Dear editor,

We thank the Reviewer #1 for his/her comments. It seems to us that none of them present substantial criticism to any of our interpretations. Therefore, we will be able to swiftly incorporate all of his/her suggestions in our revised version, as we outline below in our replies to all of his/her individual comments.

Sincerely, on behalf of all authors,

Appy Sluijs

Sluijs et al. used the previously analyzed samples which were stored for over a decade. As I am interested in GDGTs, I was curious how the old and new GDGT data would differ, although I assume the offset would be small if stored properly and measured in good condition of the HPLC/MS. Figure 3 shows the result and regression analysis between the old and measured GDGTs results. Both TEX86 and BIT look comparable. However, I found that there are few outliers in the TEX86 dataset from the supplementary data. I plotted all their new vs old TEX86, and the R^2 value is lower to 0.66. Still comparable statistically, however, the authors did not mention about the outliers.
REPLY: These outliers represent data points for which the intensity of some isomers was insufficient in our reruns for proper quantification. For these 5 samples, TEX86 values were anomalously low as a consequence. These were the open fields in the spread sheet of the raw data but for clarity we have now marked them 'below detection' for the revised version of the manuscript. This further clarifies based on which data the 0.82 R^2 of Figure 3 is based.

I appreciate the authors for providing their valuable dataset and kindly included the spreadsheet calculation for the readers to follow. For RI (ring index), however, I found that the calculations were all missing while it can be calculated from the dataset. I calculated again from their data but the values were slightly different. The maximum difference between the reported value (column BX) and the calculation I did is up to 0.11 RI unit. Although the difference is small, this would impact on some of the samples that have _RI near 0.3, screening whether the data is reliable or unreliable near its cutoff value.
REPLY: We thank the reviewer very much for noticing this. The discrepancy was caused by an error in our excel calculations so that Cren isomer was not properly included. The numbers will be corrected in the revised supplement. The difference is indeed minor as the reviewer indicates and in fact it results in lower ΔRI and so we found no extra unreliable data points in our rescreening of the data.

Overall, I suggest a moderate revision of the manuscript, especially in the data analysis first, before it can be accepted by CP. Also, the manuscript contained plagiarism (line 160-163) and many run-on sentences which made it difficult in absorbing the information when reading, therefore, I suggest a more improvement in the scientific writing for the next version.
REPLY: We will make sure to reword this section and shorten sentences where necessary.

Some specific comments are below:
Line 20-21: add "ACEX"
REPLY: this shall be done

Line 20-52: the abstract seems to be too long and includes too much information of the study results in detail. Also, line 46-50 is just copied and pasted here from the main text (line 806-810).
REPLY: we will shorten the abstract significantly and avoid textual overlap with the rest of the text.

Line 37: the background SSTs in early Eocene generally exceed
REPLY: this shall be done

Line 71-77: run-on sentence: divided into two sentences
REPLY: this shall be done

Line 77-84: I understand citing all the references to supplement, however, 17 citations are too overloaded in one sentence for the reader. I suggest organizing the citation to where they would belong. For example, link and cite Pagani et al. (2006) with "molecular fossils" which examined the _D of n-alkane addressing the hydrologic cycle. Or breakdown the sentence and cite only the important references.
REPLY: we will cite three of the early papers that showed the potential for follow-up work.

Line 160-163: this sentence should be rephrased. It is exactly the same as written in Hollis et al. (2019) describing the BAYSPAR, but one word added here (plagiarism).
REPLY: this shall be reworded

Line 169: add the TEX86 value range of which converted SSTs differs between linear & non-linear
REPLY: This has been extensively discussed in the literature and it seems that the divergence occurs close to the maximum value in the calibration dataset (ca. 0.70), which we will include here.

Line 190-191: I suggest to remove "based on high BIT index value" and add the range of BIT results from the study after the equation.
REPLY: this shall be done and we will add a sentence reporting on a previous subjectively defined threshold assigned on the study section (Sluijs et al., 2009).

Line 207: specify the GDGT. If just GDGT, does it mean both iso- and brGDGts?
REPLY: this shall be specified to isoGDGT

Line 219-224: add the depth range of the deep contribution (Talyor et al., 2013) and also the reconstructed water depth of Site M00004A, meaning shallower shelf environ- ment, to connect the interpretation of negligible deep source.
REPLY: This will be done (>1000 m)

Line 233: use "[Crenarchaeol isomer]" for consistent compound name in all equations. this also implies for the names throughout the paper.
REPLY: This will be done

Line 234: "significant presence (or contribution) of anaerobic methanotrophy"
REPLY: This will be done

Line 242-243: provide references
REPLY: This will be done

Line 251: "Crenarchaeol isomer" for consistency of the compound name throughout the paper.
REPLY: We will stick to this wording throughout.

