters deeper down, Buckles et al. (2013) found high concentrations of GDGT-0. Based on 16S rRNA sequence data, its source was identified as the uncultured archaeal group 1.2 (also named C3 by DeLong and Pace, 2001) and the 'miscellaneous Crenarchaeota group' (MCG, also referred to as group 1.3; Inagaki et al., 2003). Presence of these archaeal sequences in the permanently stratified lower water column during a single sampling of SPM does not prove the origin of similar isoGDGT distributions in settling particles from the suboxic zone two years previously. Comparison with denaturing gradient gel electrophoresis (DGGE) performed on Lake Challa SPM taken in September 2007 (Sinninghe Damsté et al., 2009) does show that archaea in the anoxic water column also mostly fall in Group 1.2 and the MCG (MBG-C) group of the Crenarchaeota, as well as showing contributions from Halobacteriales of the Euryarchaeota. Nevertheless, given the coincidence of the GDGT-0 maximum with the massive diatom bloom of 2008 and the position of the sediment trap in the suboxic 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 oxycline may have temporarily ascended. This could have resulted in the presence of GDGT-0 producing archaea above the sediment trap, and their signal being 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 detrital mineral sediment components (Weltje and Tjallingii, 2008; Wolff et al., 2014). This Ti/Al peak is unprecedented in the Lake Challa sediment-trap record and coincided with a peak in bulk settling flux (4.0-2.0 g m^{-2} day^{-1}), very low OM content (3% C_{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). Notably, the 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 OM content of the eroded soil (Buckles et al., 2014; Table S3). If this soil erosion event did cause the observed shift in Lake Challa's GDGT-producing community, its effect must have been indirect.

Nutrients triggering the annual diatom bloom in Lake Challa during austral winter are generally sourced from its anoxic, nutrient-rich lower water column by wind-driven seasonal mixing (Wolff et al., 2011, 2014; Barker et al., 2013). In the austral winter of 2008, seasonal mixing began already in June and re-establishment of stratification was slow (Wolff, 2012; Buckles et al., 2014; Wolff et al., 2014). However, the massive diatom bloom of July-September 2008 (far larger than any other in our 4-year time series; Fig. 2C) peaked during the early months of deep seasonal mixing so the extended period of nutrient advection that year is unlikely to have been the main cause of this particularly abundant diatom bloom. We hypothesise that additional, soil-derived (micro-) nutrients delivered during intense rainfall between March and May 2008 may have been the primary driver for the unusually large diatom productivity later that year. Nutrients released by the decomposition of soil organic material in the lake would amplify the (annual) 2008 austral winter diatom bloom (Fig. 2B; Wolff et al., 2011). The delivery of large quantities of organic matter could result in higher levels of ammonium, disturbing the competition of nitrifying archaea and bacteria. As nitrifying bacteria have a competitive advantage over nitrifying archaea at higher ammonium levels (Di et al., 2009) and vice versa (Martens-Habbena et al., 2009), increased ammonium could suppress Thaumarchaeota and its production of crenarchaeol and result in the absence of the quasi-annual thaumarchaeotal bloom in the 2008-2009 austral summer season. Considering that the coincident peak flux of GDGT-0 is especially clear in the IPL lipids, we infer that a certain (eury)archaeal community developed in the oxic/suboxic water layer below the euphotic zone, simultaneously with the diatoms in the epilimnion.

