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Interactive comment on "Predominance of hexamethylated 6-methyl branched glycerol dialkyl glycerol tetraethers in the Mariana Trench: Source and environmental implication" *by* Wenjie Xiao et al.

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Interactive comment on "Predominance of hexamethylated 6-methyl branched glycerol dialkyl glycerol tetraethers in the Mariana Trench: Source and environmental implication" by Wenjie Xiao et al. Anonymous Referee #1 Received and published: 31 October 2019

The manuscript by Xiao et al. presents a very interesting and welcome dataset on the distribution of brGDGTs in the Mariana Trench sediments. The results show distinct

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predominance of hexamethylated 6-methyl brGDGTs in this deepest ocean, suggesting brGDGTs are produced in situ by indigenous bacteria. This provides new insights into the composition of brGDGTs in “uncontaminated” marine and its usefulness as an endmember for brGDGTs source trace. It is suitable for Biogeosciences, but I have some suggestions/comments that could possibly improve the manuscript. Response: We appreciate the reviewer 1 to acknowledge the merit of our work. He/She also provided valuable suggestion to improve our manuscript. The follows are our response to each comment.

General Comments: 1. The current introduction dwells heavily on the history of brGDGTs, addressing their application as biomarkers for paleoclimate reconstruction, but falls short on giving much insight into the microbial ecology of this mysterious marine system. Oxygen concentration almost certainly plays a strong role in structuring the GDGT-producing microbial communities. Most brGDGTs were found produced by anaerobic bacteria. I strongly recommend the authors devote more space in the manuscript to discussing their data in the hydrologic and biogeochemical context of the MT.

Response: This is a good comment. Currently, there are two main themes of GDGT studies. 1) Calibration and application of GDGTs-derived proxies as temperature, pH and OC source indicators. 2) Mechanism of GDGT biosynthesis by microbes (archaea or certain bacteria) using molecular biology techniques. In the original submission, we paid more attention to the first theme. In the revised manuscript, we added the contents about microbial ecology of GDGTs in introduction part. For example, from line 62-66, we wrote as “In addition, oxygen (Qin et al., 2015) and moisture (Dang et al., 2016a) was found to play a profound role in GDGT-biosynthesis besides temperature and pH. By examining vertical patterns of brGDGTs and bacterial 16S rRNA gene in a deep meromictic Swiss lake (Lake Lugano), Weber et al. (2018) suggest that brGDGTs are synthesized by multiple groups of bacteria thriving under contrasting redox regimes.” We also cited these references. In the session 4.3, we added the discussion about

biogeochemical context in the Mariana Trench. From line 635 to 642, we wrote as "However, it should be pointed out that the bottom of Mariana Trench has the hydrostatic pressure > 100 MPa and is overlain by oligotrophic water masses with surface primary productivity of ca. 50 g OC m-2 yr-1 (Jamieson, 2015). Consequently, the unique microbes have been evolved in this extreme environment, such as proliferation of hydrocarbon-degrading bacteria (Liu et al., 2019), that may response to temperature and pH in a different way as their counterparts dwelling in shallow water regions. Nevertheless, the investigation of microbial community and intact polar lipids in the Mariana Trench is needed for understanding the source and environmental implication of brGDGTs in the deepest ocean." Please see our revised manuscript for the details.

2. I think that the paper would benefit from less discussion about the global brGDGT index application, which was already deeply discussed in Xiao et al. (2016). It dilutes the main conclusion of the paper, which is the distinct predominance of hexamethylated 6-methyl brGDGTs but also the absence of 5-methyl brGDGTs in the deepest ocean, this is an exciting result and again I hope the authors could devote more space on the explanations. Although some papers have already reported the dominance of hexamethylate brGDGTs in the marine sediments, none of them found the absence of 5-methyl brGDGTs. To me, it should be mainly driven by specific producers in this extreme environment than the environmental condition changes.

Response: We accepted this suggestion. BrGDGTs-derived proxies such as MBT, CBT and IBT are all developed based on terrestrial samples, and their correlations with environmental factors (e.g., temperature, pH) may be not suitable in marine settings. Thus, in the revised manuscript, we removed relevant contents about global application and calibration of brGDGTs. Specifically, we removed the figure 7 about a) CBT', b) IBT, c) CBT5me and d) #ringstetra index versus measured pH of globally distributed soils. We also deleted figs. 9b, c and d about global distributions of brGDGTs. The result and discussion about correlation between brGDGTs and environmental factors (temperature and pH) were also removed from the main text (e.g., the second

paragraph of session 4.4). Overall, in the revised manuscript, we emphasize the point about the predominance of 6-methyl brGDGTs and the absence of 5-methyl brGDGTs in the Mariana Trench.

