

Referee #1

Summary

The main finding of this manuscript is that the Lake Magadi archaeal community has abruptly shifted between periods with prominent methane cycling and periods without over the last ~500 ky. The authors use organic geochemical measurements, isoGDGT indices and bulk organic matter $\delta^{13}\text{C}$ values, as indicators of methane cycling, with evidence for both methanotrophy and methanogenesis. Periods with strong methane cycling were associated proportionally lower hydrothermal inputs, indicated by lower REE abundance.

General comments

The research findings are new and interesting, and the methodology is sound. As previously stated, exploring unusual lakes like soda lakes and providing a context for organic geochemical applications is useful for future work on other unusual lakes (which there are many of in the world). I don't believe there are any major/pressing problems in the current version of the manuscript as they've largely been corrected during the first round of revisions. However, there are some smaller issues that should be resolved prior to publication. See below.

Specific comments

L 80-81: The optimum temperature of what?

Author Comment: This is supposed to read "However, the optimum temperature for crenarchaeol production is closer to 40-45 °C (Zhang et al., 2006)." This has been changed and rearranged on lines 78-79 for clarity.

Sect 2.1, paragraph 2-3: For the formations/beds named, is it known how deep the lake was when these formations/beds were deposited? Or more specifically, is the presence of a formation at certain place evidence of a specific lake depth? The phrasing of paragraph 2 implies that the formations/beds were deposited at a specific time at some point within very large age ranges (e.g., "Oloronga Beds in outcrop have been dated between ~800 and 300 ka" sounds like the beds were deposited sometime within this 500 ky range which is very large). Is this actually the case or does it actually mean the formation starts/ends in this range? If it's the former, then such large age range estimates are not very useful for any kind of chronological determinations. If it's the latter, the phrasing of paragraph 2 needs to be changed (e.g., "Oloronga Beds in outcrop spans ~800-300 ka" or "Oloronga Beds in outcrop were laid down from ~800 to 300 ka"). Later in sect 4.1.1, L 413-414 states, "Earlier water depths are also unclear because accommodation space was changing as the axial rift developed with faulting and subsidence." Is this true for most of the time period within which these formations/beds were laid down? If so, then it merits explaining early on (e.g., somewhere in sect 2.1) why past determinations of lake depth is so difficult.

Author Comment: The clarity has been added by saying that "The chert-bearing Oloronga Beds in outcrop have been dated were laid down between ~ 800 and 300 ka..." for clarity. This is only for the early formation of the lake and not for our samples collected where the lakebed has been relatively tectonically stable from ca. 540 ka to present.

Sect 2.1, L 116-120: The part starting from "More recently..." to "... High Magadi Formation" is not super relevant and can likely be condensed (into the previous paragraph). Discussing renaming/reassignment of beds/formations without giving additional context makes it confusing as there were multiple new names (e.g., "Evaporite Series") not mentioned in the previous paragraph."

Author Comment: This text has been removed and only the text which reads “Although Lake Magadi is situated near the equator, it lies in a rain shadow. Consequently, today it has a large moisture deficit (2400 mm evaporation versus 500 mm precipitation annually: Damnati and Taieb, 1995).” Was kept and was moved to the end of paragraph 1 of Section 2.1 per the reviewer’s suggestion.

Sect 2.1, end of paragraph 4: Per responses to the first round of reviews, the manuscript may benefit from explicitly stating that the reason for the non-ideal sampling strategy is due to poor core recovery/poor core quality. This can be done at author's discretion.

Author Comment: This has been added on L 131.

Sect 2.2: Remove the 2.2 heading "Leaf wax and bulk organic preparation and analysis. Now that the leaf wax protocol is gone, sections 2.2.1 and 2.2.2 can probably be their own sections, that is, 2.2 and 2.3. Subsequently, the following subsections will have to be renumbered.

Author Comment: We believe that the sections are sufficient to stay as-is per the suggestions of Referee #2 who suggested that we change the section title from “Leaf wax and bulk organic preparation and analysis” to “GDGT and bulk organic preparation and analysis”.

