Referee comments #1 (Dr. David Naafs):

Summary

In this manuscript the authors reconstruct east Asian monsoon dynamics over the last 130 kyrs using biomarkers preserved in loess. Newly generated records of changes in the distribution of brGDGTs are combined with published records of plant wax d2H from the same section. The authors develop a quantitative method to reconstruct changes in MAP using brGDGTs preserved in loess and demonstrate that precipitation in the Chinese loess plateau varies at the precession and obliquity scale, the former indicating the northern hemisphere as a main driver of monsoon precipitation.

Main assessment

The manuscript provides a large amount of new data that are used to support novel insights into our understanding of the east Asian monsoon. This type of manuscript will be of interest to the readers of CoP. In addition, the newly proposed method to quantify precipitation using brGDGTs in loess will be of interest to organic geochemists. The manuscript was pleasant to read and the figures clear and informative.

However, my main criticism is that the manuscript and main conclusions rely on a limited set of brGDGT-based indices: DC and, to some extent, IR. However, other brGDGT indices are influenced by pH (and thus precipitation), for example the well-established CBT index for brGDGTs. In addition, other GDGTs like crenarchaeaol and the BIT index can be used to infer changes in hydrology in terrestrial sections, as highlighted in the introduction of this manuscript. However, these complementary methods are not used here. Rather, the manuscript relies on the application of less often used indices like DC. There is no explanation why these other GDGT-based indices are not used, while they are measured. I

assume they are excluded because they show different results? But these other proxies could provide additional insights into changes in hydrology in this region.

I therefore recommend moderate revisions. In the revised manuscript I would like to see an expanded discussion on the other GDGT based proxies (e.g., CBT, BIT, %cren) and justification for why they are not used here to assess changes in hydrology. Or better, they are included to obtain a more holistic reconstruction of EASM dynamics across the late Quaternary.

Reply: We thank Dr. Naafs for his positive evaluation of our work and constructive feedback. We have taken their suggestions into careful consideration and will make changes in the revised manuscript accordingly. Please find our point-by-point response below in italic.

A similar comment has also been made by Referee #2. Note that the focus on just the DC and the IR as potential proxies for monsoon precipitation is clearly motivated in the introduction of our manuscript. Specifically:

i) Although the BIT index and Ri/b have been linked to hydroclimate, in loess-paleosol sequences, they are used as indicators of mega-drought events and only in a qualitative way (e.g., Xie et al., 2012; Yang et al., 2014; Tang et al., 2017). Since the aim of this manuscript is to reconstruct monsoon precipitation quantitatively, we have not included these records here. Nevertheless, the BIT index and Ri/b in the Yuanbao section are relatively invariable and do not exceed the established threshold values (i.e., 0.5 for the R_{<i>i/b}; *Tang et al., 2017) that indicate the occurrence of mega-drought events at this site over the past 130 kyr (Fig. 1E).*

ii) The environmental controls that influence the distribution of isoGDGTs in soils is still *being studied, and no clear link between isoGDGTs in loess and environmental parameters has been demonstrated yet. As such, the %cren does not show a clear trend in the Yuanbao*

record (Fig. 1D). Nevertheless, the isoGDGT data will be provided as supplementary material upon acceptance for the community's reference.

iii) The use of CBT as a proxy for monsoon precipitation has been mentioned in the introduction (L93-96), as are the reasons for not using CBT and/or CBT′. Namely, the first application of this proxy shows that CBT reflects monsoon pacing (i.e., qualitative) rather than absolute precipitation amounts (Peterse et al., 2014). Secondly, the CBT(′) is in fact a combination of the degree of cyclization (DC) of brGDGTs and the relative abundance of 6 methyl isomers (IR). However, the clearly opposite correlations of the DC and the IR with soil pH in soils with pH >7.5 raises concerns about the meaningful interpretation of CBT(′) in loess sequences (e.g., Xie et al., 2012; Guo et al., 2022). Hence, to determine the relationship between hydroclimate and brGDGT distributions, we have deliberately split the CBT(′) into the DC and the IR. As can be seen in Fig. 1B, the CBT(′) follows the trends in IR more than in DC, indicating that the occurrence of 6-methyl brGDGTs exerts a larger influence on this proxy compared to the degree of cyclisation, and the CBT(′), therefore, does not align with the independent precipitation indicator $\delta^2 H_{\text{max}}$ *(Fig. 1A). Therefore, we prefer to keep the focus of the manuscript on DC and IR as potential proxies for monsoon precipitation, and to make both brGDGT and isoGDGT data available to facilitate community efforts in improving our understanding of the key parameter(s) driving their relative abundances in loess.*

