

### **Referee comment #3:**

Guo et al. generated new records based on branched glycerol dialkyl glycerol tetraethers (brGDGTs) from the Chinese Loess Plateau (CLP) over the last 130 kyrs. The authors found that two pH-sensitive brGDGT-based indices, DC and IR, showed contrasting temporal changes at the same site. After a comparison of the new brGDGT-based records with several published records from the same site, such as another biomarker-based record (ice-corrected  $\delta^2\text{H}_{\text{wax}}$ ), the authors found that DC showed promise as a mean annual precipitation (MAP) proxy. Then, the authors investigated the relationships between brGDGT-based indices (DC and IR) and soil pH and MAP using a global modern soil dataset, as well as a CLP modern soil dataset. The authors found that in alkaline soils, including within the CLP, DC showed a strong correlation with MAP, which enabled the development of a DC-MAP calibration for quantitative MAP reconstructions. The authors then applied their DC-MAP calibration at three sites within the CLP, including the study site, and investigated spatial differences in MAP within the CLP and their changes through time. The authors also did spectral and cross-spectral analyses and found that (i) their DC record showed precession and obliquity signals, contrary to  $\delta^2\text{H}_{\text{wax}}$  and  $\delta^{18}\text{O}_{\text{speleorecords}}$  which only showed the precession signal, and (ii) their DC record was in phase with  $\delta^2\text{H}_{\text{wax}}$  and  $\delta^{18}\text{O}_{\text{speleorecords}}$  at the precession scale. The authors thus concluded that Northern Hemisphere summer insolation was a direct forcing of precipitation amount rather than the result from confounding factors on precipitation records based on isotopes.

As a paleoclimatologist with expertise on GDGT-based proxies, I have read this manuscript with interest for several reasons. First, the authors tackled a topical and important subject, namely the understanding of the East Asian Monsoon which also has its own controversies and subjects of debate, as the authors stated in the Introduction. Second, this manuscript presents an interesting use of a brGDGT-based proxy, namely DC as a (quantitative) MAP

proxy, as brGDGTs are classically used for quantitative reconstructions of land temperature and soil pH. Third, the authors reconstructed past MAP changes using a proxy that does not involve hydrogen or oxygen isotopes, contrary to  $\delta^2\text{H}_{\text{wax}}$  and  $\delta^{18}\text{O}_{\text{speleo}}$ , which strengthens the independence of DC relative to  $\delta^2\text{H}_{\text{wax}}$  and  $\delta^{18}\text{O}_{\text{speleo}}$ . Furthermore, I found the manuscript easy to read and well-organized. Overall, this piece of work would be a great addition to the literature and is worth publishing in *Climate of the Past*.

However, I have several comments, questions, and suggestions for revision, which I detail below.

*Reply: We thank the reviewer for their positive evaluation of our work. Please find our point-by-point response below in italic.*

General comments:

1) Recently, Zhao et al. (2020) introduced a precipitation index (PI) as follows:  $\text{PI} = (\text{Ia} + \text{Ib}) / (\text{Ia} + \text{Ib} + \text{IIIa} + \text{IIa}' + \text{IIIa}')$ . Like the redefined DC, the PI takes advantage of the improved separation of 5- and 6-methyl brGDGT isomers. Importantly, Zhao et al. (2020) and Zhang et al. (2024) proposed PI-MAP calibrations in cancellous bones and soils, respectively, for brGDGT-based quantitative MAP reconstructions. Accordingly, I would like to know the authors' thoughts concerning the PI. Specifically, the authors may check how well the PI would behave as a (quantitative) MAP proxy compared with e.g., DC and IR in the CLP and, in case of similar trends between DC and PI, which MAP reconstructions the PI would yield in the CLP. However, the authors do not need to switch to the PI, especially if DC has a better motivation and/or shows more meaningful results compared with the PI in the authors' view.

Zhang, T., Han, W., Tian, Q., Zhang, J., Kemp, D. B., Wang, Z., Yan, X., Mai, L., Fang, X., and Ogg, J.: Tectonically controlled establishment of modern-like precipitation patterns in East and central Asia during the early late Miocene, *Journal of Geophysical Research: Atmospheres*, 129, e2024JD041025, <https://doi.org/10.1029/2024JD041025>, 2024.