Line 299: I would rather suggest starting with 'brGMGTs' and supplement that this was previously reported as H-shaped brGDGTs, since the former is the major compound referred throughout the manuscript. Also, I suggest removing any description of 'Hshape brGDGTs' afterwards, as it makes it more confusing.
REPLY: This suggestion will be followed

Line 344: the precision of TEX86 unit or converted SSTs unit?
REPLY: We will explain that this regards the uncertainty calculated to the SST domain.

Line 351-353: same comment with line 344. In addition, I am confused with what "both" labs means.
REPLY: We will delete this confusing sentence.

Line 409: interval should be between 371.0 to “369.0” mcd, based on Figure 4 and Sluijs et al. (2009),
REPLY: Indeed, thank you for noticing.

Line 416-417: add the linear regression line in Figure 4 and supplement what “explaining 26 % of the variation” means
REPLY: This will be done and we will clarify the statement on variation; this number is taken directly from the R^2 (0.26) of the linear regression.

Line 428: I suggest to cite “Figure 6” in the first sentence, so the reader can easily compare the visualized data with the text, starting from the beginning of section or paragraph.
REPLY: This will be done

Line 442-452: Rather than directly moving on to the discussion of the method and result, I suggest to add a brief explanation of what lignite is and why lignite was used as the representative of terrestrial source for the readers to easily understand the concept.
REPLY: This will be done. We use the peat and lignite databases because they represent comprehensive datasets and therefore allow a rough calculation of the potential isoGDGT contribution.

Line 445: supplement how the absolute concentration is calculated (e.g. what standard used).
REPLY: The reference to absolute concentration was incorrect. It will be changed to raw signal intensity.

Line 467: “GDGT-2 and -3”. Suggest describing the compounds be consistent throughout the paper.
REPLY: This will be done

Line 473-478: This is true based on the isoGDGT distributions of Paleogene lignite. The reported lignite samples’ paleolatitudes are located within 57 _S to 48 _N, outside the Arctic region. Is there any lignite record from the Arctic that could be a more direct source to constrain the isoGDGTs distribution? If not, then how can this anomalous abundance of terrestrial isoGDGTs be explained in the Arctic where terrestrial input (especially from peats) is highly suggested while it has not been recorded elsewhere?
REPLY: We are not aware of any study that describes such high abundances of GDGT-3 nor a study that describes GDGT distributions from a northern high latitude Paleogene lignite, such as those described by Suan et al. (2017). In addition, we do not argue that peats are the main contributor to the terrestrial isoGDGT contribution. We merely include this exercise as a crude model for the potential terrestrial contribution to the isoGDGT pool in our ACEX samples, as we will better explain in the next version of the manuscript. Ideally, the analyses we perform here are also conducted using the abundance of isoGDGTs relative to brGDGTs in mineral soils to provide an even more complete picture, but those paired data are not available.

Line 486-487: add the threshold value of GDGT-2/Cren (Weijers et al., 2011), as it is shown as MI’s cutoff in the following.
REPLY: As far as we are aware, a formal threshold or cut off was never defined, but our values are clearly within the safe range of values described by Weijers et al. (2011), which is what we indicated here.

Line 492: I suggest the authors add a short interpretation of why these biomarker results are contrasted to the suitable depositional environment for abundant anaerobic archaea (methanotrophy and methanogen) which they indicated in the beginning of the section.
REPLY: This will be done

Line 508-510: interpreting BIT index with a distal position from the shoreline is problematic. Even in coastal marine or lacustrine settings, the BIT shows a large variation (Hopmans et al., 2004). Is the change of position interpreted from sea-level rise, similar to Sluijs et al. (2006)? Then what caused the sea-level rise (thermal expansion?) while the temperature proxy does not indicate significant warming?

REPLY: We do not only rely on the BIT index but also on palynological evidence that is consistent with a relative drop in terrestrial organic matter contribution. Relative sea level rise is clearly the simplest explanation for the observed changes. Sluijs et al. (2006) described sea level rise during the PETM that was later shown to be eustatic (Sluijs et al., 2008a). The interval described here regards an episode of relative sea level rise some time before ETM2. We are not aware of literature that has seen similar relative sea level rise elsewhere so we presume this relative sea level rise was of local, perhaps tectonic, origin. We will rephrase as follows:

"At ~371.2 mcd a drop in BIT index and a change in the palynological assemblages corresponds to an interval of greenish sediment, suggestive of pronounced amounts of glauconite. These changes are consistent with local relative sea level rise, causing a somewhat more distal position relative to the shoreline. However, the sediment remains dominantly siliciclastic and organic terrestrial components, particularly pollen and spores, remain abundant still indicating a shallow setting (Sluijs et al., 2008a; Sluijs et al., 2008b)."

