

Utrecht, 4 March 2024

Dear Prof. De Jonge,

We thank you for reviewing our proposed changes and for the invitation to resubmit the manuscript titled, “Disentangling influences of climate variability and lake-system evolution on climate proxies derived from isoprenoid and branched GDGTs: the 250-kyr Lake Chala record” to Biogeosciences.

Both reviewers praise the extensiveness of our dataset, consisting of both branched and isoprenoid GDGT distributions for ~950 sediment horizons, and detailed interpretation of the time series of individual GDGTs and derived proxies. Reviewer #1 provides a few suggestions for further clarification of our text, whereas the main concern of reviewer #2 is the length of our manuscript, and suggests moving (parts of) our section summarizing previous work on Lake Chala (i.e., section 2) to supplementary files.

Besides incorporating the textual suggestions made by the reviewers, we have made the following changes to the new version of our manuscript. All our changes are marked in a “track changes” version of the complete manuscript file, with blue underlined text indicating new material and red strikethrough text indicating deleted material. Note that relocated text is present twice: in red (initial location) and blue (revised location).

- We have improved section 5.1 which describes the development of the chemical gradient in the lake over time by adding more explicit description of i) the evolution in lake hydrology over time due to gradual infilling of the crater basin with sediments and ii) the expected effects of major lake-level changes in the water column’s dissolved ion content.
- We have made a clear effort to shorten (parts of) the manuscript, as requested by reviewer #2 and also suggested by the editor. Specifically, we have removed or condensed several parts of sections 1 (**Introduction**) and 2 (which now has the better fitting title ‘**The modern system and history of Lake Chala**’). This has led to a total reduction of ~120 lines from the main text (~1700 words; ~11% reduction). This condensing effort includes the section summarizing previous work (now an undivided subsection 2.5 with new title ‘**GDGT biochemistry**’). Therefore, we opted not to move that section to supplementary material for the following reasons: i) We feel that a (shortened!) overview of previous research is essential to understanding our interpretation of the data. Moving this part to the supplements would add references to this material in the discussion, which would alter the current flow of the text and likely lengthen the discussion. Removing this part would in turn require the reader of our work to first read a set of previous publications from different disciplines in order to be able to follow our interpretation of the GDGT data presented here. Hence, keeping the overview of previous work as part of our manuscript ensures that it can be read as a stand-alone publication by all disciplines.
ii) This paper is the first publication on the full ICDP-DeepCHALLA core. Hence, we feel that this is the appropriate and necessary moment to summarize the relevant research from this lake, providing the evolutionary and environmental context for all

forthcoming publications on this core. In addition, the fact that our system-level study deals with both geological and biological processes affecting organic geochemical climate proxies calls for an interdisciplinary journal with a wide audience, like Biogeosciences.

iii) Finally, we deliberately targeted Biogeosciences for publication of our work because it is open access, but also an electronic journal where publications only appear online. As such, publication should not be limited by the length of a manuscript as is the case in paper issues. In fact, moving sections from the main text to a supplement does not result in a net text reduction.

We hope that you find our revised version suitable for publication in Biogeosciences.

Sincerely,

Dr. Allix Baxter,

On behalf of all coauthors

Detailed list of changes made in response to the reviewers:

Anonymous Referee #1:

I find a few places in the discussion where it seems that the authors are speculating. For example, is there other evidence for the development of the strong chemical gradient over time? This is certainly possible, but I didn't see any solid evidence for that. I don't think it is a problem to speculate in this type of paper, but they should be clear when they are doing so.

We have clarified the explanation on the development of the chemical gradient in section 5.1.

The authors used the Pearson 2011 calibration to estimate temperature from brGDGTs, ultimately concluding that this "MST" reconstruction that combines the 5 and 6 Me GDGTs appears to be more appropriate to use in Lake Chala than MBT'_{5ME}. This is somewhat surprising as there is substantial evidence that separating the two groups improves temperature reconstruction, though the logic they use is clearly laid out. I am very curious to see if either the Zhao et al 2023 (QSR), O'Beirne et al 2023 (GCA), or Wang et al 2024 (EPSL) calibrations provide more reasonable results. I fully understand that these calibrations were not yet published when this work was being performed, but now that they are out there I wonder if they do a better job than the Pearson calibration, which has issues of its own that compromise its application.

