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Climate of the Past Discussions

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.

Anonymous Referee #3

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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 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.

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.

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.

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.

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 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).

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