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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.

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We would like to thank the referees for their valuable comments. We are prepared to address and/or clarify all points raised by the reviewers in a revised version of our manuscript. Both reviewer 1 and 3 addressed the problems associated with a general applicability of our approach due to the number of assumptions and parametrizations which we feed into the model. We completely agree with the reviewers on this point and stress that it is not our intention to introduce an approach which is universally applicable. Rather, we see our manuscript as a proof-of-concept, i.e. an idea what is potentially possible, when certain conditions are met and constraints can be made to parametrize the model. We show what we would need to get quantitative information from biomarker hydrogen isotope data. For this reason, as an example we choose an exceptionally well studied and detailed archive (Lake Meerfelder Maar), where we can constrain the model parameters as accurately as currently possible, and show that we obtain reasonable data, albeit with significant uncertainties. We are prepared to discuss this aspect in a revised version in more detail, so that the manuscript is not mistaken as a blueprint which can be applied to any lacustrine archive. We do understand our contribution as a first step towards quantitative reconstructions from biomarker data.

In the following we provide a point by point answer to the specific comments of referee #1. The original comments of referee #1 you will find below. Our answers are in italic letters below each comment.

Anonymous Referee #1

Rach et al. present a mechanistic leaf-water isotope model to quantify for the first time past changes in relative humidity using lake sedimentary leaf-wax dD records. The model, which is suited for reconstructions from temperate regions where lake water evaporation is negligible, is tested using previously published proxy data from the type site of Meerfelder Maar (MFM) in Western Germany, with a focus on the period spanning the climate transitions into and out of the Younger Dryas stadial. Personally, I'm glad to see this work finally coming along after several years of development. I think this study constitutes a long overdue leap towards a more quantitative estimate of lipid-based stable water isotope reconstructions, which will ultimately be of interest to a broad readership including organic geochemists, paleobotanists, paleoclimatologists and climate modellers.

The construction of the model leans upon a large number of unavoidable assumptions involving both plant physiology and environmental conditions. While the majority of the physiological assumptions are reasonably satisfied through empirical evidence, I think some of the assumptions surrounding past environmental and climate conditions are less convincing. Nonetheless, due to the lack of sound proxies that allows reconstructing parameters such as local vegetation cover, atmospheric pressure, wind strength and seasonal air temperature (among others), the model formulation presented in this paper should be considered –overall– as good as it can get. However, I think there is still much room for improvement and I have a number of serious concerns, primarily involving the methodological approach, the data-uncertainty treatment, and the contents of the paper that the authors should address before publication.

Main comments 1) I am very surprised the authors decided to design this study directly around downcore data without attempting validation of their model using core-top samples from MFM. I think the model should first be tested using lipid δD measurements from surface sediments in tandem with meteorological observations before a deglacial Rh reconstruction is discussed. Whether this unusual direction was taken intentionally or not, I think the authors should provide some explanations.

Answer from authors: We do understand the reviewer's concerns, but we deliberately choose this approach. In a general sense, there are two possibilities of validating climate proxy approaches. The most straight forward is to test the proxy under modern conditions and compare results with actual instrumental data. This can be done either along a modern climatological gradient or over the time period where instrumental data are available. Potential problems of this approach are a limited comparability of environmental changes in space and time. The second approach is to use a past time period with known major changes in the parameter to be tested for. We employ the second approach here, because the first approach is unsuitable for the DUB model. This is for the following reasons: 1) Testing the DUB approach with modern core top sediments from MFM (or any other lake) is not feasible, as over the instrumental period (roughly the last 150 years) no major changes in relative humidity occurred. Lake MFM is also not annually laminated (varved) during this period (due to human interference), so that MFM would not be suitable. We don't know of any other lake sediment record which is varved and was subject to major rH changes during the last 150 years. Using only core top sediments (i.e. only one data point integrating the last decade or so) would not allow for testing the performance of the DUB approach, which aims to reconstruct relative changes in rH, not absolute data (see discussion in the manuscript). 2) We considered testing the DUB approach along a climatic gradient i.e. (Sachse et al., 2004). We concluded this approach to be unsuitable, because we cannot assume that the source of aquatic biomarkers (in our case nC23) is always the same aquatic macrophyte in different lakes and ecosystems. For that matter, it was impossible to find enough lake systems where the sources of aquatic biomarkers are comparable and which cover a large enough aridity gradient. As such, we rejected the modern calibration approach and opted to use a past sediment record with the following constraints: a) Which covers a time period where major changes in relative humidity have been identified before (the YD as the last major abrupt climate change in Europe, see (Brauer et al., 2008; Isarin et al., 1998; Rach et al., 2014) and b) Where sources of in particular aquatic (but also terrestrial) biomarkers can be constrained. In lake MFM detailed pollen records (Brauer et al., 1999) allow this source assignment, as outlined in (Rach et al., 2014), see also the supplementary material of this paper. In brief, during the studied core section of MFM for the aquatic biomarker we observed a covariation of nC23 concentrations and the abundance of Potamogeton remains, so that we concluded nC23 during the study period at MFM is mainly produced by this aquatic macrophyte (Rach et al., 2014). We are aware of the limitations of our approach, in particular it's potential applicability to other records, where such constraints are more uncertain. Our intent is to show that the DUB model is a potential approach to reconstruct quantitative hydrological information, if the sediment record, in particular biomarker source assessments, are well constrainable. In that sense, the well studied MFM YD record is a perfect record to put our hypothesis forward. Our approach is supported by the tight correlation between our ΔrH reconstructions and pollen concentrations of aridity adapted land plants (i.e. Artemisia), see Fig. 4, i.e. two completely independent aridity proxies. In a revised version of the manuscript we would explain this reasoning in more detail and outline the requirements for the applicability in other records. We also outline other potential approaches, i.e. choosing more source specific aquatic biomarkers (which however are not present in all lake records or not over the whole of the period of interest).

