Reply to comments by Reviewer #2

We thank Dr. Dunkley-Jones for his review of our paper. He raises two points. Point 1 regards an as yet unpublished paper by Eley et al., and Point 2 regards the shape of the calibration between SST and TEX₈₆. We discuss these points below.

This is an excellent and thorough reassessment of organic biomarker temperature records for the latest Paleocene and early Eocene, derived from sediments recovered from the central Arctic Ocean. As demonstrated within the manuscript, this time of peak Cenozoic warmth is a key interval of interest to the paleoclimate community. Considerable proxy data and climate model efforts are focusing on this interval to address questions of climate sensitivity and the persistent problem of extreme polar warmth, which is indicated by the proxy data but is still problematic for climate model simulations. The late Paleocene to early Eocene also includes multiple hyperthermal events with millennial-scale onsets, which allow for the study of climate warming and ecosystem responses that approach the rates of modern environmental change. Two of these hyperthermal events are recovered within the ACEX record (PETM and ETM2). The biomarker-based temperature data from the Lomonosov Ridge is a critical latitudinal "end-member" for an assessment of polar warmth during the latest Paleocene and early Eocene. The unusual GDGT assemblages extracted from these samples, and the initial efforts to use these to estimate sea surface temperatures - which by necessity were non-standard - left some concern within the community about their reliability as predictors of absolute temperatures. This study re-evaluates this critical record with new analyses, including of glycerol monoalkyl glycerol tetraethers (GMGTs), and places this new data within the context of the past decade of studies on the calibration and use of GDGT-based thermometry.

Reply: We thank Dr. Dunkley-Jones for his support of our work.

Point 1. This study should be accepted for publication in Climates of the Past, although I do have one recommendation that I would like the authors to consider engaging with. Within this study they do a very thorough job of testing the potential controls and biases on GDGT assemblages using a range of indices and co-occurring markers for terrestrial-derived brGDGTs. The general aim of this is to screen GDGT assemblages, such that they can be separated into those that are formed in broadly analogous conditions to the modern marine system – and hence where the modern temperature dependency of assemblage composition can be well-modelled by the modern core-top calibration and those samples where the GDGT assemblage is significantly altered, by terrestrial input, methanogenesis or other processes, such that resultant estimates of SSTs may be biased. In their comprehensive treatment of this question of non analogue behaviour and biases, my only recommendation is that the authors also consider the methods proposed by Elev et al. (Climates of the Past Discussions, 2019) for the detection of ancient GDGT assemblages that are significantly nonanalogue to the modern calibration dataset. Below I include calculations of their Dnearest metric and OPTIMAL SST estimation for the new GDGT data presented by Sluijs et al. These results confirm some of the key findings of Sluijs et al. - that the pre-PETM GDGT assemblages are anomalous relative to the modern calibration dataset (Dnearest> 1); that there two clear shifts towards GDGT assemblages more "typical" of the modern at _385.0m, and then again at _375.0m. There is also an interval after the PETM, where TEX86 based temperatures remain high (>20 C), whilst OPTiMAL temperatures are considerably lower, with values in the high single figures (375 to 371 mcd). Sluijs et al. show that pre-ETM2 GDGTs have high BIT indices (377 to 371 m) and do not consider TEX86 derived temperatures from this interval to be robust because of the potential bias from terrestrial-derived material. The OPTiMAL methodology, however, indicates that these pre-ETM2 GDGT assemblages are relatively closely analogous to GDGT assemblages in the modern core top data (Dnearest <0.5), and that these "near neighbours" are formed in locations with modern MAT SSTs below 10 C.

The Eley et al. (2019) methodology – and the one applied by me below (Figure 1) – includes all modern core top data within the Tierney & Tingley (2015) database, including Arctic data associated with SSTs <3_C. These data were excluded from the standard BAYSPAR calibration ("NoNorth" /