3. The application of soil pH index CBT' and mean annual air temperature index MATmr in this marine setting is unconvincing. To be noticed, both CBT' and MATmr were established using stepwise forward selection, see De jonge et al. (2014) and Loomis et al. (2012), which are only suitable for terrestrial regions and have no mechanism behind compare to CBT/IBT or MBT.

Response: As mentioned above, we accepted the reviewer's suggestion and removed all relevant figures and discussion. It is true that brGDGTs-derived proxies such as MBT, CBT and IBT are all based on soil dataset, and could not be used for marine sediments directly if these compounds are produced by marine organisms. We keep this issue in our mind when revised the manuscript.

4. Results and discussion not completely separated. There is some overlap here, with data appearing in the discussion.

Response: We partly accepted this suggestion. We tried our best to move some unnecessary data presentation into the result part. However, the discussion can be benefited by brief data presentation.

5. The cross plot of acyclic hexa-/pentamethylated brGDGTs ratio and fractional abundance of brGDGT-IIIa' as a new approach to distinguish the terrestrial vs. marine provenance of brGDGTs (Fig. 10) can be removed. As the relationship between the (IIIa+IIIa')/(IIa+IIa') index and the BIT index (Fig. 6; Xiao et al., 2016) have already clearly separated the Mariana Trench sediments from the other samples. I think that it could be eliminated with no loss to the manuscript (If it remains, its value should be made clearer, why it's important compare to the index set before).

Response: We think the reviewer did not catch the point why we proposed the new

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source indicator for brGDGTs in marine settings. It is true that Xiao et al. (2016) already proposed the (IIIa+IIIa')/(IIa+IIa') index to distinguish source of brGDGTs (soil vs. marine with and without terrigenous influence). However, there is still overlap for the (IIIa + IIIa')/(IIa + IIa') values between soils and marine sediments, as shown in Fig. 6. So, it is necessary to develop more sensitive indicator. In our study, we found the combination of the (IIIa + IIIa')/(IIa + IIa') ratio and fractional abundance of brGDGT-IIIa' can completely separate samples with different terrigenous influence. The cross-plot of these two indicators results in distinct difference in the slope among soils, marine sediments with different terrigenous influence (please see fig. 9). Thus, our new approach provides two dimensional resolution to assess source of brGDGTs, whereas the (IIIa + IIIa')/(IIa + IIa') ratio by Xiao et al. (2016) is only one dimensional resolution. Furthermore, we found the slope of the (IIIa + IIIa')/(IIa + IIa') ratio and fractional abundance of brGDGT-IIIa' is applicable for sediment cores by compiling literature data. Given these facts, we keep figure 9 and discussion about our new approach to distinguish source of brGDGTs (Session 4.4: Deciphering brGDGT provenance in marine sediments) in the revised manuscript.

6. There is an excessive number of figures, leading to some redundancy. I would delete some overlapping ones and focus more on the main point (see specific points of clarification below).

Response: We accepted this suggestion and deleted the fig.7, 9b, 9c, and 9d in the revised manuscript.

Specific comments: Line 15: Leave space between numbers and symbols, keep consistent format for the left manuscript. Keep one decimal place for the relative abundance of brGDGTs for the following parts.

Response: We accepted this suggestion and added the space between number and unit throughout the manuscript.

Line 22-24: See my general comments

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Response: Please see our responses to the fifth general comment.

Line 36: Change the literature order in the reference. The early shown one should be named Weijers et al. (2007a)

Response: We made this correction in the revised manuscript.

Line 43-45: One sentence missing: While isoGDGTs were mainly produced in the marine realm. Thus...

Response: We added the sentence as “while iGDGTs are mainly produced in the marine realm” in line 52 of the revised manuscript.

Line 57: Schouten et al. (2007) is misused here.

Response: we removed Schouten et al. (2007) here.

Line 60: brGDGTs with cyclopentanes are not called hexa- or pentamethylated brGDGTs. Please correct this sentence.

Response: we accepted this suggestion and removed “hexa- or pentamethylated brGDGTs” here.

Line 111: Please cite Huguet et al. (2006) here.

Response: we accepted this suggestion and cited Huguet et al. (2006) in the revised manuscript.

Line 112: (3:1, v/v)

Response: Done.