2) One of the main assumptions upon which the results depend is that pollen records are indicative of the local vegetation cover and that pollen counts scale linearly with waxes concentration in MFM sediments. If this were the case, one would expect to observe a clear correlation between the abundance of n-alkanes and pollen records. I would therefore like to see the distribution of n-alkanes plotted together with selected pollen data (e.g. relative distribution of trees, shrubs, herbs). Ideally, the authors should also present some correlation statistics or at the very least a visual correlation using for instance the average n-alkane chain length (and other chain length ratios) to make a qualitative distinction between graminoids and woody plants that would confirm the authors' hypothesis.

Answer from authors: We disagree with the reviewer. The results of our general approach do not at all depend on the assumption that pollen records are indicative of local/regional vegetation. Only for the vegetation correction approaches we rely on this assumption, as we outline also in the paper. We note again, that the vegetation correction does not substantially alters our results, see also below. We do think that at MFM pollen concentrations do record local to regional vegetation, due to the small catchment size of the maar lake etc. (see discussion in (Brauer et al., 1999). However, results from the DUB model with and without (which doesn't rely on the pollen data at all) the vegetation correction shows very similar results. We discuss leaf wax biomarker sources in the manuscript and in more detail in Rach et al. (2014), see supplementary material, where we also show pollen and leaf wax concentrations. As such, we don't think it is necessary to provide this here again. Correlations between the amount of biomarkers and the amount of pollen would not support the assumption, as different plant types produce different amounts of n-alkanes. As such, an increase in grass pollen may be accompanied by a decrease or an increase in n-alkane concentrations, if grasses produce less or more n-alkanes respectively, than the previously dominant species.

3) The authors argue that monocots integrate a varying evaporative δD enrichment signal in response to changes in humidity conditions. Following this reasoning, the mixing ratio between leaf water and un-enriched xylem water in C3 grasses will change as a function of the aridity level in the catchment. Therefore, the weighing of the fraction of grass cover used to correct εterr-aq should also vary over time, whereby a much higher fraction of C3 grass integrated the leaf-water evaporative δD enrichment during the Late Allerød (AL) and Early Holocene, when climatic conditions were presumably wetter. The authors instead apply a constant weighing factor of 18% throughout the record in their **correction. This issue should be addressed and the weighing factor should vary over time according to the humidity conditions as inferred using an independent proxy (for instance Artemisia pollen percentages).

Answer for authors: The comment above is based on a misunderstanding and we will clarify this in a revised version of our manuscript. We did not apply a constant weighing factor in our **correction (but in our *correction we did). The model weighs the leaf water isotope enrichment (i.e. εterr-aq) with only 18% for the fraction derived from grasses (estimated through the pollen record). This results in a lower reconstructed relative humidity during the dry YD, as opposed to the * and uncorrected model versions, since according to the hypothesis which forms the base of this correction, leaf water enrichment under dryer conditions is larger than recorded in grass leaf wax n-alkane δ2H values, as opposed to wetter conditions (see discussion in Kahmen et al. 2013 and in line 499-522 in our manuscript). That no constant weighing factor was applied in the **correction (as opposed to the *correction) is also visible in Fig. 3, where the * and the **correction show different degrees of changes, in particular during the YD. If ** would have used a constant weighing (as was used for *), then both curves should covary and feature a constant offset.