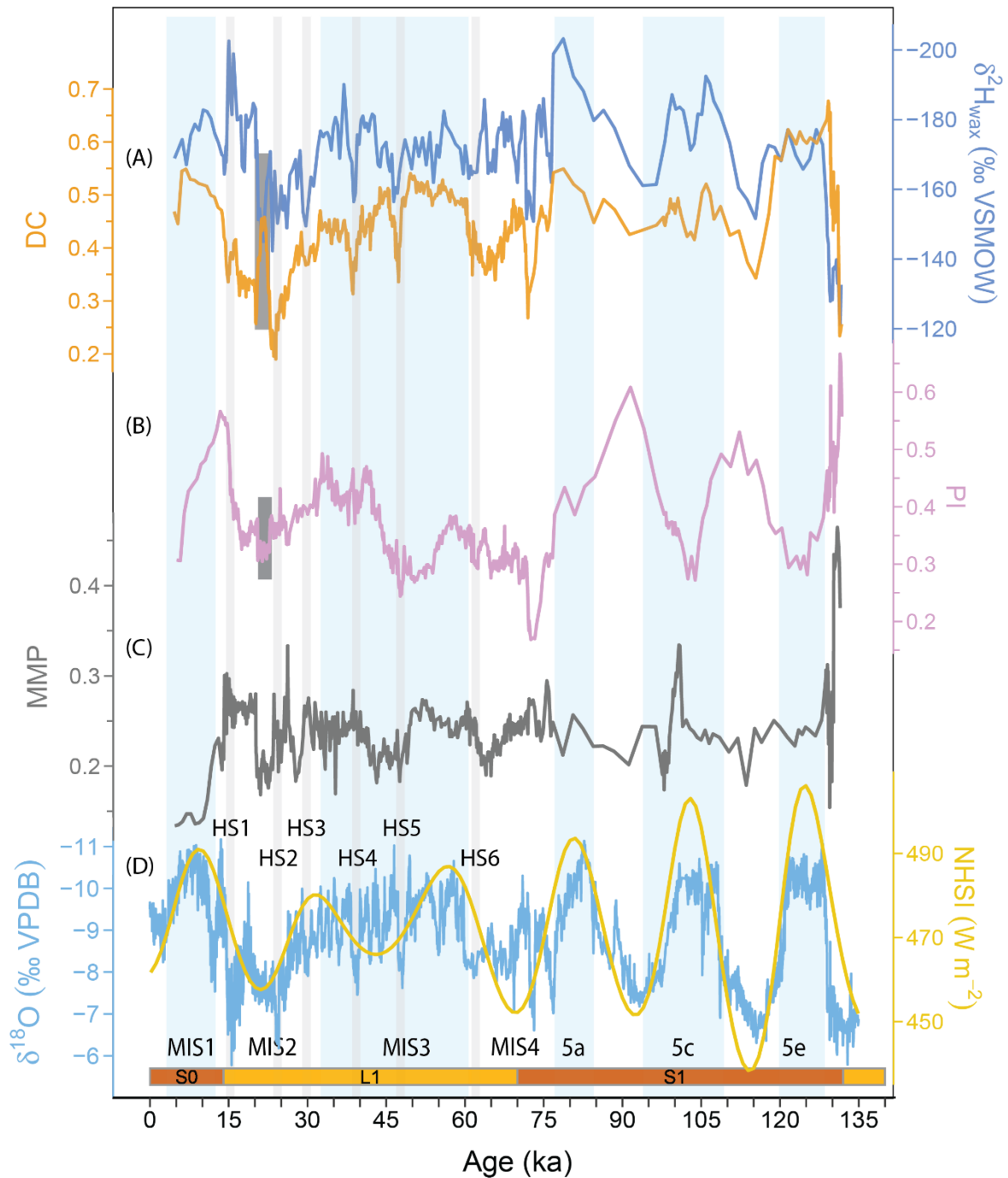
Zhao, J., Huang, Y., Yao, Y., An, Z., Zhu, Y., Lu, H., and Wang, Z.: Calibrating branched GDGTs in bones to temperature and precipitation: application to Alaska chronological sequences, *Quaternary Science Reviews*, 240, 106371, <https://doi.org/10.1016/j.quascirev.2020.106371>, 2020.

*Reply: We thank the reviewer for their comments and providing the references. In previous studies, the precipitation index (PI) has shown a positive correlation with precipitation in bones and surface soils from China (Zhao et al., 2020; Zhang et al., 2024). The application of the PI in the Yuanbao section, however, results in a record with higher values during stadials than interstadials during MIS5, which is inconsistent with other hydroclimate proxies from the same section (Fig. 1B), such as MagSus and  $\delta^2H_{wax}$ . In addition, compared to the DC, the PI fails to capture millennial-scale events during the last glacial period.*

*Upon closer examination, the PI equation combines the degree of methylation (MBT), the position of the methylation (IR), and the degree of cyclization (DC), making it complex to interpret which membrane adjustment is driving the changes in the PI record. Specifically, having the tetramethylated brGDGTs in the numerator and the penta- and hexamethylated brGDGTs in denominator resembles the structure of the  $MBT'_{5Me}$ , which is linked to temperature -and not precipitation- in the global soil dataset (De Jonge et al., 2014). In addition, the PI was initially developed based on distributions of brGDGTs in bones, where bacterial community compositions might differ from those in (arid and alkaline) soils and loess and paleosol materials.*

For these reasons, we have decided not to include the PI in our revised manuscript.