4.4. Connection between the BIT index and precipitation

Before the discovery of substantial in situ brGDGT production in Lake Challa (Buckles et al., 2014a), it was thought that precipitation-triggered soil erosion transported brGDGTs to the lake and settled in the sediments against a background of aquatic crenarchaeol, thus increasing the BIT index (Sinninghe Damsté et al, 2009; Verschuren et al., 2009). Since soil-derived brGDGTs entering Lake Challa during March-May 2008 did not discernibly affect the brGDGT distributions and abundances in particulate matter settling at that time (Buckles et al., 2014), the event is barely registered in the BIT index of those settling particles (Fig. 2F). Then how does this evidence support use of the sedimentary BIT index as hydroclimatic proxy in this system (Verschuren et al., 2009)? We propose that in this permanently stratified and (most often) unproductive tropical lake, episodic injection of extra nutrients derived from eroded soils creates a positive feedback loop leading to the suppression of Thaumarchaeota, via changes in the lake's planktonic and microbial communities. Variation in the relative proportions of crenarchaeol and brGDGTs in the 25,000-year sediment record (Sinninghe Damsté et al., 2012a) had already indicated that variability in crenarchaeol is the main driver of BIT index changes in Lake Challa. Also in the higher-resolution record studied here, the BIT index correlates (negatively) with the concentrations (μg g^{-1} C_{org}) of crenarchaeol (r = - 0.69) and its regioisomer (r = - 0.68) but does not correlate significantly with brGDGT concentrations (Table S2). Thus, the hypothesised suppression of Thaumarchaeota following an event of intense precipitation increases the BIT index via its reduction of crenarchaeol deposition (cf. Fig. 6B). We postulate that the strongly seasonal nature of Thaumarchaeota production in this system and the dependence of Thaumarchaeota on the suboxic niche in the water column (Buckles et al., 2013) leaves it more vulnerable to exceptional events. The BIT index can thus be considered to reflect the frequency of 'extreme' soil-erosion events, which in this semi-arid region have a threshold relationship with rainfall extremes.

Several other hypotheses can be put forward to explain the general match between the BIT index and seismic-reflection evidence in Lake Challa (Verschuren et al., 2009). First, lake-level fluctuation alters the relative volumes of the portions of the water column that are either annually or less frequently mixed, thus changing the availability of niches favourable to either Thaumarchaeota or brGDGT-producing bacteria. For example, an increase in accommodation space for Thaumarchaeota in the suboxic zone when lake level is high (cf. Sinninghe Damsté et al., 2012a) may result in conditions more favourable to lacustrine brGDGT production than to Thaumarchaeota, and vice versa.

More relevant at short time scales is the relationship between annual precipitation and strong or prolonged windiness during the dry austral winter season at Lake Challa (Wolff et al., 2011). Stronger wind and its lake-surface cooling result in deeper mixing, enhancing both the regeneration of nutrients from the lower water column to the photic zone as well as delaying the recovery of water-column stratification. Since stronger austral-winter winds are associated with a weak southeasterly monsoon compromising the main rain season during March-May (Wolff et al., 2011), 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.

4.5. A high-resolution record of monsoon precipitation

Wolff et al. (2011) produced a high-resolution, 3,000-year record of varve-thickness variation in Lake Challa bottom sediments using the same composite sediment sequence that we analysed at 1-cm resolution (Fig. 8B), and showed that the thicknesses of varves deposited over the last 150 years correlate both with indices of the ENSO (Niño3.4 SST and the Southern Oscillation Index: Ropelewski and Jones, 1987; Kaplan et al., 1998) and with sea surface temperature (SST) anomalies averaged over the western Indian Ocean (Rayner et al., 2003). Specifically, thick varves are deposited during prominent La Niña years, during which East Africa tends to experience anomalous drought; and thin varves tend to correspond with El Niño years, which are often characterized by high rainfall. Noting that most of the varve-thickness variation resides in variation of the light laminae, which are mainly composed of diatom frustules, Wolff et al. (2011) proposed that prolonged dry and windy conditions during the austral winter season of La Niña years promotes the deep water-column mixing of Lake Challa required to supply surface water with adequate nutrients for diatom growth. As a consequence, La Niña conditions create more prominent annual diatom blooms and thus result in thicker varves. Lake Challa varve thickness thus appears to be an indicator of the inter-annual component of East African rainfall variability that is under control of its tele-connection with ENSO, with greatest sensitivity for the anomalously dry conditions typical of La Niña events.