Line 122: Italic m/z, correct followings.

Response: we accepted this suggestion and made correction throughout the manuscript.

Line 195: A description of HPLC/MS method in the Method Section is missing. Either

add in or cite a paper. If this is not a method developed by the authors, please rephrase the sentence.

Response: we accepted this suggestion. In the revised manuscript, we added a sentence as “The detailed instrumental parameters were described in Hopmans et al. (2016).”

Line 197: “...and hexamethylated (m/z 1050) brGDGTs in sediments of the Mariana Trench. This feature shows a distinct difference...”

Response: We accepted this suggestion. In the revised manuscript, we rewrote as “This feature is distinctly different from previous studies for other environmental settings that two or more peaks (5-methyl, 6-methyl and even 7-methyl isomers) were identified”.

Line 248-253: This part should go to results.

Response: we did not accept this suggestion. Because here we discuss the difference of brGDGT compositions between Mariana Trench and other globally distributed samples. We think it is needed to supply some summarized data about individual brGDGTs, so the readers can easily catch this point. In addition, we only used one sentence to describe the difference. Considering these facts, we still keep this sentence in the discussion part.

Line 242-263: As the authors mentioned two times in both introduction and method, this deepest trench is remote from any mainland and has no significant terrestrial influence. Either shorten it using one or two sentences or delete it.

Response: We accepted this suggestion, and shorten this as one sentence “This difference may reflect a difference in terrestrial influence since most marine samples in literatures are from continental margins where significant contribution of terrestrial-derived brGDGTs may mask the marine signal.” (line 468-471).

Line 264: ...is similar to those of distal marine sediments... Schouten et al. (2013) is

not a good reference the way it is written here.

Response: We accepted this suggestion and removed the reference of Schouten et al.(2013) here.

Line 266: Recheck the numbers you used here. 1354 soils and 589 marine sediments should be.

Response: We have corrected this mistake, and changed the numbers into “By compilation of globally distributed 1354 soils and 589 marine sediments”

Line 272-278: see my general comments. Section 4.2: Would it be nice to add in the data of this study for comparison, since all of the data the authors choose are sea sediments.

Response: Please see our responses to the general comments #2 and #3 above.

Line 333-336: Again results.

Response: we accepted this suggestion and deleted the data here. In the revised manuscript, we wrote the sentences as “This adaption mechanism may be extrapolated to marine organisms. In the Mariana Trench, in-situ production yields brGDGTs with the strong predominance of 6-methyl. The cyclopentane-containing brGDGTs (Ib, Ic, IIb, IIb’, IIc, IIc’, IIIb, IIIb’, IIIc, IIIc’) comprise less than 10% of total brGDGTs, and the #ringstetra index is low (Table 2)”.

Line 341-346: This is more a comparison of the indexes than the discussion of mechanisms behind. I would suggest to delete it. Only talking about the reasons for the predominance of IIIa’ here, 1) sedimentary in-situ brGDGTs-producers produce more hexamethylated brGDGTs to adapt to the low temperature/poor nutrient conditions (Sinninghe Damsté, 2016; Ding et al., 2018); 2) brGDGTs-producers adopt a strategy of the carbon skeleton isomerization of brGDGTs to live at alkaline seawater, resulting in a distribution in which 6-methyl brGDGTs are abundant (Ding et al., 2015; Xiao et al., 2015).

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Response: We agree with the reviewer on the explanation of adaption mechanism of brGDGTs-producers to environmental factors. In the revised manuscript, we emphasize the unique feature of brGDGTs in the Mariana Trench and tried to explain its potential reason in context of the extremely environmental condition in this deepest ocean. We also deleted all contents about calibration of brGDGT parameters at the global scales, such as removal of figure 7, 9b,c and d as well as relevant contents in main text.

Line 349-372: See my general comments, I would condense this part and delete the discussion that dilutes the main findings.

Response: Please see our responses to the general comments above.

Line 435: Recheck the format of references. Some DOI cannot be opened.

Response: we already double checked our references and format in the revised manuscript.

Fig. 2: Set a boundary between brGDGTs from Crenarchaeol, most external readers will not understand.

Response: We have highlighted Crenarchaeol with a rectangular.

Fig. 7: Not needed, see the general comments above

Response: We accepted this suggestion and deleted Fig.7.

Fig. 9: Delete Fig. b, c and d since they are showing the same results as a.

Response: We accepted this suggestion and removed Fig. 9b, c, and d.

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