Fig. 1 Biomarker- and loess-based records for the past 130 kyr at Yuanbao. (A) Degree of cyclization (DC) of brGDGTs and ice-corrected $\mathscr{S}H_{\text{max}}$ *based on plant waxes in the same lipid extracts (Fuchs et al., 2023). (B) Cyclization of branched tetraethers (CBT, CBT′). (C) Isomer Ratio (IR). (D) Fractional abundance of crenarchaeol to total isoGDGTs (%Cren). (E) Branched and Isoprenoid Tetraether (BIT) index and ratio of iso- and brGDGTs (Ri/b). (F) Grain size (GS) and magnetic susceptibility (MagSus). (G) NHSI at 35°N (Berger et al.,* 2010) and the composite speleothem oxygen isotope ($\delta^{8}O$) record (Cheng et al., 2016). Dark *grey intervals (~23‒21 ka) in brGDGT-related records (DC, IR, CBT, and CBT′) indicate the transition from the outcrop to the pit and are not considered in the interpretation of the records.*

Minor comments:

Line 1: The method proposed here to quantify precipitation is explorative and needs to be verified at other sections. Remove "quantitative" from title to reflect the uncertainty surrounding this method. *Reply: We thank the reviewer for their suggestion. Note that we do test the DC at two other sections for which brGDGT data are available (see section 4.2 and Fig. 5 in our manuscript). Therefore, we feel that the DC can be considered as precipitation indicator in loess-paleosol*

sequences. Regardless, we are willing to change the title into: "Towards quantitative reconstruction of…." if the editor agrees.

Line 14: state here that both the speleothem and plant wax d2H records are already published.

Reply: We will make this clear in the revised manuscript.

Line 30-31: I am not an expert, but NH summer insolation also has an obliquity component, especially when we look at 65 oN and higher. In this manuscript the focus is on 35 oN insolation (e.g. figure 2), but this nuance of low versus high-latitude NH summer insolation

needs to be explained here and elsewhere in the manuscript. Also, the spectra of NH summer insolation (as shown in figure 2, so 35 oN) should be added to figure 3.

Reply: We thank the reviewer for pointing this out. It is true that the obliquity signal in Northern Hemisphere Summer Insolation (NHSI) becomes stronger at higher latitudes. We will clarify this in the revised manuscript when discussing the precession and obliquity signals in our proxy records. We will add the spectrum of NHSI at 35°N to Fig. 3 in the revised manuscript.

Line 32-34: this sentence seems to be crucial for the later interpretation of the data, but the reasoning behind this conclusion is not very clear for non-experts (like myself). The importance of this lag and why this argues against a NH insolation control needs to be explained a bit more here. This will help clarifying the discussion and conclusion later on.

Reply: We will extend this section in the revised version of the manuscript and clarify that not only the presence of cyclicity in a proxy record but also the phasing with respect to the orbital parameters points to the forcing mechanism.

Initially, Kutzbach (1981) has proposed that enhanced summer monsoon intensify is consistent with a stronger northern hemisphere summer insolation (NHSI) and would therefore vary in phase with precession cycles. Past variations in EAM climate have been inferred from proxy records such as the oxygen isotope composition ($\delta^{18}O$ *) of cave speleothems (Cheng et al., 2016; Wang et al., 2008, 2001) support this hypothesis (Kutzbach, 1981). However, other proxy records such as a stacked summer monsoon record from Arabian Sea shows a 6-8 kyr lag of monsoon maximum intensity with respect to the precession minima (NHSI maximum). This led to the suggestion that monsoon variations are driven by latent heat fluxes from southern hemisphere as well as global ice volume (Clemens*

and Prell, 2003). These discrepancies between proxy records opened the discussion on the interpretation of e.g. speleothem $\delta^{18}O$ *records (Clemens et al., 2010).*

Lines 46-48: Similarly, expand here to explain why a strong 23 kyr cycle is indicative for NHSI.