Wolff et al. (2011) also noted broad visual agreement between (multi-)decadal variability in the Challa varve-thickness record (represented by its 21-point running average) and the last 3000 years of the low-resolution Challa BIT index record (Verschuren et al., 2009); and similarly between a 7-point running average of the Challa varve-thickness record and a 1100-year moisture-balance reconstruction from Lake Naivasha in central Kenya (Verschuren et al., 2000), 400 km northwest of Lake Challa. Broad correspondence between the hydroclimatic histories of lakes Challa and Naivasha is not unexpected, since both sites are located within the broader 'Horn of Africa' region of coastal East Africa where (multi-)decadal variation in monsoon rainfall is strongly tied to changes in SST of the

Indian Ocean (Tierney et al., 2013). More importantly, this correspondence seems to imply that a substantial part of the (multi-)decadal variation in this region's monsoon rainfall can be attributed to the compound effect of alternating increases and decreases in the frequency of La Niña events, possibly mediated by changes in the regional geometry of atmospheric convergence (i.e., ITCZ migration; Wolff et al., 2011) and/or Indian Ocean SST patterns. However, the mechanisms of external climate forcing known to influence ENSO dynamics at these longer time scales (solar irradiance variation, temporal clustering of volcanic activity; Mann et al., 2005) may also, and simultaneously, exert a direct influence on Lake Challa diatom productivity, for example through temperature effects on the seasonal cycle of water-column mixing and stratification. Importantly, this direct influence is not necessarily synchronized with or even of the same sign as the relationship between varve thickness and rainfall at the inter-annual time scale. This complexity of proxy-signal attribution warrants caution in the extraction of multi-decadal and century-scale rainfall trends from the Challa varve-thickness record, and leaves room for other sediment-derived proxies with appropriate sensitivity and range of variation to more reliably capture these longer-term trends in the region's hydroclimate.

Focusing on such (multi-)decadal hydroclimate variability within the last two centuries, both our decadal-resolution BIT index record and the Challa varve-thickness record are highly congruent with independent historical data and previously available paleoclimate records from equatorial East Africa. The most prominent negative excursion in the BIT index time series within this period, here dated to between 1779±14 and 1816±11 AD and consisting of four consecutive data points with BIT index values of 0.48-0.52 (Fig. 8A), matches the episode of extreme aridity that ended the region's generally moist Little Ice Age climate regime (Verschuren and Charman, 2008). In most paleoclimate records employing an age model based on a combination of ^{14}C and ^{210}Pb dating (Verschuren et al., 2000; Stager et al., 2005; Bessems et al., 2008; Kiage and Liu, 2009; De Cort et al., 2013) this dry episode is situated sometime during the late 1700s to early 1800s. High-resolution ^{210}Pb-dating on the sediment record from Lake Sonachi near Naivasha had earlier constrained the end of this drought to 1815±8 AD (Verschuren, 1999a), i.e. indistinguishable from our age estimate for the end of the prominent BIT anomaly at Lake Challa. Within analytical and age-modelling error, both of these radiometric ages are also indistinguishable from the date of 1822-1826 AD on a prominent Ba/Ca peak in a coral from Kenya's Indian Ocean coast (Fleitmann et al., 2007), which is inferred to reflect increased soil runoff from the Sabaki River catchment caused by drought-breaking flood events.