"TT13" model of Tierney & Tingley, 2014), because in the Arctic region "TEX86 has a near-zero sensitivity to SST and therefore little predictability" and "incorporation of these data can negatively affect TEX86 predictability in the North Atlantic" (Tierney & Tingley, 2014). Although it would need to be tested – with OPTiMAL being run with and without these modern high-latitude data points and then applied to the ACEX core – it is possible that modern Arctic GDGT assemblages are the "nearest neighbours" of the pre-ETM2 GDGT assemblages, whereas above _371 mcd, assemblages shift to a more normal open marine assemblage, as inferred by Sluijs et al. on the basis of BIT indices. This may, in part, account for the significant warming suggested by OPTiMAL across this transition, and further work would be needed to investigate the inclusion or exclusion of modern Arctic GDGT assemblages in the modern calibration for OPTiMAL, and the ability to extract temperature information from these GDGT assemblages using proxy formulations other than the TEX86 index. Regardless of this, the consideration of the OPTiMAL approach con- firms - through an independent approach that is agnostic about the form or "model choice", of the GDGT – SST relationship - that Arctic SSTs around ETM2 were in the region of _20_C (OPTiMAL) or higher (TEX86H, BAYSPAR).

Reply to point 1. We are aware of the Eley et al. manuscript and their new indicators are certainly potentially interesting for this paper. As far as we can see on the CP website, however, this paper is under evaluation and, considering the online discussion, may be subject to significant revisions. This compromises the use of this method in its present form.

We have nevertheless considered the Dnearest and OPTiMAL records kindly provided by the reviewer. Certainly, as the reviewer indicates, some of the results are consistent with our results (e.g. during the latest Paleocene) and could in principle be used as support for our statements. Other aspects are inconsistent with what we find. For example, the interval 377-371 m discussed by the reviewer is particularly interesting. Although the Dnearest metric suggests that TEX₈₆ values are robust, the delta Ring Index, a simple metric to evaluate whether GDGT distributions are similar to modern ocean core top data (Zhang et al., 2011), clearly indicates the GDGT assemblage is compromised for reliable TEX86 paleothermometry. The BIT index (>0.4 to 0.8), the palynological assemblage (dominated by terrestrial organic matter) the dinoflagellate cyst assemblage (almost completely freshwater-tolerant) and bulk organic carbon stable carbon isotope ratios are also inconsistent with a dominant marine source of the organic matter (Sluijs et al., 2008; Sluijs et al., 2009; Sluijs and Dickens, 2012). This would call into question the reliability of the Dnearest metric as an indicator of "normal" marine isoGDGT assemblages, even if they are consistent with some of the modern core top data. Therefore, it seems to us that any discussion on this topic would rather serve as a test to the as yet unpublished new methods proposed by Eley et al. rather than contributing to the main goal of our paper, to reconstruct late Paleocene - early Eocene Arctic paleotemperature.

Collectively, we therefore prefer to not discuss these results at this point.

Point 2. Section 2.1 and especially Lines 145 – 147: suggest that culture and mesocosm experiments and surface sediment data indicate a linear relationship but without a clear citation of these studies. Rather, the citations seem to be of the studies that demonstrate a deviation from linearity. As the authors implicitly acknowledge - with statements like "suggest a linear relation" (line 146) or "assumes a linear relationship" (line 160) - the most appropriate form of the TEX86 – SST relationship is uncertain, with current calibration models making some degree of assumption about the best fit relationship between core top TEX86 data and SSTs. I would suggest a slight rephrase to acknowledge this uncertainty and appropriate citations to back up any arguments made about the form of the TEX86 – SST relationship. There is extensive discussion of the assumptions that can be made about the form of the TEX86 – SST relationship within the online discussion to Eley et al. (2019) that address this issue, between those who argue for an assumed linear response (Tierney) and those who question this

assumption (Eley et al.) – some of the relevant response to Tierney quoted below from Eley et al. (https://www.climpast-discuss.net/cp-2019-60/cp-2019-60-AC1-supplement.pdf):

"We agree that there is a basic underlying trend for more rings within GDGT structures at higher temperatures (Zhang et al. 2015; Qin et al., 2015). What we dispute is that this translates into a simple linear model at the community scale (core top calibration dataset), or is yet reproduced with consistency between strains in laboratory cultures, including the temperature-dependence of GDGT ring numbers within the marine, mesophilic Thaumarchaeota in Marine Group 1 (broadly equivalent to the old Crenarchaeota Group 1) (Eilling et al., 2015; Qin et al., 2015; Wuchter et al., 2007). Wuchter et al. (2004) and Schouten et al. (2007) show a compiled linear calibration of TEX86 against incubation temperature (up to 40_C in the case of Schouten et al., 2007) based on strains that were enriched from surface seawater collected from the North Sea and Indian Ocean respectively. Like Qin et al., (2015) we note the nonlinear nature of the individual experiments in Wuchter et al., 2004 (see Wuchter et al., 2004 Fig. 5). Moreover, the relatively lower Cren' in these studies yield a very different intercept and slope (compared to core-top calibrations e.g. Kim et al. 2010) meaning that the resulting calibrations for TEX86 cannot be applied to core-tops. This was recognised by Kim et al. (2010), who state "but we may speculate that Marine Group I Crenarchaeota species in the enrichment cultures are not completely representative of those occurring in nature...