4) Despite the authors' effort to address all the potential errors that accompany their data, there is a large source of uncertainty that has been neglected and that has been

allegedly circumvented by simply including the analytical uncertainty of the δD measurements. This source of uncertainty is the variability associated with the apparent isotope fractionation values of C3 dicots and C3 monocots as observed for modern plants (e.g. Sachse et al., 2012). This variability should generally be accounted for when estimating Rh. It should also be taken into consideration when calculating the vegetation corrected εterr-aq*, as the authors simply apply the mean εapp difference between dicots and monocots disregarding the related uncertainty, which can be quite substantial (as high as 30‰ at 1 sigma) for each plant group. While I understand that quantifying this uncertainty would disproportionally increase the error bounds of their Rh reconstruction, I think the authors should at least discuss this issue more openly in the text. Furthermore, along these lines I believe that the authors should attempt at estimating how well the pollen records represents the local vegetation cover and include this source of uncertainty in the error propagation equation.

Answer from authors: Firstly, we note that this discussion only affects the reconstructions derived from the "vegetation corrected" DUB* model (not the DUB** or the not vegetation corrected results). We respectfully, but decisively disagree with the reviewer, that adding the variability of the apparent fractionation (i.e. the isotopic difference between source water and n-alkanes) would result in more accurate estimations of DUB* model uncertainty. This is because the observed variability in the apparent fractionation for higher plants is due to at least two different processes: a) differences in the amount of plant transpiration, i.e. leaf water isotope enrichment during leaf wax synthesis, which likely cause the larges variability b) differences in biosynthetic fractionation among different species see (Kahmen et al., 2013a; Kahmen et al., 2013b; Sachse et al., 2012)

With the DUB approach we actually account for the large part of the variability caused by these processes: we assume that δ2Hterr does not represent source water, but leaf water of plants, as such removing the variability caused by process a) from the model. As of now, we have not enough information to estimate the amount of variability originating from process b), but it's likely significantly smaller than from a) in particular when only a limited number of species are n-alkane producers. Because of this uncertainty we also used another approach for obtaining "vegetation corrected" results (DUB**), which relies on a completely different hypothesis (see above). We note that all three DUB model runs (with and without vegetation correction) show quite similar results. So we rather see the range of results from the 3 approaches 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"

5) I am quite sceptical about the use of chironomid-based temperature reconstructions to infer air temperatures. Although I recognize that as yet lacustrine midge assemblages are one of the best proxies for summer air temperature, they have shown to incorporate a number of environmental signals apart from air temperature alone, most importantly catchment vegetation, nutrient levels, lake depth and seasonality (Eggermont and Heiri, 2012; Luoto, 2010). It is therefore likely that this biological proxy was very sensitive to the major environmental and seasonality shifts that occurred at the onset and termination of the YD. Especially, colder and longer winters during the YD might have resulted in relatively colder surface water temperatures during the growing season relative to the air (e.g. replenishment of the local aquifer via snow thawing), in contrast to the preceding warm AL phase. As chironomids are sensitive to water temperatures, it is hard to say to what extent the proxy is biased towards colder temperatures during the YD, not mentioning that the authors use a Dutch record thus making impossible to assess local versus regional factors. I think some of these issues should be openly discussed in the paper. Moreover, if I remember correctly the chronology that underpins the record from Hijkermeer is based on 14C dating of regional pollen boundaries that have been dated elsewhere. This implies that the temperature record comes with fairly large age uncertainties as compared to the proxy records at MFM,

where the chronology is much more accurate. Assuming that the alignment between Hijkermeer pollen records and the regional pollen stratigraphy is precise, I wonder if the author can include in their error propagation estimate the age uncertainty associated with the temperature reconstruction. Even though I understand temperature plays only a minor role in the DUB model, in my opinion, this would result in a much more rigorous estimation of the true uncertainties that accompany the reconstructed Rh. In addition, I suggest that for reference the authors plot the temperature record together with the Rh reconstruction.

Answer from authors: We are aware of the possible uncertainties of chironomid-based temperature reconstruction to infer air temperatures, as well as of the age model uncertainties and local vs. regional differences, since we compare 2 different sites. The uncertainties in temperature estimations, including a potential seasonal bias, are somewhat included in the uncertainty of the results, which we propagated into the DUB model. Of course, ideally a local T record should be used, but as none is available from MFM, we used the closest record. Also here we are interested in relative changes, i.e. differences between temperatures before and during the YD. Newer studies suggest that these temperature patterns are spatially different between N, Central and S Europe, but very consistent within these regions, which is also supported by modelling exercises (Heiri et al., 2014). As such, we see it as a well supported assumption, that temperature differences between the YD and the preceding and following time periods are well represented by this Hijkermeer record.