A second prominent BIT index minimum at Lake Challa, consisting of two data points with values of 0.54-0.55 dated to between 1873±7 and 1893±6 AD (Fig. 8A), matches diverse historical and proxy evidence for a prolonged late 19th century episode of anomalous drought throughout East Africa (Nicholson et al., 2012). In lake records from Kenya's rift-valley region, this drought is dated to between the 1870s and early 1890s (Verschuren, 1999; Verschuren et al., 1999, 2000; De Cort et al., 2013), and it is also recorded in the sediments of Lake Abiyata in the rift-valley region of Ethiopia (Legesse et al., 2002). There, major late-19th century drought is at least partly responsible for the "Great Ethiopian Famine" of 1888-1892 AD, which is said to have cost the lives of one third of Ethiopia's population (Pankhurst, 1966). In agreement with the Challa BIT index time series, historical and proxy evidence from throughout the region indicate that this drought ended in the late 1880s (clearly not yet in Ethiopia, cf. above) or early 1890s, with generally much wetter conditions prevailing at the end of the 19th century and the first decades of the 20th century (e.g., Verschuren et al., 1999; Nicholson & Yin, 2001; Verschuren, 2004; Nicholson et al., 2012).

Unresolved data-quality issues concerning the few historical and/or active rain-gauge stations in the wider Challa region preclude a detailed comparison of either the Challa BIT index or varve-thickness records with the instrumental record of annual-mean rainfall at this time, and are beyond the scope of this study. Here we only highlight the exact match between a third BIT index minimum, here dated to between 1963±2 and 1974±2 AD (Fig. 8A), and the cluster of seven thick varves (each of which exceeds 1.4 mm in thickness; Fig. 8B) deposited during a period of near-continuous strong La Niña conditions between 1968 and 1974 (Niño3.4 SST; Kaplan et al., 1998).

Strong visual agreement between the Challa BIT index and varve-thickness records during these three sub-recent drought periods is confirmed by the significant inverse linear correlation between BIT index values and a 9-point running mean of varve-thickness values over the period 1800-2000 AD ($r = -0.55$; $n = 18$). However, this general agreement between the two hydroclimate proxies is not sustained through the earlier part of the record, so that we find no correlation between them for the entire 2,200-year period analyzed in this study ($r = -0.09$; $n = 159$). One obvious difference between the two proxy records is their degree of variance over the entire record compared to that during the 'historical' part of the record (i.e., the period 1780-2005 AD). For the varve-thickness record, these variances are respectively 0.046 and 0.061 (a ratio of 1.33), whereas for the BIT index record these variances are respectively 0.0053 and 0.0088 (a ratio of 1.68). This is so because the varve-thickness time series, with the exception of a cluster of thin (< 0.6 mm) varves deposited during the early 18th century, mostly displays a single trend of gradually increasing thickness throughout the 2,200-year record, with low-frequency variability not much greater (or more extreme) than that realized during the last two centuries. The high-resolution BIT index time series, in contrast, displays several pronounced fluctuations at the (multi-)decadal and century time scale, with minima and maxima inferring the occurrence of past hydroclimatic conditions during the past 2,200 years that were both substantially drier and wetter than the historical extremes. Specifically, the Challa BIT record is consistent with the general temporal pattern of East Africa's climate history during the last millennium, which features a medieval period of prolonged aridity (here, the driest episode is dated to 1170-1300 AD) followed by generally wetter conditions during the East African equivalent of the Little Ice Age (Verschuren, 2004; Verschuren and Charman, 2008).

According to our Challa BIT index record, easternmost equatorial Africa enjoyed its wettest period of the last 2,200 years between ca. 600 and 1000 AD (Fig. 8A). Although quite variable in its expression among the set of presently available records, a distinct period of inferred higher rainfall occurring towards the end of the first millennium AD has also been reported from several other lakes across East Africa: Lake Naivasha in central Kenya reached peak lake level (and minimum salinity) around 900 AD (Verschuren et al., 2000; Verschuren, 2001); low %Mg values in sedimentary carbonates from Lake Edward in western Uganda infer a positive moisture balance between AD 900 and 1000 (Russell and Johnson, 2007); and sedimentary carbonates from Lake Hayq in northern Ethiopia reach minimum $\delta^{18}O$ values around AD 700 (Lamb et al., 2007). Given large uncertainty on the timing of this episode in most East African lake records (at least compared to Lake Challa), the reported proxy signatures may well represent the same, and region-wide, event of elevated rainfall. The first half of the first millennium AD appears to have been rather dry by comparison (mean BIT index value 0.63±0.06 SD (standard deviation), $n = 45$; Fig. 8A), following generally wet conditions during the second half of the first millennium BCE (mean BIT index value 0.71±0.04 SD, $n = 17$; Fig. 8A and Verschuren et al., 2009). The timing of the abrupt drying trend which forms the transition between these two contrasting climate states, here dated to between 45 BCE and 57 AD (7±50 AD; Fig. 8A), matches that of a century-scale episode of pronounced aridity near the start of the Common Era that impacted several other East African lakes whose hydroclimatic history has appropriate late-Holocene age control: Naivasha (shortly before the 2nd century AD; Verschuren, 2001), Edward (1st century AD; Russell and Johnson, 2005) and two crater lakes in western Uganda (early 1st century AD; Russell et al., 2007).

