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Interactive comment on "A dual-biomarker approach for quantification of changes in relative humidity from sedimentary lipid D / H ratios" *by* Oliver Rach et al.

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We would like to thank the referees for their valuable comments. In the following we provide a point by point answer to the specific comments of referee #3. The original comments of referee #3 you will find below. Our answers are in italic letters below each comment.

Anonymous Referee #3

General: Rach et al. use the hydrogen isotopic difference between mid-chain nC23 alkanes and long-chain nC29 alkanes, which they interpret to be mainly derived from aquatic and terrestrial plants, respectively, to infer changes in relative humidity based

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on a so-called DUB (dual biomarker) model. While I agree that a step forward towards quantitative estimates of changes in terrestrial hydrology based on lipid biomarker hydrogen isotope compositions is needed, I think that the authors underestimate the uncertainties in their approach and underlying assumptions so that the calculated estimates in changes of relative humidity cannot be regarded as precise or even accurate. I agree that the approach should be presented but only with a broader discussion of potential sources of uncertainty. My main comments are on the assumptions which go into the consideration and the model. Some of them are shortly discussed in the manuscript while others are only 'between the lines'. I think this should be discussed more broadly and openly and would then add to the strength of the paper.

Lipid distributions in plants: The authors assume that the nC23 reflects a signal from the aquatic macrophytes while the nC29 reflects a signal of the integrated terrestrial plant ecosystem. n-Alkane distributions are, however, not so distinctive in plants. Terrestrial plants also make nC23 and macrophytes also make nC29 albeit in smaller amounts. Due to the current lack of isotope data of the smaller abundant compounds it cannot be assumed that the nC23 has the same hydrogen isotope composition as the nC29 in terrestrial plants and macrophytes, respectively. nC29 and nC31 as most abundant alkanes in terrestrial plants often show slightly different hydrogen isotope compositions in the same plant so this would also be expected for nC23 and nC29. In sedimentary mixtures of various alkane sources this is difficult to disentangle. Even if a sediment sample would only contain alkanes from a single plant species such a difference would be interpreted by the model to reflect a difference in evaporative enrichment in leaf waters which would clearly not be the case.

Answer from authors: We are well aware of the issues the reviewer touches here. In that sense, MFM as a well studied site does allow us to constrain lipid sources to a degree which is likely more difficult to obtain at other sites. We are prepared to outline our general aims with the model, the problems with a universal applicability (see answer to Rev. #1) as well as the assumptions and model parametrizations in more

detail in a revised version (i.e. a table with the assumptions etc., see answers to Rev. #1). With regard to source assignments of biomarkers: we did discuss lipid sources and how they can be constrained at MFM in detail in the supplementary material of (Rach et al., 2014), and believe that we should not repeat the whole discussion here. In brief, the detailed pollen record allows for biomarker source assessment. There we also show the tight covariation between nC29 and nC31 alkane δ2H values, which we regard as being derived from a similar terrestrial source. These two compounds have similar absolute δ2H values, whereas nC23 δ2H values are more enriched, suggesting a different source (i.e. macrophyte).

Ecosystem integration: Sediments will collect alkanes from a variety of sources including ones that are derived from distant sources. As alkanes from different plants can have very different hydrogen isotope values depending on used water sources, different biosynthetic fractionation and variable sensitivity to leaf water enrichment any changes in the relative proportions of the supplied alkanes to the sediment, either by changes in the ecosystem composition around the lake or changes in local versus distant sources of alkanes can lead to changes in the recorded signals which have nothing to do with changes in relative humidity at the site. Ecosystem changes might occur due to changing temperature and CO2 levels next to relative humidity. Source water isotopes can change due to shifts in moisture sources and transport pathways. Changes in aeolian-derived alkanes might occur due to changing wind patterns and strengths. These factors would introduce uncertainty in relative humidity estimates.

Answer from authors: We agree with the reviewer on the potential relevance of these processes, but we can either rule them out or we account for them: We do discuss ecosystem changes (which are reconstructed from pollen data) as potential factors influencing our model outcome (see also the supplementary material of Rach et al. 2014) and in the present manuscript we present an approach to quantify the influence on the model outcome (vegetation correction). During the YD no major changes in atmospheric CO2 occurred, as such we can rule this process out. Source water δ2H values

can (and likely did) change due to moisture source changes during the onset and termination of the YD, but our approach, i.e. using terrestrial and aquatic biomarkers, which use the same source water, is therefore not dependent on this factor. In addition, the lake's small catchment area (5,76 km2) with steep, vegetated crater walls, sheltering the lake from wind makes a dominant input from more distant sources unlikely vs. the importance of local sources.