Line 591: suggest the citation as "Figure 7b". This applies to other figure citations in the text to be more specific, when available, rather than just citing the whole figure. Another example is - line 606 to change to "Figure 7d"

REPLY: This will be done

Line 633-635: suggest to divide the two methods with (1) and (2), which the dashed line makes it confusing, and remove the linear/non-linear calibration description since these are already explained previously.

REPLY: We will include the 1) and 2) suggestion but we choose to keep the reminder to the reader regarding the linear vs non-linear calibrations.

Line 739: I find "lower temperature mean annual air temperature" very unclear.

REPLY: We will delete the first 'temperature' to solve this issue.

Figure 1: (1) the word 'using' is used repeatedly – remove or organize with a different word (2) add gplate webpage link for the readers and reference (3) describe or indicate what the brownish lines in the map

REPLY: We will rephrase the caption accordingly

Figure 2: (1) I suggest removing GDGT-4 since it is not discussed in the text nor measured in this study (see supplementary spreadsheet). Moreover, GDGT-4 is generally not included when calculating the relative fraction of isoGDGTs among the whole isoGDGTs pool. (2) add Crenarchaeol regioisomer's structure or note together with the Crenarchaeol (3) suggest changing "chemical structure" to "molecular structure"

REPLY: We will change accordingly. Specifically, we will remove isoGDGT-4 from the figure as suggested. We note in the caption of figure 2 that the Crenarchaeol isomer differs from Crenarchaeol in the stereochemistry of a cyclopentane moiety (Sinninghe Damsté et al., 2018), and replace 'chemical' with 'molecular' as suggested.

Figure 5: (1) describe the "modern peats" into two in the caption. (2) describe what the box and line, error bar, circles indicate (3) add the number of samples for statistical meaning

REPLY: This will be done

Figure 7: (1) I suggest 7d and 7e switch the order, since it is the
REPLY: This will be done

Supplementary material Data table: (1) a lot of blanks in the sample data, as well as an unknown words or sample core names below the data seat (see row 153-157).
REPLY: These open fields in the spread sheet of the raw data will be marked 'below detection' in the revised version of the manuscript.

(2) in "iGDGTs in peats" sheet, cite the references
REPLY: This will be done

(3) in "Lignite crenarchaeol", Sluijs et al. reported the GDGTs (iso- and br-) data originally from Naaf et al. (2018) and their newly measured 'Cren. Isomer'. Here, I suggest the authors to report the other iso and br-GDGTs abundances (here which I assume is HPLC/MS integrated peak area) together since they clearly mentioned in 'Material and Methods' that they re-analyzed the polar fraction of the lignite samples. Although I expect that this will not significantly change the result, still comparing only the newly measured 'Cren. Isomer' with reported GDGT dataset is not acceptable. This is because even measuring the same sample in the same method, the peak area can be different among interlaboratory measurements, the analytical parameter of the analytical instrument etc.
REPLY: We did not re-analyse these samples, but instead revisited the original chromatograms where we determined the peak area for the crenarchaeol isomer (i.e., Naafs et al., 2018). We have amended the text to make this clearer.

In addition, I suggest to add the calculations and results of the 'fraction of isoGDGTs' in all lignite samples. Lastly, minor comment on style of the table (e.g. missing cell borders, missing compound names) to be consistent. Describe 'n.d.' and 'b.d' too.
REPLY: We will add steps in our calculated values for fraction of isoGDGTs in lignites to our data supplement. In addition, to further facilitate reproducibility, we add an example calculation of the data presented in Supplementary Figure 1 and include the meaning of the abbreviations.

References

Naafs, B. D. A., McCormick, D., Inglis, G. N., and Pancost, R. D., 2018. Archaeal and bacterial H-GDGTs are abundant in peat and their relative abundance is positively correlated with temperature, *Geochim Cosmochim Ac*, 227, 156-170.

Sinninghe Damsté, J. S., Rijpstra, W. I. C., Hopmans, E. C., den Uijl, M. J., et al., 2018. The enigmatic structure of the crenarchaeol isomer, *Org Geochem*, 124, 22-28.

Sluijs, A., Schouten, S., Pagani, M., Woltering, M., et al., 2006. Subtropical Arctic Ocean temperatures during the Palaeocene/Eocene thermal maximum, *Nature*, 441, 610-613.

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