The combined evidence on East Africa's hydroclimate variability during the last two millennia, as well as excellent agreement between BIT index minima and prominent episodes of regional drought within the last 250 years, suggests that our high-resolution, and well-dated, BIT index time series from Lake Challa represents a trustworthy reconstruction of multi-decadal and century-scale trends in the hydroclimatic history of easternmost East Africa. This conclusion, together with the contrasting character of long-term variability displayed by the BIT index and varve-thickness records, supports our proposition that the Challa BIT index is principally a precipitation proxy. However, this is so only on time scales long enough to average out the occurrence of relatively infrequent, rainfall-driven soil-influx events that are sufficiently massive to affect the community structure of aquatic microbiota, and hence the balance of GDGTs deposited in finely-laminated profundal sediments. As is often the case, the mechanism by which the climate parameter of interest in translated into variability of a climate-sensitive sedimentary proxy in Lake Challa is contingent upon site-specific conditions: permanent stratification of the lake's lower water column (creating a permanent but shifting oxycline), dominance of *in-situ* produced brGDGTs, strongly seasonal rainfall of high intensity, and intermittent mobilisation of soil from a semi-arid tropical landscape. While these conditions appear to make the BIT index an effective precipitation proxy at Lake Challa, we recommend its application to other lakes only when factors controlling the crenarchaeol production by Thaumarchaeota as well as brGDGT production are well understood.

5. CONCLUSIONS

Loose soil material transported to Lake Challa by intense precipitation between March and May 2008 stimulated diatom productivity during the subsequent dry season of July-September 2008 and set in motion a sequence of events that shifted the composition of GDGTs exported to profundal bottom sediments. It included a suppression of the seasonal Thaumarchaeota bloom 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 2,200 years results from fluctuations in crenarchaeol production against a background of high *in-situ* brGDGT production. Integrated over approximately 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. Decadal-scale trends in our high-resolution BIT index time series show no significant correlation with those in the annually-resolved rainfall reconstruction for the Lake Challa region based on varve thickness, but capture the three most prominent known episodes of prolonged regional drought during the past 250 years, and are broadly consistent with the hydroclimatic history of East Africa of the last two millenia as presently known.

We propose that the BIT index value of Lake Challa sediments is primarily controlled by variation in the annual Thaumarchaeota bloom during the austral summer, which is suppressed when excess nutrient input associated with occasional rainfall-driven soil erosion events result in these Thaumarchaeota being outcompeted by nitrifying bacteria. Whereas rainfall-triggered events of Thaumarchaeota suppression may occur rather infrequently at inter-annual timescales, 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 local rainfall. Marked decade-scale maxima and minima in the sedimentary BIT index are smoothed further by integration over longer intervals, such as the approximately 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 of Lake Challa sediments reflects the amount of monsoon precipitation indirectly, as is also the case with varve thickness (Wolff et al. 2011) and many other climate proxies extracted from lake sediments. Prior to application elsewhere, we strongly recommend ascertaining the local situation of lacustrine brGDGT production and of variables affecting the productivity of Thaumarchaeota.