Reply: We will clarify this in the revised manuscript.

Line 68-onwards: somewhere in this section of the introduction of the manuscript explain where (and when) the biomarkers that are found in loess are produced. Do this for both the GDGTs and the plant waxes. For example, are the plant waxes produced in situ or transported with the loess? This nuance is important for the later discussion.

Reply: We thank the reviewer for raising this important point. In the loess-biomarker literature, both GDGTs and n-alkanes are commonly assumed to reflect an in situ signal. This is based on the absence of GDGTs in material from loess source regions, mainly due to the unfavorable growth conditions for GDGT producers in these arid deserts (Gao et al. 2012). In addition, GDGTs were below detection limit in wind-transported dust, suggesting that they are not commonly transported through the atmosphere (Hopmans et al., 2004).

Similarly, the plant wax signals can be interpreted as a local signal. Specifically for Yuanbao, the carbon isotope signal of the plant waxes indicates a consistent dominance of C³ vegetation, which aligns with the high elevation and cold, dry winters at Yuanbao that are unfavorable for C⁴ plants (Fuchs et al., 2023). Secondly, vegetation is sparse in the main dust source for the CLP, located northwest of Yuanbao. During summer, when the main wind direction is east-to-west, dust-associated transport of biomarkers is unlikely due to the higher *vegetation cover and increased precipitation towards the east, which largely prevents dust mobilization. These processes have also been discussed and confirmed by previous studies (Liu et al., 2005; Thomas et al., 2016; Zhou et al., 2016).*

Line 83: this is a bit of a NIOZ/UU centered list of papers. Lots of other groups have worked on this, I suggest diversifying the reference list here.

Reply: We thank the reviewer for their comments, we will add other works that highlight the influence of growing season temperature on brGDGT production to the revised manuscript.

Line 86-89: I was surprised that CBT was not discussed at all here (and not used at all in the entire manuscript). CBT is one of the most common methods to reconstruct soil pH. It needs to be introduced here. In this context, I wonder whether changes in the accumulation rates of brGDGTs hold any paleoclimatic information. The GDGTs were quantified using the C46 std, so this data is available.

Reply: We thank the reviewer for pointing this out. The CBT and CBT′ are definitely on our checklist, as always. However, as we mentioned in our response to your earlier comments, and also explain in the introduction of our manuscript, both CBT and CBT′ are combinations of the degree of cyclization (DC) and the isomer ratio (IR) (Fig. 1B). In arid and alkaline soils, however, it has been found that the DC and the IR exhibit opposite correlations with soil pH > 7 (Guo et al., 2022; and Fig. 4A and B in this manuscript). This finding is in accordance with the previously observed abrupt change in the relationship between CBT and soil pH at pH = 7.5 (Xie et al., 2012). Furthermore, the abundance of brGDGT compounds shows different optimal pH ranges (e.g., Supp Fig. 3 in De Jonge et al., 2021). Hence, the environmental parameter(s) that control the CBT in alkaline soils (like loess) deviates from

the global trend and is not well understood. As we also explain in the Introduction, we have, therefore, decided to focus on DC and IR. With this approach, we aim to improve our understanding of the key factors influencing all different aspects of changes in brGDGT distributions in loess.

As for the GDGT concentrations, these data are indeed available. As shown in Fig. 2A and B, GDGT concentrations follow the same trend as magnetic susceptibility (MagSus, Fig. 2D), indicating that they are similarly impacted by sedimentation rates (dilution) and/or the rate of soil formation (production) as MagSus. As such, this record does not provide additional paleoclimatic information beyond what MagSus already indicates. If the editor deems this useful, we can add this data to Fig. 2 in the revised manuscript.

Fig. 2 Biomarker- and loess-based records for the past 130 kyr at Yuanbao. (A) Concentration of isoprenoid GDGTs. (B) Concentration of brGDGTs. (C) Grain size (GS) and magnetic susceptibility (MagSus). (D) NHSI at 35°N (Berger et al., 2010) and the composite speleothem oxygen isotope ($\delta^{18}O$) record (Cheng et al., 2016).

