Response to Editor and reviewer comments

Dear Editor,

We thank you and the reviewers for your suggestions.

In our revision, through a third case study, we demonstrate how incorporating the distribution of the apparent fractionation factor (ε_a) between plant wax and precipitation in three plant groups can be used to reconstruct precipitation δ^2H. We present the case study with a three-end mixing scenario using published chain length distribution, $\delta^{13}C$, and δ^2H of four n-alkanes from a marine core off the Zambezi River Mouth. The model priors consist of user-defined endmembers and their associated parametric distributions of 1) *n*-alkane concentrations, 2) $\delta^{13}C$, and 3) ε_a.

By leveraging $\delta^{13}C$ and relative abundance from multiple chains, the model produces mixing ratios of rainforest C_3, savanna C_3, and C_4 end members, which are similar to a vegetation correction on n-alkane δ^2H in conventional methods. Moreover, the model produces estimated precipitation δ^2H with relatively small uncertainty limits using the end member mixing ratios, ε_a distributions among the end members, and the measured δ^2H of the four chains. We also provide a comparison between the reconstructed precipitation δ^2H and the δ^2H from two dominant alkanes, and interpretations of the reconstructed precipitation δ^2H curve with published sedimentary sourcing patterns in the region. We also provide a qualitative comparison between our vegetation reconstruction and the pollen record of a nearby marine core in the supplementary document, as suggested by reviewer 1.

We think that case study 3 demonstrates model utility in paleoclimate reconstructions, which makes the article more aligned with the aim and scope of the journal. Overall, we think that the three case studies highlight the flexibility and strengths of the Bayesian framework in disentangling multivariate mixing scenarios in plant wax lipids. We hope that our revision provides substantial improvements to the manuscript and addresses most of the issues raised by the reviewers.

We also provide specific responses to the minor issues raised by reviewer 2 in the sections below.

Please let us know if there are any other issues or concerns.

Thank you very much for your assistance!

Deming Yang and Gabe Bowen

Authors' response to specific comments by Reviewer 2:

l27: I would say mid-chains are of likewise importance (just consider the many studies using Paq), i.e. I´d rather span the range to "C23 – C35".

> Edits were made as suggested.

l59 and other: general comment: I think references should be sorted by year, i.e. here start with Collister and end with Liu and An. (relevant for whole text)

Figure 1: even they are explained elsewhere, abbreviations (FLMC, etc) should be explained here in the Figure caption

> Abbreviations made as suggested

Table 1: so high Paq means high influence of aquatic sources onto C27 (visible at somewhat higher d13C in this sample). Even though the focus is on C27, C29, C31, would it make sense to report data also for C23, C25?

> The data included in case study 1 and Table 1 are originally from Liu et al., 2015, which do not include C_{23} and C_{25} alkanes. This is also the reason why we could only use C_{27}, C_{29}, C_{31} alkanes in the case study.

l117: "we expect n-alkane δ 13C to follow a group-specific distribution pattern": I´m wondering how well this works for aquatic plants, because those have shown to be quite variable in their d13C values, even within similar species. This is also visible on the right panels in Figure 2 (pretty broad Gaussian blue curve overlay).

> Although the uncertainty in the prior distribution of $\delta^{13}C$ for aquatic macrophytes is relatively high, it is worth mentioning that such uncertainty is already included in the model and properly represented in the results. The two benefits of our Bayesian approach are 1) simultaneous evaluation of data from multiple n-alkane chains, and 2) straightforward propagation of uncertainty in the model results. Analyzing multiple chains at the same time effectively reduces the overall uncertainty in the model results by leveraging common sensitivities among the chains to the same variables (mixing ratios and concentrations). By contrast, If the conventional linear mixing method is used, the overall uncertainty of the results would be much higher due to the high uncertainty of the selected chain. Therefore, the width of the Gaussian curve may not be the main concern. Instead, the fit of the curve might be, especially if the empirical data deviate from the assumed unimodal distribution. For example, if a bimodal distribution of $\delta^{13}C$ is observed, it is better to consider splitting the assumed group into two.

l245: micro or macroalgae? Macroalgae (e.g. Charophytes) further complicate the issue because they mainly produce mid-chains. Or are Chara sp. here included to

macrophytes (because they are listed in the table EA-2 at github)? This needs clarification.