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Figure legends

Figure 1: Map of Lake Challa and its volcanic crater catchment with bathymetry (Moernaut et al. 2010) and sampling sites relevant to this study: the sediment trap suspended at 35 m water depth; the composite sediment sediment sequence covering the last 2,200 years; and intact profundal surface sediments collected in August 2007 (CH07) and January 2010 (CH10). The outermost bold black line denotes the catchment area boundary, which coincides with the crest of the crater rim except in the north-western corner where it is breached by a 200-meter ravine (see text).

Figure 2: Fluxes and GDGT parameters of approximately monthly sediment-trap samples of settling particles, from 18/11/2006 to 31/08/2010. (A) monthly precipitation over the 0.5° × 0.5° grid which includes Lake Challa, from the Global Precipitation Climatology Centre data set, version 6 (GPCC-v6; Schneider et al., 2014), and the Ti/Al ratios of settling mineral particles (Wolff et al., 2014); also indicated are the episode of heavy rainfall in March-April 2008, and the estimated period covered by the 0-1 cm interval of surface-sediment samples CH07 and CH10; (B) Fluxes of bulk sedimenting particles and bulk percent organic carbon (%C_{org}) content; (C) Settling fluxes of diatoms, and of the IPL and CL fractions of 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 average CL BIT indices of surface sediment deposited over the time periods corresponding with CH07 and CH10. In panels C-F, geochemical data from 18/11/2006 to 01/12/2007 are by Sinninghe Damsté et al. (2009) and data from the following months until 31/08/2010 are by Buckles et al. (2014).

Figure 3: Bulk and GDGT parameters in the 213-cm long composite sediment sequence from Lake Challa, against sediment age in years AD. (A) C_{org}, (B) GDGT-0 concentration, (C) crenarchaeol (GDGT-V) concentration, (D) the percentage of crenarchaeol regioisomer concentration relative to crenarchaeol, (E) summed brGDGT concentration, and (F) BIT index. Points connected by a thin line represent raw data and the thicker black lines denote 5-point running averages. A few data points are missing for GDGT concentrations because these were not quantitatively measured.

Figure 4: MBT vs. DC plot for the 0-213 cm sediment record (circles), for CH10 surface sediments (squares) from Buckles et al. (2014), and settling particles (triangles; data from 18/11/2006 to 01/12/2007 are by Sinninghe Damsté et al. (2009) and data from the following months until 31/08/2010 are by Buckles et al. (2014)). Black triangles represent settling particles from March-April 2008, during the episode of intense rainfall when the lake was reported to be turning brown.

Figure 5: Boxplot of fractional abundances of (A) CL GDGT-0 (I), (B) CL crenarchaeol V, (C) CL summed brGDGTs and (D) of the BIT index, for respectively surface sediments CH10 (n=7, 0-1cm depth) collated from Buckles et al. (2014), CH07 collated from Sinninghe Damsté et al. (2009) and the 2,200 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.

Figure 6: IPL and CL GDGT distributions from surface sediments (A) CH10 from Buckles et al. (2014) with (B) corresponding weighted average IPL and CL GDGT distributions from summed fluxes of settling particles between 30/01/2008 and 30/01/2010, also from Buckles et al. (2014). (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 18/11/2006 and 24/08/2007.

Figure 7: Comparison of BIT index values from our decadal-resolution time series, averaged over four adjacent 1-cm sections, against the BIT index measured on integrated 4-cm sections of the same sediment core analysed earlier by Verschuren et al. (2009).

Figure 8: (A) Decadal-resolution time series of BIT index variability in the 2,200-year sediment record from Lake Challa, with black symbols and lines representing the raw data and the thick grey line a 5-point running average. (B) Time series of varve-thickness in the same record (Wolff et al., 2011), with purple symbols and lines representing the raw data and the thick black line a 9-point running average. Orange-shaded bars highlight the approximate duration of documented periods of drought in East Africa (see text): 1) 1780-1820 AD; 2) 1870-1895 AD; and 3) 1968-1974 AD.