Line 88: methylation can also occur at C7, see for example (Ding et al., 2016)

Reply: We will add this to the revised manuscript.

Line 90: we also discussed this in (Naafs et al., 2017)

Reply: We will include this in the revised manuscript.

Line 113: and is this benthic d18O record tuned to astronomical cycles like the LR04 stack is?

Reply: This benthic $\delta^{8}O$ *record used for our age model is LR04, we will specify this in the revised manuscript to avoid any confusion.*

Line 116: change to "...corresponding to a sedimentation..."

Reply: We will change this in the revised manuscript.

Line 144: Explain here why IIc and IIIb-IIIc are not used in the DC index. Is their abundance too low?

Reply: These compounds are indeed mostly below detection limit in the Yuanbao sequence. In addition, the equation for the DC presented in Baxter et al. (2019) does not include these compounds, see Eq. 2 in their paper. Regardless, including the (low contributions of) brGDGT-IIc(′), -IIIb(′) and -IIIc(′) in the DC does not affect the overall trend of the DC record for Yuanbao (Fig. 3), and the offset between DC with and without hexamethylated brGDGTs are mainly induced by brGDGT-IIIa. For the DC-MAP calibration, using the same

DC equation for both calibration and downcore records calculation will not impact the absolute precipitation reconstructions.

 $DC = (fIbI + 2*fIcI + fIIbI + fIIb'I)/(fIaI + fIbI + fIcI + fIIaI + fIIa'I + fIIbI + fIIb'I)$

DC (with hexamethylated brGDGTs) =

([Ib]+2[Ic]+[IIb]+[IIb′]+2*[IIc]+2*[IIc′]+[IIIb]+[IIIb′]+2*[IIIc]+2*[IIIc′])/([Ia]+[Ib] +[Ic]+[IIa]+[IIa′]+[IIb]+[IIb′]+[IIc]+[IIc′]+[IIIa]+[IIIa′]+[IIIb]+[IIb′]+[IIIc]+ IIIc′])*

Fig. 3 Degree of Cyclization (DC) over the past 130 kyr at Yuanbao using the original equation (orange curve, Baxter et al., 2019) and including the hexamethylated brGDGTs (purple curve). The dark grey interval $\left(\frac{23-21}{10}\right)$ *ka) indicates the transition from the outcrop to the pit and are not considered in the interpretation of the records.*

Line 153: what is this assumed standard deviation based on? Can repeat analysis of for example a lab standard provide a data-supported value? If not, what is the impact of selecting a slightly different value? How does this impact the MAP reconstructions?

Reply: The standard deviation of the DC in our lab is 0.02 based on an in-house standard run every ~10 samples. For this study, we chose 0.05 to ensure that potential between-lab

variations are better accounted for in the calibration. Nevertheless, using a standard deviation of 0.02 or 0.05 does not significantly impact the estimated MAP, as both fall within the calibration uncertainty (± 125 mm). Specifically, assuming a given DC of 0.5, a standard deviation of 0.02 yields a MAP of 784 mm, while a standard deviation of 0.05 yields a MAP of 804 mm.

Line 177-179: show cross plots of DC versus GS and d2H to provide a statistical basis for this "match"

Reply: The cross plots of DC vs GS and DC vs δ^2H_{max} *are shown in Fig. 4. We have decided not to add them into the manuscript because: i) It is evident in the time series that the low DC corresponds with more negative* $\delta^2 H_{\text{max}}$ *and high GS, and that the timing and direction (but not necessarily the amplitude) of changes in these records is similar. ii) The main focus in the discussion here is on the Henrich stadials (HS; i.e., millennial-scale events) during the last glacial period (Fig. 4C and D), rather than the entire record (Fig. 4A and B). As we have discussed in the manuscript (e.g., Line 285-293 in the original manuscript), the evapotranspiration and changes in moisture source impact the trend of* $\delta^2 H_{\text{max}}$ *which is not related to the precipitation amount recorded by DC. Regardless, the HS are clearly present* in GS, DC and $\delta^2 H_{\text{max}}$.