Sect 2.4: REE was defined as rare earth elements during the second instance it's mentioned (L 226) and not the first (L 222).

Author Comment: We have changed this discrepancy on L 217 and L 221.

Sect 2: Since pyrite evidence is mentioned frequently, the methods for pyrite measurement/detection should be included somewhere in section 2.

Author Comment: A description of pyrite assessment was added at the end of Section 2.4

Sect 4.1.1, paragraph 1 and 2: Suspected SMTZ presence is mentioned for the first time in paragraph 1, but evidence for a potential SMTZ at Lake Magadi is not presented until the next paragraph. L 317-319 states where SMTZ is suspected, we expect increases in methane GDGT indices, but L 340-342 states high methane GDGT indices contributes to evidence for SMTZ presence. The way this is written has some circular reasoning. The latter (using biomarker and geochemical evidence to make a statement) is more logical than the former (using an assumption to suggest expected patterns in biomarker and geochemical evidence).

Author Comment: Our logic of flow was to state the likely environment (SMTZ) which would result in our observations and then we explain how our results fit the environment and then conclude by stating that the combined evidence is likely pointing to it being an SMTZ environment. This seems logical to us.

L 385: If meromixis is a present characteristic of Lake Magadi, should explicitly state this fact in sect 2.1 study location description.

Author Comment: It is not a present characteristic of Lake Magadi, and the language has been clarified on L 381 by adding “in this study” to help define that these are not modern interpretations but paleo interpretations.

Sect 4.1.2, paragraph 3: Both Ca/Na and REEs are mentioned here. It may merit briefly

reminding/explaining what each represent (e.g., REEs come from hydrothermal inputs) as neither was explicitly defined in the context of Lake Magadi prior to this section.

Author Comment: Section 3.3 had the line “Increased values of REEs are characteristic of sodic systems influenced by hydrothermal springs, namely such as Mono Lake in California and this system (Johannesson and Lyons, 1994; Owen et al., 2019).” We have added an additional description of Ca/Na and why it is a useful metric for the core’ interpretations on L 286.

Sect 4.1.2, last paragraph: Delete "The CPI_{alk} and CPI_{FA} 510 averages were 4.6 and 5.0, indicating more terrestrial input." Consider also deleting the following sentence, "So, while these values are lower than Interval 3, and closer to the values in Interval 1, these still indicate a higher terrestrial input during this timeframe," as now there's no longer a discussion of terrestrial inputs. If this is deleted, then the only sentence left in the paragraph (starting with "Lastly, ...") would better be moved to the end of the previous paragraph to prevent having an orphan sentence.

Author Comment: The reviewer’s comments have been implemented in the manuscript.

Technical Corrections

Author Comment: All technical corrections were implemented and as for the L 586 comment about the manuscript number being “#XXX”, yes this will be changed once it is finally accepted for publication. This may take some extra time as our collaborator (Andrew S Cohen) who archived these numbers has recently passed away. The issue with coloration on Fig. 2 has also been resolved.

L 17-18: Replace "MI "switches off" ($MI < 0.2$); and on ($MI > 0.5$)" with "The MI switches "off" ($MI < 0.2$) and "on" ($MI > 0.5$)"

L 43: Change "generation of" to

"the"

L 69: Remove "Moreover"

L 110: Remove "More recently" as it is used to start the next

paragraph. L 120: Remove "Various"

L 121-122: This line, "Although Lake Magadi is situated near the equator, it lies in a rain shadow. Consequently, today it has a large moisture deficit (2400 mm evaporation versus 500 mm precipitation annually: Damnati and Taieb, 1995)" would better fit as the last line of sect 2.1, paragraph 1, rather than where it is currently at.

L 211: Change "was used here" to "is used here"

Equation 4: Un-bold the text

L 222: Change "intervals of focus (i.e., Intervals 1, 3, and 5)" to "intervals of focus in the sediment core (designated as intervals 1, 3, and 5 in the results and discussion sections)"

L 227-231: Condense these lines, starting from "Prior to performing...", to "The data was tested for normality via the built-in "Normality and Lognormality Tests" function in GraphPad®. Tests yielded lognormal distributions of each dataset and found the data to be non-normally distributed."