Appendix: Key to GDGT structures. The number of cyclopentane moieties in the isoprenoid GDGTs is indicated by the nuber following GDGT as indicated. This is not used for crenarchaeol (V) and its regioisomer (V') since these GDGTs contain a cyclohexane ring. BrGDGTs are subdivided by their principal number of methyl substituents: four (VI), five (VII), or six (VIII). Each group consists of the parent brGDGT (a) and brGDGTs with one (b) or two (c) cyclopentane moieties formed by internal cyclization.

Table 1: Mean (±σ) GDGT concentrations, indices, and distributions 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. (2014).

	Depth interval (cm)	**No. of cores**	**Water depth (m)**		**GDGT-0 (μg g^{-1} dry wt.)**	**Crenarchaeol (μg g^{-1} dry wt.)**	**Σ[brGDGTs] [a] (μg g^{-1} dry wt.)**	**BIT**	**MBT**	**DC**
CH10	0-1	7	68-92	**IPL**	14.5 (± 5.3)	0.3 (± 0.5)	2.2 (± 1.1)	0.90 (± 0.10)	0.34 (± 0.06)	0.15 (± 0.01)
				CL	8.7 (± 3.2)	1.2 (± 1.5)	8.5 (± 2.0)	0.87 (± 0.13)	0.36 (± 0.06)	0.17 (± 0.02)
CH07	0-1	1	94	**CL**	5.1	7.4	6.5	0.50	n.m.[b]	n.m.

					Fractional abundance[c]						
	Depth interval (cm)	**No. of cores**	**Water depth (m)**		**GDGT-0 (I)**	**GDGT-1 (II)**	**GDGT-2 (III)**	**GDGT-3 (IV)**	**Crenarchaeol (V)**	**Cren isomer (V')**	**ΣbrGDGTs[a]**
CH10	0-1	7	68-92	**IPL**	0.85 (± 0.10)	0.00 (± 0.00)	0.00 (± 0.00)	0.00 (± 0.00)	0.02 (± 0.02)	0.00 (± 0.00)	0.12 (± 0.07)
				CL	0.49 (± 0.13)	0.01 (± 0.01)	0.01 (± 0.01)	0.01 (± 0.01)	0.07 (± 0.08)	0.00 (± 0.00)	0.42 (± 0.04)
CH07	0-1	1	94	**CL**	0.25	0.02	0.06	0.02	0.32	0.01	0.32

[a] sum of brGDGTs VIa, VIIa, and VIIIa; [b] n.m. = not measured; [c] fractional abundances of the GDGT indicated

Table 2: Fluxes and indices of GDGTs in settling particles throughout the estimated deposition periods of CH10 and CH07 surface sediments. GDGT data from 18/11/2006-01/12/2007 is collated from Sinninghe Damsté et al. (2009) and from 31/12/2007-31/08/2010 is collated from Buckles et al. (2014).

Settling particles	**Deployment date**	**Collection date**	**C$_{org}$ (%)**	**Bulk flux (g m^{-2} day^{-1})**	**GDGT-0 (μg m^{-2} day^{-1})**		**Crenarchaeol (μg m^{-2} day^{-1})**		**Σ[brGDGTs] [a] (μg m^{-2} day^{-1})**		**BIT**		**MBT**		**DC**	
					IPL	**CL**	**IPL**	**CL**	**IPL**	**CL**	**IPL**	**CL**	**IPL**	**CL**	**IPL**	**CL**
2008-2010 (CH10)	30/01/2008	30/01/2010	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)	18/11/2006	24/08/2007	18.1	1.5	n.m.[b]	0.4	n.m.	2.3	n.m.	2.3	n.m.	0.50	n.m.	0.25	n.m.	0.18

[a] sum of brGDGTs VIa, VIIa. and VIIIa; [b] n.m. = not measured