Fig. 4 Cross plot of (A) DC vs GS over the past 130 kyr and (C) the last glacial period; (B) DC vs $\delta^2 H_{\text{max}}$ *over the past 130 kyr and (D) the last glacial period. The color gradients indicate age of downcore samples.*

Line 179: also show cross plot for NH insolation and the IR record for comparison (and for other proxies used, see main comment)

Reply: We have decided not to include the cross plots of NHSI and GDGT-based proxies because DC and other GDGT-based proxies record sub-Milankovitch and millennial-scale variability. NHSI is influenced by orbital cycles and contains no sub-Milankovitch variability. For this reason, we included the bandpass filters of DC (Fig. 2B in the original manuscript) to directly compare with NHSI and highlight the signal of orbital forcings in our DC record.

Line 182: this grey is hard to see, add arrow to figure of where this splicing occurs *Reply: We will adjust this in the revised manuscript.*

Figure 3: How do you get 100 kyr cyclicity in a 130 kyr long record?

Reply: We agree that discussing glacial-interglacial cycles with a record that only extends back 130 kyr is challenging. Therefore, we consistently state that the interpretation of glacial-interglacial cycles in our record is restricted by the length of our record (e.g., Line 299 in the original manuscript). Nevertheless, the presence of a 100 kyr cycle in loess proxy records (GS and MagSus) is well described in earlier work based on longer time series (e.g., Sun et al., 2022).

Line 195: but besides precession, the DC record also shows a strong 41 kyr signal *Reply: Indeed, both precession and obliquity signals are evident in the DC record (Fig. 3A in the original manuscript). However, in this part of the manuscript, our primary focus is the*

comparison between DC and δ^2H_{max} *, we have therefore mainly discussed their shared characteristics, particularly those in the precession band. The discussion of obliquity cycles occurs later in the manuscript, i.e., Line 295 in the original manuscript.*

Line 199: to fully determine whether the DC and IR records are different, show a spectral analysis of the IR record to highlight that it lack precession and obliquity cycles as seen in the DC record.

Reply: The interpretation of IR in Yuanbao section is not clear (yet), therefore we have not included any timeseries analysis of this record in the previous version. Here we show spectral analysis of IR (Fig. 5). Although the IR shows a precession signal based on the spectral analysis, its spectral density is comparatively smaller than that of the other proxies (Fig. 5F). If the editor deems this useful, we can add a spectral panel to Fig. 3 in the revised manuscript.

Fig. 5 Spectra of time series of proxy records from the Yuanbao loess section, cave speleothems in southeast China, and Northern Hemisphere Summer Insolation (NHSI) at 35°N.

Line 207: could the brGDGTs be used to quantify these variations in hot and cold conditions? *Reply: Unfortunately, no. The MBT′5ME index shows an abrupt and large increase in parallel with the change in IR. Next to the fact that this would suggest that conditions changed from cold to warm, which is opposite to what is suggested by the loess proxies and isotopic signals based on speleothem* $\delta^{18}O$ *and* $\delta^{2}H_{\text{max}}$ *, the MBT'_{5ME} also shows non-analogue behavior in part of the core. This introduces uncertainty in the brGDGT-temperature relationship. The MBT′5ME record for this section and an extensive assessment of its environmental controls is part of a manuscript that is currently under review with Organic Geochemistry.*

Line 211: doesn't Ca2+ affect pH and that influences brGDGT production? Is there clear evidence that it is Ca2+ and not the resulting change in pH?

Reply: We agree with the reviewer that distinguishing the impacts of Ca2+ and soil pH on brGDGT distributions is challenging. Nevertheless, loess is generally rich in carbonate, which will dissolve after rainfall. The then released Ca2+ could possibly contribute to an increase in soil pH. However, this mechanism points at the amount of available Ca2+ as the primary driver of the production of cyclic brGDGTs in loess. In addition, available Ca2+ was found to be a more important factor explaining the relative abundance of cyclic brGDGTs in an Arctic elevation transect (Halffman et al., 2022) as well as a suite of mid-latitude soils (De Jonge et al., 2021) compared to soil pH or free acidity.