> The reviewer asked about the treatment of Characeae (genus *Chara*) in our data sheet shared on GitHub. The genus *Chara* is treated as algae while we did not specify whether it is micro or macro algae. This treatment is following Aichner et al., 2010 and Liu & Liu, 2016. When we were putting the data together, we observed that the genus *Chara* display much lower $\delta^{13}C$ values than macrophytes as reported by Aichner et al., 2010. This further justifies the reason why *Chara* is not grouped with macrophytes, which display higher and more variable $\delta^{13}C$ values according to Aichner et al., 2010.

Table 3: is "MAP" the official abbreviation for Maximum A Posteriori probability estimates, or could an alternative shortening be used? Just because it´s easily confused with mean annual precipitation.

> This has been changed to MAPE to avoid confusion.

l313 and 313: what precisely is a "trade-off correlation"?

> A short explanation is added in parentheses.

Supplement:

l12 and ff: it is unclear what EA-2, 3, 4 are referring to. Those tables appear in the github data sources. If this is the case, those should be referenced here in the supplement.

> EA-2, 3, 4, etc. are now referenced as "Supplementary Data EA-x, Yang 2022", where "Yang, 2022" refers to the code and data repository on Zenodo for this project.

l16: two time "in"

> This has been corrected.

Supplement tables: some "n" in n-alkanes are not italic

> This has been corrected.

List of all relevant changes made in the manuscript

1. The title has been changed to reflect the inclusion of case study 3.
2. The abstract has been changed to reflect the inclusion of case study 3.
3. Some minor details have been changed throughout the manuscript to reflect the inclusion of case study 3.
4. Section 2.2.3 has been added for the specific methods in case study 3.
5. Section 3.3 has been added for the results of case study 3.
6. Section 4.1.3 has been added for the discussions of case study 3.
7. Both the discussion and conclusion sections have been modified to reflect the inclusion of case study 3.
8. The in-text citation and the reference list have been updated.
9. In the supplementary document, Section 4 has been added for the qualitative comparison to a pollen record related to case study 3
10. An updated version of the code, prior data and visualization is now available on Zenodo.

Archived responses during the open discussion phase

Response to Reviewer 1

We thank the reviewer for taking the time to review our manuscript, and the constructive suggestions. The reviewer suggested that we should elaborate on three aspects of the manuscript. Below is our response to the reviewer's suggestions point by point.

1) To paper would benefit from a more thorough comparison to existing techniques (e.g., linear mixing-model approaches of Gao et al., 2011). You state that your results appear to provide alternative interpretations to the same n-alkane records – please elaborate!

In our case studies (CS1 and CS2), we did provide alternative interpretations to the published data. In CS1, we compared the conventional interpretation of aquatic plant input based on the P_{aq} index and our interpretation based on $\delta^{13}C$ and chain length distribution of three n-alkane chains. In CS2, we compared the interpretation of vegetation composition based on $\delta^{13}C$ of one n-alkane chain, and our interpretation based on $\delta^{13}C$ and chain length distribution of three n-alkane chains. Both case studies provided comparisons with existing interpretation techniques and we highlight where the new method's results are similar to those methods as well as where they provide alternative or more nuanced interpretations.

We can not provide a direct comparison with the approach by Gao et al. (2011), specifically, based on the case studies we currently include, because these datasets do not contain all the information required by Gao's method. As discussed in the next response we do plan to add additional comparisons to established methods for vegetation reconstruction by adding an additional case study.

2) It is essential to validate this approach in a sediment core with independent vegetation reconstructions. I would take a look at the African records published by Sarah Feakins (e.g., Feakins 2013 P3) - these include n-alkane chain length distributions, n-alkane carbon isotopes and the % of shrub, graminoids and tree pollen. This seems an ideal site to test your approach. However, I am sure there are dozens of other suitable sites.

We agree that the approach would be much more convincing if the results are consistent with an independent vegetation reconstruction, e.g., by pollen analysis. We have done a literature search and identified data that will allow us to complete such a comparison. We are planning to include such a comparison in the supplementary document. We would like to note that the comparison won't be a true validation,

because the two approaches are associated with different potential biases such as pollen/n-alkane production and transportation. For this reason, some differences between the approaches are to be expected.

