Biogeosciences Discussions

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Reply on RC1

Jonathan H. Raberg et al.

Author comment on "Revised fractional abundances and warm-season temperatures substantially improve brGDGT calibrations in lake sediments" by Jonathan H. Raberg et al., Biogeosciences Discuss., https://doi.org/10.5194/bg-2021-16-AC1, 2021

Response to Reviewer 1 Comments (RC1):

We thank Reviewer 1 for their insightful comments and suggestions. A detailed response is provided below:

Review of Raberg et al.,

General comments

The manuscript of Raberg and colleagues reports the investigation of the impact of different environmental parameters on the distribution of branched GDGTs (brGDGTs) in a globally distributed set of lake sediment samples. Following this analysis, the authors propose new calibrations for the use of brGDGTs as paleo-proxies. Branched GDGTs are lipid biomarkers that are ubiquitous in continental settings and are increasingly used in paleo-studies to reconstruct past air temperatures (and sometimes pH) from lacustrine archives. However, the organisms producing brGDGTs are still unknown so the relationships between their distribution in a sample and environmental parameters (temperature, pH ...) remain empirical. In this context, Raberg and colleagues provides a very comprehensive analysis of the relationships between the lipid distribution and a wide range of environmental parameters. Furthermore, they extend the latitudinal coverage of the worldwide sample set typically used to establish brGDGT calibrations to high latitude lakes. This study is thus of great interest for the community. The manuscript is clear and well written although it would benefit from reducing some parts (see below). I thus consider the manuscript to be suited for publication in Biogeosciences after minor revisions. The authors will find below a list of specific scientific comments and a list of technical ones.

Specific comments

-The approach of dividing the compounds into subsets to isolate each structural variation is interesting and valuable as it enabled the authors to reveal some physiological links between the lipid structures and the environmental parameters. I would suggest the authors to more specifically explain the rationale for their groupings in the introduction (l. 111-112). It would also be interesting to further discuss the relationships revealed by their approach in light with the literature on other biomarkers, such as isoprenoid GDGTs or alkenones. Are the observed lipid structure adaptations coherent with the homeoviscous membrane adaptation theory?

We thank the reviewer for these suggestions. We will briefly outline our rationale behind the brGDGT groupings in the introduction, leaving the main discussion for Section 3. We will discuss connections to homeoviscous membrane adaptation theory for isoGDGTs and alkenones as a potential explanation for brGDGT structural distributions. A more involved discussion is beyond the scope of this work, but readers are directed to the Weijers et al. (2007) reference for a more detailed treatment of this subject.

-Moreover, I strongly recommend the authors to better emphasize why the calibrations they set up are better than the previously established ones, especially the one, still under review but available as preprint, proposed by Martinez-Sosa and colleagues. I am, up to now, not convinced that the authors temperature calibration would perform better than others. Eventually, the paleo-community needs to know which calibration is the best suited for their archive(s). The authors should thus clearly and specifically state in their introduction and in the discussion part, the benefits for the paleo-community of using the calibrations they defined in the present manuscript over the others previously published calibrations.

We thank the reviewer for this concern. We agree that "better" and "best" are ambiguous terms for evaluating our results that should be avoided. It is more accurate to say that our calibrations have some advantages in comparison to previous ones. In terms of R^2, the structural set calibrations perform comparably well to those derived using the standard approach (slightly worse for temperature and pH, slightly better for conductivity). The main advantage of using the structural set calibrations is that they can perform this well using a smaller number of compounds chosen to isolate variations in a single structural variable. In theory, this selective approach could provide some protection against the unwanted influences of other environmental parameters on different compounds (e.g. L618-621). Furthermore, subset-specific calibrations using only the most abundant compounds make it easier to apply brGDGT proxies to organic-lean samples, where only some (major) compounds are typically present above detection limit. However, we fully recognize the need to test these calibrations in paleoclimate archives, ideally with independent proxies for comparison. Such tests will be the subject of future work from our group, and we hope from the broader paleoclimate community as well. We have included the Full set temperature calibration (Eqn. 11) for such comparisons, but will emphasize this point in the text.

-The proposed calibration with the conductivity of the lake water column is novel and of

high potential for paleo-studies. The authors note that, in their dataset, pH and conductivity covary but they never suggest an explanation for this covariation and treat them separately all along the manuscript. In fact, it is not surprising to observe links between pH and conductivity in a lake water column. This aspect should be further discussed in the manuscript.