Line 215: I don't understand how Ca2+ affects brGDGT production. The direct impact of Ca2+ on brGDGT producers needs explanation. If Ca2+ drives pH and that impacts brGDGT producers, explain that here

Reply: Unfortunately, the exact link between available Ca2+ and the production of cyclic brGDGTs is currently based on empirical correlations in soils from Scandinavia (Halffman et al., 2022) as well on more global soil datasets (De Jonge et al., 2021, 2024). However, microbial ecological studies have shown that Ca rather than pH is a key predictor in shaping the soil microbiome as well as its functionality (e.g., Shepherd and Oliverio, 2024; Neal and Glendining, 2019; Allison et al., 2007). We will add the information on the influence of Ca on the microbial diversity in soils to the revised manuscript.

Line 222: doesn't a r2 of 0.06 indicate no correlation?

Reply: We will rephrase this in the revised manuscript.

Line 225: although the overall community might change, this doesn't mean that the brGDGT producing community changes. The next sentence should state that this is speculation.

Reply: We will clarify this in the revised manuscript.

Figure 4: do I understand correction from this figure that pH is not correlated to MAP because IR is strongly correlated with pH, but not with MAP? Does that not undermine some of the earlier text of this manuscript where MAP and pH are suggested to be linked? *Reply: As the figure below shows, MAP and soil pH stills show a negative correlation globally (Fig. 6). In general, IR also relates to MAP if we do not separate the data into different groups based on soil pH.*

Fig. 6 Cross plots of observed mean annual precipitation (MAP) vs soil pH and isomer ratio (IR). The pink, blue, and yellow symbols indicate soils with pH < 7 and pH > 7 in the global soil dataset (Raberg et al., 2022), and modern soils from the Chinese Loess Plateau (Wang et al., 2020), respectively.

Line 243: cite reference for modern soil CLP GDGT data here

Reply: We will include the reference in the revised manuscript.

Line 245: how does this uncertainty of ± 125 mm compare to other quantitative proxies used for the CLP? Is this correlation much better, worse, or similar to other methods? This context would be good for the non-expert.

Reply: The uncertainty of quantitative precipitation reconstructed by ¹⁰Be is 190 mm (1) standard deviation) (Beck et al., 2018), we will specify these in the revised manuscript. The uncertainty of microcodium Sr/Ca ratio is not reported (Li et al., 2017).

Line 266: and how does this gradient compare to the modern gradient?

Reply: The modern MAP at Yuanbao, Xifeng and Weinan is 500 mm, 470 mm and 600 mm, respectively. The downcore records reflect a similar spatial gradient as seen in the modern observations. We will clarify this in the revised manuscript.

Line 268: is the difference in reconstructed MAP between Holocene optimum and MIS 5e statistically not different? State statistically proof for this statement.

Reply: In this sentence we aimed at making the point that reconstructed MAP was spatially similar during interglacials, i.e., between sites, but not between the Holocene and MIS5, as our data indeed suggest that MAP was higher during MIS5 than during the Holocene. We will rephrase this sentence for clarification.

Line 283: the manuscript states a "close resemblance", but the DC has a strong 41 kyr signal that is lacking in the d2H record.

Reply: We thank the reviewer for pointing this out, we will rephrase this in the revised manuscript to ensure that the description is more precise.

Lines 283-308: For this comparison with the d2H record, it is important to in the introduction explain where and when the different biomarkers are produced. Is there a possibility for a spatial and/or temporal offset between production of the plant waxes and bacterial membrane lipids?

Reply: As explained in our response to your earlier comments, both the brGDGT-based proxies and ²Hwax at Yuanbao reflect in-situ signals and this should not have spatial or temporal offsets. We will clarify this in the Introduction in revised manuscript.

Figure 6: why is the IR data not included here? Does that not have a clear precession forcing? *Reply: The main reason we decided not to include IR in the phase wheel is that it remains unclear which climate aspect IR indicates in this downcore record, while the phase wheel is intended to show the leads and lags of precipitation-related proxies on the precession band. In addition, as the reviewer has pointed out, although the IR shows a precession signal based on the spectral analysis, its spectral density is comparatively smaller than that of the other proxies (Fig. 5).*

Line 343: ensure that the individual GDGT data is available for future usage

Reply: We thank the reviewer for their reminder. The related dataset has been submitted to PANGAEA database. We will add the DOI link as soon as it becomes available. In addition, we will ensure that the dataset is attached as a supplement file upon acceptance.