...As we state above, although we agree that there is a basic underlying trend of increasing ring number with increasing growth temperature, we do not agree that this is well enough known to be quantified into a "basic relationship" that can be "enforced" as a particular model form. Rather, there is uncertainty in the appropriate form of the relationship even within the modern calibration data (see Kim et al. 2010) which becomes substantial beyond the calibration range. The spatial structuring of residuals in global models of modern TEX86 temperature dependence (Tierney & Tingley, 2014) and clear structuring of residuals with temperature in our and other GDGT- temperature calibrations, are likely indications of transitions in the ecology, community make-up or habitat of modern GDGT producers that are not well constrained. We argue that this complexity in the GDGT temperature responses in the modern oceans should be grounds for caution when applying empirical models from the modern to ancient conditions, especially when working with the subset of ancient assemblage data for which there is no modern analogue."

Reply to point 2. We fully agree with the reviewer here. In fact, some of us have argued for the possibility of a non-linear relation ourselves in a recent paper (Cramwinckel et al., 2018). We will adapt the text in section 2.1 to fully reflect the current status of the discussion (e.g., Hollis et al., 2019).

References

- Cramwinckel, M. J., Huber, M., Kocken, I. J., Agnini, C., Bijl, P. K., Bohaty, S. M., Frieling, J., Goldner, A., Hilgen, F. J., Kip, E. L., Peterse, F., van der Ploeg, R., Röhl, U., Schouten, S., and Sluijs, A.: Synchronous tropical and polar temperature evolution in the Eocene, Nature, 559, 382-386, 10.1038/s41586-018-0272-2, 2018.
- Hollis, C. J., Dunkley Jones, T., Anagnostou, E., Bijl, P. K., Cramwinckel, M. J., Cui, Y., Dickens, G. R., Edgar, K. M., Eley, Y., Evans, D., Foster, G. L., Frieling, J., Inglis, G. N., Kennedy, E. M., Kozdon, R., Lauretano, V., Lear, C. H., Littler, K., Lourens, L., Meckler, A. N., Naafs, B. D. A., Pälike, H., Pancost, R. D., Pearson, P. N., Röhl, U., Royer, D. L., Salzmann, U., Schubert, B. A., Seebeck, H., Sluijs, A., Speijer, R. P., Stassen, P., Tierney, J., Tripati, A., Wade, B., Westerhold, T., Witkowski, C., Zachos, J. C., Zhang, Y. G., Huber, M., and Lunt, D. J.: The DeepMIP contribution to PMIP4: methodologies for selection, compilation and analysis of latest Paleocene and early Eocene climate proxy data, incorporating version 0.1 of the DeepMIP database, Geosci. Model Dev., 12, 3149-3206, 10.5194/gmd-12-3149-2019, 2019.

- Sluijs, A., Röhl, U., Schouten, S., Brumsack, H.-J., Sangiorgi, F., Sinninghe Damsté, J. S., and Brinkhuis, H.: Arctic late Paleocene–early Eocene paleoenvironments with special emphasis on the Paleocene-Eocene thermal maximum (Lomonosov Ridge, Integrated Ocean Drilling Program Expedition 302), Paleoceanography, 23, PA1S11, doi:10.1029/2007PA001495, 2008.
- Sluijs, A., Schouten, S., Donders, T. H., Schoon, P. L., Röhl, U., Reichart, G. J., Sangiorgi, F., Kim, J.-H., Sinninghe Damsté, J. S., and Brinkhuis, H.: Warm and Wet Conditions in the Arctic Region during Eocene Thermal Maximum 2, Nature Geoscience, 2, 777-780, 2009.
- Sluijs, A., and Dickens, G. R.: Assessing offsets between the δ^{13} C of sedimentary components and the global exogenic carbon pool across Early Paleogene carbon cycle perturbations, Global Biogeochemical Cycles, 26, GB4005, doi:10.1029/2011GB004224, 2012.