We thank the reviewer for this suggestion. Indeed, it is not surprising to find that conductivity and pH covary in our dataset. Variations in the concentration of H_3O+ (or OH-) will affect conductivity both directly and indirectly through altering the solubility of various ions. Furthermore, both pH and conductivity can be controlled by shared factors such as rock dominance and precipitation chemistry (Wetzel, 2001). While it is therefore not surprising that these variables covary in our dataset, the exact nature of their connection is complex and varies from one site to the next. An in-depth analysis of these connections is beyond the scope of this study; however, an understanding of modern pH and conductivity controls on a given site would be crucial for interpreting a downcore reconstruction of these variables using our proposed calibrations. We will therefore discuss the connection between pH and conductivity in the revised manuscript, both in the context of our modern dataset and for consideration during future downcore applications.

Wetzel, R.G., 2001. Salinity of Inland Waters, in: Wetzel, R.G. (Ed.), Limnology. Academic Press, pp. 169–186.

-In line with the previous comments, parts three and four seemed often redundant and sometimes too descriptive. The manuscript will benefit from a reorganization/condensation of these two parts. This reorganization should put forward comparisons of the study results with previous literature.

We thank the reviewer for this feedback. We recognize that the separation of Section 3 ("Partitioning brGDGTs into structural sets for FA calculations") from Section 4 ("Results and Discussion") results in some redundancies; however, we choose not to merge/reorganize these sections because they serve distinct purposes. Section 3 aims to outline the new structural set methodology, while Section 4 applies the methodology to the lake sediment dataset. We will, however, work to condense and/or eliminate any redundancies in these sections where we are able.

Comparisons of our results with previous literature are generally deferred to Section 4.4.4 ("Recommended Calibrations"), which aims to gather all applicable calibration results for clarity and convenience.

-In the introduction, the authors should briefly describe the four temperature indices they used and the differences between them (l. 114).

We thank the reviewer for this suggestion. We will add a brief description of the four temperature indices and their differences in the introduction, leaving a more detailed discussion for Section 2.4.

-Also, the authors should mention in the introduction the previous studies that evidenced the multiple sources of brGDGTs in lake sediments: from the lake catchment but also from in situ production in the water column or in the sediment. This aspect will have implications to define the environmental parameters the producers are effectively experiencing and could warrant further discussion in their discussion part notably in l. 592 (can the depth habitat of the producer have a role?) and in l. 601-614 (could the export mechanism also play a role here?).

We thank the reviewer for this suggestion and agree that a discussion of the various sources of brGDGTs to lake sediments would be valuable to include. We will do so in the revised manuscript.

-In the material and methods part, I wonder if the paragraph 2.3 really belongs there. Maybe the authors could put it in the supplementary material instead. Also, some details on their statistical procedure are missing. It would be important to know if their variables were all normally distributed or if the authors transformed and centered them before defining the linear models, for example.

Paragraph 2.3 (Comparison of ASE and BD extraction methods) outlines a methodological result that is auxiliary to the main points of the paper, but we believe will be of interest to the brGDGT community. We opted to put it in the main text for this reason (and because it is within the space constraints of the manuscript), but have no qualms moving it to the Supplement and will leave the final decision to the discretion of the editor.

The dataset was constructed from our own data and the published studies available to us and were therefore not normally distributed. Logarithmic transformations for some variables (e.g. conductivity) resulted in a much more even spread of the data. BrGDGT data were normalized through fractional abundance calculations. No other transformations were performed.

Technical comments

- 54: move “indices” before “methylation”
- 63-64: “e.g.” should be added before the reference cited
- 67: “of the dependencies of brGDGTs on...”
- 114: no S at “temperature”
- 377: provide reference for the existing correlation.

Figure 7: replace r2 by R2 in the caption. In the legend p-value should be italicized and

with a small p.

- 428: R2 is relevant only to evaluate the quality of a regression model. To discuss the correlation between two variables it is more appropriate to mention the correlation coefficient (r) and its p-value.
- 459: explain what DC stands for l. 593: same remark for HP5

We thank the reviewer for these technical comments. All will be addressed in the revised manuscript.