Nevertheless, it would be interesting to test the PI in different types of material in future work.



**Fig. 1** Biomarker-based records for the past 130 kyr at Yuanbao. (A) Degree of cyclization (DC) of brGDGTs and ice-corrected  $\delta^2H_{wax}$  based on plant waxes in the same lipid extracts (Fuchs et al., 2023). (B) brGDGT-based precipitation index (PI) (Zhao et al., 2020). (C)

*isoGDGT-based mean monthly precipitation (MMP) record (De Jonge et al., 2024). (D) NHSI at 35°N (Berger et al., 2010) and the composite speleothem oxygen isotope ( $\delta^{18}O$ ) record (Cheng et al., 2016). Dark grey intervals (~23–21 ka) in brGDGT-related records (DC and PI) indicate the transition from the outcrop to the pit and are not considered in the interpretation of the records.*

2) Even more recently, De Jonge et al. (2024a) proposed another GDGT-based proxy which may track precipitation changes, specifically mean monthly precipitation (MMP) changes:  $MMP = (isoGDGT-1 + isoGDGT-3)/(isoGDGT-1 + cren)$ . I recognize that the involved GDGTs are isoGDGTs rather than brGDGTs and that the alternative GDGT-based index would likely yield uncertain reconstructions as well (see the [Eq. 14] versus MMP plot in Supp. Fig. 8 in De Jonge et al., 2024a). Nevertheless, provided that isoGDGTs are abundant enough to yield peak areas above quantification limit, I feel that the authors may consider this isoGDGT-based ratio as well.

De Jonge, C., Guo, J., Hällberg, P., Griepentrog, M., Rifai, H., Richter, A., Ramirez, E., Zhang, X., Smittenberg, R. H., Peterse, F., Boeckx, P., and Dercon, G.: The impact of soil chemistry, moisture and temperature on branched and isoprenoid GDGTs in soils: a study using six globally distributed elevation transects, *Organic Geochemistry*, 187, 104706, <https://doi.org/10.1016/j.orggeochem.2023.104706>, 2024a.

*Reply: We thank the reviewer for highlighting this isoGDGT-based precipitation proxy. We have applied the proxy to our loess section (Fig. 1C). However, the resulting record does not show a trend that could be linked to any climate events, regionally or globally. Although the work of De Jonge et al., (2024) suggests that isoGDGTs in soils have potential as a proxy for precipitation reconstructions, the drivers of isoGDGT distributions in arid and alkaline soils, like loess, remain elusive. At least, the mean monthly precipitation (MMP) record we obtained for Yuanbao suggests that precipitation may not be the dominant control. We have therefore decided not to include this record into our revised manuscript.*

Detailed comments:

Main text

Line 11: In “Chinese Loess Plateau”, “loess” is not capitalized in the abstract, but is in line 40 in the Introduction.

*Reply: We will correct this in the revised manuscript.*

Line 48: Which paper by Guo et al. (2022) is cited here? The one published in Geology (Guo et al., 2022a)?

*Reply: It is the Geology one indeed. We will make it clear in the revised manuscript.*

Line 98: Which paper by Guo et al. (2022) is cited here? The one published in Organic Geochemistry (Guo et al., 2022b)?

*Reply: We thank the reviewer for their careful check, this is the one published in Organic Geochemistry. We will make it clear in the revised manuscript.*

Fig. 1 (also Fig. S3): Readers may find it hard to read the coordinate labels for the inset which shows the relevant wind patterns: the authors should consider changing the color from black to white, as they did for other labels within the larger map, and/or increasing the font size.

*Reply: We will modify this accordingly in the revised manuscript.*

Lines 142–143: Replace “(De Jonge et al., 2014a)” with “De Jonge et al. (2014a)”.

*Reply: We will replace this in the revised manuscript.*

Fig. 2: For panel A, the authors should pick a color pair different from the current green-orange one for the sake of accessibility to color-blind readers. For panel B, the authors could consider picking a color pair with a stronger contrast in terms of hue and/or lightness for the sake of readability.

*Reply: We will change this in the revised manuscript.*

Line 204: Replace “this event only last” with “this event only lasts”.

*Reply: We will correct this in the revised manuscript.*

Fig. 4: For panels A–D, the authors should consider revising the colors to avoid the green-orange confusion for color-blind readers. Alternatively, the authors should distinguish the CLP datapoints from the other ones using a different symbol type, for instance with squares, diamonds, or triangles rather than circles. If the authors pick the second option, then the change in symbol type for CLP should be reflected in panels E and F as well.