Sediment integration: Sediments represent not only spatial but also time-integrated signals. The authors apply their model not to plants but to lipids from sediments which integrate over several years with inter- and intra-annual variability. The investigated sediment samples in Rach et al. (2014) are 1 cm thick. With a sedimentation rate of 0.5 to 3 mm per year in Meerfelder Maar these samples reflect a few to about 20 years at least. The signals recorded by the aquatic and terrestrial lipids could vary from year to year as well as their relative contributions into the sediments which would then lead to signals that are not directly comparable between aquatic and terrigenous lipids regarding the recorded environmental conditions. The signals of both aquatic and terrestrial lipids alone would reflect averaged conditions over the sample integration interval but it seems questionable to me if these are then directly comparable. Although difficult to predict the effect of time-integration might add additional uncertainty to the model results.

Answer from authors: Again, we agree with the reviewer that these processes can be important. But at MFM, we can rule out several of these effects due to the small catchment size and steep morphology: the residence time of n-alkanes in this small and steep catchment (i.e. from leaf or soil) is likely short, we assume within our sampling resolution (i.e. decades). In this catchment we assume that potential differences in the temporal integration between aquatic and terrestrial biomarkers are small, i.e. smaller than our sampling resolution (decadal on average), which is supported by the similar sample to sample (i.e. decadal) variability in their lipid δ2H values. If, for example, terrestrial leaf wax n-alkanes would have a substantially longer residence time in the

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soils before being transported into the lake, then the decadal variability should be much smaller, as the soil would already deliver a more integrated signal into the lake. We will outline this argument in a revised version of the manuscript.

Dependence on setting: The authors assume that isotopic enrichment due to lake water evaporation and surface soil water evaporation does not occur. While this may be true for the Meerfelder Maar site it certainly is not true on a larger scale. Surface soil water enrichment occurs in semi-arid to arid areas and shallow-rooting plants incorporate this signal. Lake surface water isotope enrichment occurs in arid areas and then offsets the recorded aquatic signals. Lakes may also be fed by groundwater and can thus be isotopically offset from precipitation. Also the assumption that the isotopic enrichment in terrigenous lipids is due to leaf water enrichment may be questionable on a larger scale. In settings with very short rainy and growing seasons the vegetation might not be sensitive for leaf water enrichment as assumed here. These are clear restrictions of the model to humid regions with rain-fed lakes and should be made clear in the discussion. It can thus not be assumed that the isotopic offset between aquatic and terrestrial solely arises from leaf water enrichment, which in my view is an oversimplification.

Answer from authors: We agree with the Reviewer on the relevance of these issues, as these were part of the reason why we applied to model to the MFM record. In a revised version of the manuscript we will expand the discussion on the problems of a universal applicability of the model under different environmental and hydroclimatic boundary conditons. See also answers to Rev. #1.

In summary, I think the approach to apply a plant physiological model to sedimentary lipid isotope composition is interesting as an exercise to test if the outcome makes sense but highly challenging as sedimentary lipids cannot be treated in a similar fashion as lipids directly derived from plants. The environmental factors regarding variable lipid sources, spatial and temporal integration of signals, and the dependence on the particular setting need to be taken into account and discussed openly. Although the

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environmental processes which lay between plants and sediments tend to be often ignored in literature it cannot simply be assumed that plant lipids and sedimentary lipids can be treated similarly. An adequate discussion of these environmental processes and associated uncertainties needs to be included. Although likely impossible to quantify, I expect the associated uncertainties to be much larger than the 3.4% in rH based on the model alone probably exceeding the total amplitude of the reconstructed changes in rH. The model results should be discussed in the context of the environmental processes to avoid the risk of an over-interpretation of the model output. In this respect, I wonder if the data derived from the model actually indeed provide more quantitative information than the comparison of the two 'raw' isotopic signals alone as shown in Rach et al. (2014)

Answer from authors: As mentioned in the 2nd comment we are aware of these limitations and choose the MFM record for exactly those reasons, as we can rule out a number of these processes. We again state, that we do not believe the DUB model is universally applicable, but only at locations, where these boundary conditions are met (we plan to provide a checklist of boundary conditions for applicability in the revised version). The calculated uncertainty of 3.4% includes all errors which can be quantified in some way, but likely not the full uncertainty of the model due to the incomplete understanding of some processes. That is why we rather see the range of results from the 3 approaches (uncorrected and corrected) as the current uncertainty of model results (we state this in line 563): "Tentatively, the lower variability in Δrh** within the YD as well as the less pronounced shift in particular at the onset and termination of the YD (Fig. 3A) provides are more realistic scenario. But as of now, we regard the differences in predictions as the error of quantitative predictions from the DUB approach".

Rach, O., Brauer, A., Wilkes, H., Sachse, D., 2014. Delayed hydrological response to Greenland cooling at the onset of the Younger Dryas in western Europe. Nature Geoscience 7, 109-112.

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