3) The authors state that their approach could be used to assess the interpretation of associated proxies such as n-alkane δ2H. This would be a great tool for organic geochemists and paleoclimatologists. However, the authors did not explore this any further. The authors should demonstrate - if they can, this paper will be far more valuable to the paleoclimate community.

We agree with the reviewer that adding information on how the approach can help to interpret associated n-alkane δ^2H will add tremendous value to the significance of the framework. Doing so would make the manuscript more aligned with the aims and scope of the journal. We are planning to add one more case study to demonstrate how our framework can be implemented. The case study will be based on n-alkane records in a marine core off the Zambezi River Mouth (Wang et al., 2013, GCA), which has δ^2H, $\delta^{13}C$, and chain length distribution data on n-C_{27}, n-C_{29}, n-C_{31}, and n-C_{33} chains. Please note that doing so will require further development of the existing model structure, and discussion of model outputs. Please allow us some time to implement the case study in the revision.

Response to Reviewer 2, AC1

We appreciate the reviewer's recognition of the novelty of the model approach, and the recommendation for publication. We also thank the reviewer for the detailed comments and constructive suggestions.

The main issue raised by the reviewer is whether the manuscript is a good fit for Climate of the Past. The reviewer also mentioned that a case study analyzing a paleorecord will help to address the issue. We would like to note that reviewer 1 raised a similar issue.

In our response to reviewer 1, we agreed to include an additional case study to demonstrate the model's utility in analyzing a paleorecord, with n-alkane δ^2H as an additional model component. We agree that adding this case study will make the manuscript of interest to both organic geochemists and paleoclimatologists and make it a much better fit for the journal. We are finalizing the case study right now.

The specific comments by reviewer 2 seem to have raised some minor issues. We appreciate the reviewer thoroughness in the review, and we will do our best to address them in the revision.

Response to Reviewer 2, AC2

We thank the reviewer for the follow-up questions, and we apologize for any negligence of the issues raised. We are delighted to see the topics brought up by the reviewer as we think that such a discussion is needed beyond the manuscript itself.

The reviewer asked about the treatment of Characeae (genus *Chara*) in our data sheet shared on GitHub. The genus *Chara* is treated as algae while we did not specify whether it is micro or macro algae. This treatment is following Aichner et al., 2010 and Liu & Liu, 2016. When we were putting the data together, we observed that the genus *Chara* display much lower $\delta^{13}C$ values than macrophytes as reported by Aichner et al., 2010. This further justifies the reason why *Chara* is not grouped with macrophytes, which display higher and more variable $\delta^{13}C$ values according to Aichner et al., 2010.

The reviewer also asked about how to make this approach more accessible to a broad range of researchers, as the approach may seem rather complicated. We would like to add to this question by emphasizing the main benefits of using such a Bayesian approach, which are 1) getting the most out of compound-specific isotope analysis by offering mixing solutions of multiple sources, and 2) straightforward handling of uncertainty in the analysis. It is intuitive that researchers would only choose to use this approach if the benefits outweigh the "costs". We would like to start with the notion that there are potentially huge benefits in using this approach, some of which are outlined in section 4.2. Case study 3 will specifically demonstrate the benefits of handling uncertainty and add to what is already in section 4.2. We really hope that this publication can increase the awareness on the benefits that this approach may bring to the community.

In terms of lowering the hurdle (cost), there are several things that can be done. One is to develop more user-friendly versions of the code into perhaps an R package and distribute it with a detailed tutorial. We are planning to do this, but we would also like to receive some community feedback first through this publication. Another is to organize workshops online or at conferences for the technical details of this approach if there is enough interest. Again, community feedback is key. We are also open to inquiries and potential collaborations if necessary.

The reviewer also asked about the handling of prior distributions with examples of whether to use regional compilations or global averages. The model itself is flexible enough to accommodate either choice, so it ultimately depends on the user and the reasons to justify it. We chose to use regional compilations in case studies 1 and 2 because we think that this is more consistent with what we know about the production, transport and mixing processes of plant wax lipids in lake sediments. If using global

averages can be justified in certain situations, we see no reason why they cannot be used. Related to this and the reviewer's last comment, we think that having a centralized lipid database is important because it can make this model approach much easier to use from a user's perspective, whether the need is for a regional compilation as demonstrated, or a global one.