*Reply: We will modify this accordingly in the revised manuscript.*

Lines 247–248: The  $r^2$  value represents the percentage of variance explained by the regression, not the correlation strength which is represented by the  $r$  value.

*Reply: We will change this in the revised manuscript.*

Lines 255–258: This is an important and welcome remark.

*Reply: We thank the reviewer for their positive feedback.*

Line 294: “(i.e.,  $\delta^{2}\text{H}_{\text{wax}}$  (Fuchs et al., 2023), speleothem  $\delta^{18}\text{O}$  (Cheng et al., 2016))”: A few parentheses should be removed so that only a pair of parentheses remains.

*Reply: We have carefully checked and will make sure they are well in order in the revised manuscript.*

References: Could the authors recheck their reference list? The formatting appears a bit suboptimal at places, for instance in lines 367–368 (Baxter et al., 2019) where I spotted a “ScienceDirect” which appears out of place there, as well as in lines 537–539 (Wang et al., 2001) where I spotted an unexpected “(80-. ).” just after the journal name.

*Reply: We thank the reviewer for their careful check. We will carefully go through the reference list and correct the formatting.*

#### Supplementary Figures

Fig. S1: It would be great if the authors could write the  $m/z$  values of  $[\text{M}+\text{H}]^+$  ions with at least one decimal place rather than as integer values. Otherwise, researchers who would examine GDGTs for the first time may fail to do optimal GDGT analyses for the reasons discussed by Davtian et al. (2018) and partly reminded by De Jonge et al. (2024b).



Davtian, N., Bard, E., Ménot, G., and Fagault, Y.: The importance of mass accuracy in selected ion monitoring analysis of branched and isoprenoid tetraethers, *Organic Geochemistry*, 118, 58–62, <https://doi.org/10.1016/j.orggeochem.2018.01.007>, 2018.

De Jonge, C., Peterse, F., Nierop, K. G. J., Blattmann, T. M., Alexandre, M., Ansanay-Alex, S., Austin, T., Babin, M., Bard, E., Bauersachs, T., Blewett, J., Boehman, B., Castañeda, I. S., Chen, J., Conti, M. L. G., Contreras, S., Cordes, J., Davtian, N., van Dongen, B., Duncan, B., Elling, F. J., Galy, V., Gao, S., Hefter, J., Hinrichs, K.-U., Helling, M. R., Hoorweg, M., Hopmans, E., Hou, J., Huang, Y., Huguet, A., Jia, G., Karger, C., Keely, B. J., Kusch, S., Li, H., Liang, J., Lipp, J. S., Liu, W., Lu, H., Mangelsdorf, K., Manners, H., Martinez Garcia, A., Menot, G., Mollenhauer, G., Naafs, B. D. A., Naeher, S., O'Connor, L. K., Pearce, E. M., Pearson, A., Rao, Z., Rodrigo-Gámiz, M., Rosendahl, C., Rostek, F., Bao, R., Sanyal, P., Schubotz, F., Scott, W., Sen, R., Sluijs, A., Smittenberg, R., Stefanescu, I., Sun, J., Sutton, P., Tierney, J., Tejos, E., Villanueva, J., Wang, H., Werne, J., Yamamoto, M., Yang, H., and Zhou, A.: Interlaboratory comparison of branched GDGT temperature and pH proxies using soils and lipid extracts, *Geochemistry, Geophysics, Geosystems*, 25, e2024GC011583, <https://doi.org/10.1029/2024GC011583>, 2024b.

*Reply: We thank the reviewer for their careful review and detailed suggestions. We will add this important information to the Method and supplementary figure in the revised manuscript to avoid any potential misleading.*

## **References**

Berger, A., Loutre, M.-F., and Yin, Q.: Total irradiation during any time interval of the year using elliptic integrals, *Quat. Sci. Rev.*, 29, 1968–1982, <https://doi.org/10.1016/j.quascirev.2010.05.007>, 2010.

Cheng, H., Edwards, R. L., Sinha, A., Spötl, C., Yi, L., Chen, S., Kelly, M., Kathayat, G., Wang, X., Li, X., Kong, X., Wang, Y., Ning, Y., and Zhang, H.: *The Asian monsoon over the past 640,000 years and ice age terminations*, *Nature*, 534, 640–646, <https://doi.org/10.1038/nature18591>, 2016.

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De Jonge, C., Guo, J., Hällberg, P., Griepentrog, M., Rifai, H., Richter, A., Ramirez, E., Zhang, X., Smittenberg, R. H., Peterse, F., Boeckx, P., and Dercon, G.: *The impact of soil chemistry, moisture and temperature on branched and isoprenoid GDGTs in soils: A study using six globally distributed elevation transects*, *Org. Geochem.*, 187, 104706, <https://doi.org/10.1016/j.orggeochem.2023.104706>, 2024.

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2020.