As motivated in our earlier replies to the reviewer we have chosen to not incorporate and discuss temperature reconstructions based on alternative calibrations into the present manuscript. In Baxter et al. (2023, Nature), we have already shown that any calibration based on the 'classically' defined MBT'_{5ME} will result in reconstructed temperature records that are likely incorrect, and demonstrated that 6-me brGDGTs are required to better capture past temperature change in the Lake Chala sediment sequence. However, we do plan to discuss the potential of the new calibrations mentioned by the reviewer in a forthcoming publication in preparation which focusses primarily on the Lake Chala temperature record and its climatic implications.

A few minor typos:

Line 164 needs closing parenthesis after Johnson et al., 2016

Line 351: change to "...Similar to brGDGTs, isoGDGT-0 is also produced"

Lines 361 and 372: change to "Table 1"

Line 461: Change to "...The degree of cyclisation (DC) of brGDGT was also calculated..."

Line 497: change to "...within..."

Lines 652-653: sentence is unclear, use of "respectively" twice in once sentence

Line 676: change to "confirms"

Line 722: change to "Stage"

Line 796: change to "from"

Line 886-887: used "inferred" twice

Line 923: change to "insolation"

Line 924: change to "from around"

We have made these changes.

You use both "GDGT" and "GDGTs" when referring to them as plural. Choose one and use it consistently

We have now consistently used "GDGTs" for the plural form in the revised manuscript.

Anonymous Referee #2

This paper incorporates a wealth of data and very detailed data interpretation. Finishing such a paper would be a tough task. The paper is generally well-written. However, reading a paper like this would be a challenge to a reader because it is too long and there is too much information that needs to be considered. I do not have comments on the scientific issues. Instead, I strongly suggest the authors remove some unnecessary parts of the paper (moved to the supplementary material) and make the paper succinct and easy to read. That would attract a much wider interest to read through the paper.

We have made a considerable effort to shorten the manuscript. Specifically, we have reduced the introduction and the section on the study site and previous work (section 2). As a result of this, the latter section is now entitled 'The modern system and history of Lake Chala', and summarized all previous biomarker work in one subsection rather than 3. In addition, the materials & methods section is condensed from 5 to 3 subsections. Overall, our efforts have led to a shortening of the text by ~120 lines and should have contributed to the overall readability of our manuscript.

Table 1 '%GDGT-2' A bracket is missing. Two 'iso-GDGT-2' were in the denominator.

We have corrected this.

L110 'only few' is not accurate. There are many downcore applications of brGDGTs in lakes. We have changed this sentence as follows: "However, despite strong correlation between MBT'5Me in lacustrine surface sediments and temperature (Russell et al., 2018; Martínez-Sosa et al., 2021; Raberg et al., 2021), only few down-core applications of lake-based temperature calibrations have proved successful (Feakins et al., 2019; Stockhecke et al., 2021; Zhao et al., 2021; Zhang et al., 2021; Garelick et al., 2021; Ramos-Roman et al., 2022; Parish et al., 2023), partly due to continued uncertainty about the exact source(s) of brGDGTs in lakes."

L175 I suggest that you delete the detailed descriptions of all previous results or make a summary of them. Such a detailed description of previous GDGT work in Lake Chala makes the part look like a review. The paper is too long, which eliminates the interest of careful reading. This detailed description can be moved to the supplementary files

As motivated in our general response to the editor, we have opted not to move any parts of our manuscript to the supplements. We did, however, made a considerable effort to shorten the text, as specified in our reply to the first comment.

L260 delete 'then'

L439 following the method of Hopmans et al.(2016)

L459 calculated according to De Jonge et al. (2015).

Figure 10 in e) Stage VI should be stage IV

L796 from

L924 from around

L932 where these lipids

L951 Data on this chapter? Which chapter?

L955 'review and edited the?'

L1033 Chen et al.... This is a preprint. The paper has been published in GCA.

We have corrected all these minor changes.