L 246: Is "197 to 149 ka" supposed to have a "ca." before it like the other intervals? L 249: Remove "Table SI" "Table SI"

L 262: Change "46.6815 m" to "46.68 m"

L 263: Remove spaces in "%GDGT-0 / cren"

L 277: Remove "Table SI" from the start of the sentences. L 291: Change "namely" to "such as"

L 344: This seems to refer to Figure 5 rather Figure 4.

L 345-346: This seems to refer to Figure 2 and 5 rather than 3 and 5.

L 346-347: Change "resulting from a more prevalent" to "as seen in a high".

L 354: Change "reported values" to "reported $\delta^{13}\text{C}_{\text{org}}$ values" (assuming Summons et al. (1998) reported the $\delta^{13}\text{C}$ for OM)

L 402: Put Table SI in parenthesis.

L 406-407: Split up the sentence at "and while", ending the first with "of the MI" and starting the next with "while both".

L 420: Flip "(MI-on; Fig. 2)" to "(Fig. 2; MI-on)" for consistency with the earlier parenthesis.

L 424: Change "such as hot spring mats in Thermoproteota" to "such as in hot spring mats made by Thermoproteota".

L 446-449: Shorten the two sentences (as they're currently a bit redundant) to something like "In agreement with the pollen record, the $\delta^{13}\text{C}_{\text{org}}$ values likely record a mixture of C4 grasses and C4 sedges. Similar $\delta^{13}\text{C}$ values were reported in C27 to C33 n-alkanes in equatorial regions of Cameroon, ranging from -18.2 to -17.6 ‰ and recording the signals from C4 grasses and sedges (Garcin et al., 2014)."

L 497: Can delete "(averaging 1.5 excluding the outlying value of 36.7). This average is closer to shallow Group I.1a Thermoproteota as described previously" as much of this was already stated in an

earlier sentence.

L 526-527: Add a comma between "Magadi ash" so it's "Magadi,

ash". L 545: Remove spaces in "Ca/ Na" so it becomes "Ca/Na".

L 551: This seems to refer to Fig. 4a rather than Sb.

L 558: Change "spring-runoff ratios" to "spring/runoff ratios" for consistency.

L 580: Should replace "RBO" with the full name of the co-author.

L 581: Change "special thank" to "special thanks".

L 582: Change "for providing, who provided" to "for

providing". L 584: Change "CSDF" to "CSD".

L 586: I assume "#XXX" will be replaced with an appropriate publication number once this paper is officially published?

Fig 2d: There seems to be a layer order issue with the data points in intervals 2, 4, and 6 plotting behind the yellow box instead of in front.

L 983-984: Remove the spaces between all the ratio indices (i.e., "%O/Cren" instead of "% O / Cren").

L 985-986: Remove "Checkered patterns indicate periods of tuffaceous silt deposit, which align with the low MI intervals" as there is no longer a checkered pattern in the figure.

L 991: Add "and in the Y-axis scale for 6^{13}CoM " after "X-axis scale".

L 995-996: Remove "The checkered pattern is indicative of periods of higher inferred hydrothermal flow" as there is no longer a checkered pattern in the figure.

Referee #2

I am satisfied with the changes made by the authors and would now support publication in Biogeosciences after a few final corrections:

Author Comment: Each of these have been completed. Figure S1 has been updated with the relevant information.

Abstract, l. 18: Please delete semicolon.

Title of section 2.2, l. 153: "Leaf wax" should be deleted (maybe replace by "GDGT")

Section 3.1, l. 249: Please check positioning of reference to table S1 (appears twice).

Section 4.1.1, l. 402: Please check positioning of reference to table S1.

Section 4.2, l. 551–554: References to Fig. 5a or b should be references to Fig 4a or b.

Fig. S1: Please label the main compounds in each chromatogram and add an explanation on the type of the chromatograms into the caption (e.g. TIC or ion filter?; LC chromatogram).