The age model uncertainty (from the Hijkermeer 14C based age model) is implicitly included in our calculations, due to the different temporal resolution of both records (Hijkermeer record consists of 37 datapoints, whereas the our MFM εterr-aq dataset of 106 datapoints). Therefore we calculated an equidistant time series for comparison, which results in a (time) averaged record of the lower resolution dataset. We don't see this loss of resolution as problematic, as also here we are interested in relative changes, i.e. differences before, after and during the YD. We can add this more explicitly to the method section, but refrain from further assessing the potential uncertainties in chironomid based temperature reconstructions, as this methodology has been covered elsewhere and validated in particular for temporal differences during the Late Glacial period with climate model data (Heiri et al., 2014).

6) Since the authors decided to reconstruct Rh across the Younger Dryas, I think it would be appropriate to briefly present the status of the knowledge on this climate event (as well as to better frame their results into a paleoclimatological context). I suggest to mention the ocean–sea-ice–atmosphere mechanisms that would explain the climate variability observed in European climate reconstructions during this period (e.g. Brauer et al., 2008; Lane et al., 2013; Muschitiello et al., 2016; Rach et al., 2014). I would also recommend that the authors discuss the current understanding of hydroclimatological variability at the onset and termination of the YD in Europe based on the lake sedimentary δD and $\Delta\delta$Dterr-aq reconstructions available so far.

Answer from authors: We believe an extensive discussion or analysis of the current understanding of hydroclimate variability is not in the scope of our manuscript, as we don't conclude on the mechanistic drivers of these processes here, which is done elsewhere in the literature, but rather present a new approach to estimate quantitative hydrological data. Therefore we refrain from further adding a discussion in our manuscript.

Specific comments The DUB is based on a number of important assumptions that are discussed along the text (I counted at least 14 in the paper, some of which are "hidden" between the lines). I wonder if the author can provide a summary of these assumptions in the form of a table to facilitate rapid screening. Similarly, I suggest that all the model parameters, fixed variables, and sensitivity tests are summarised in separate tables. As they are, these information are hard to piece together.

Answer from authors: We agree with the reviewer, and will provide a table with important assumption and fixed parameters in the supplement of a revised version of our manuscript.

L170: The authors adopt a constant atmospheric pressure value in their model. I would first like to know what value they use and why? Secondly, I would like to know how sensitive their model is to this parameter. Climate modelling studies have shown that there were considerable summer sea level pressure changes over Northern-Central Europe from the late AL to the YD (Menviel et al., 2011; Muschitiello et al., 2015). I would therefore be inclined to apply different sea-level pressure values across the deglaciation. Perhaps the authors can comment on this and openly discuss these problems in the paper.

Answer from authors: We applied a constant value for atmospheric pressure of 971 hPa, which is based on the barometric formula, i.e the mean atmospheric pressure at the elevation of MFM at 337m above sea level (pressure at sea level 1013 hPa), see line 207-208. The elevation of the lake has not changed during the investigated period and the model sensitivity to this value is very low (i.e. 0.05% rH change for a 100hPa changes, which encompasses the highest and lowest pressure ever measured in Germany for example), so that short-term weather related changes would not impact the results.

L176-182: Is there any empirical value that allows calculating Tleaf as a function of Tair? For the reasons I outlined in the previous comments, assuming that it is constantly (and equally) cloudy and/or windy at MFM during both AL and YD does not necessarily hold.

Answer from authors: Unfortunately there are no long-term data on this, but studies show that air and leaf temperature during daily cycles have a nearly 1:1 correlation for a temperature range between 15 – 20°C (Kahmen et al., 2011), but can deviate more at higher temperatures. Since during the cold YD the environmental temperature would not have exceeded this range significantly and our reconstruction datapoints integrate over a decade, we think this assumption is justified.

L487-488: A number of studies have shown a bi-partite structure of the YD with relatively drier conditions in Northern, Central and Southern Europe during the Early YD and relatively drier conditions during the Late YD (Bakke et al., 2009; Bartolomé et al., 2015; Lane et al., 2013). It surprises me that this mid-YD transition is not clearly captured in the Rh reconstruction at MFM. Although the authors claim that the record reveals "centennial scale excursions to higher ΔRh after 12.100 BP" I struggle to see any appreciable change in Rh variability. Critically, a marked shift in Rh after the mid-YD transition would support the reconstructed Rh, since virtually no significant vegetation shift had occurred during the YD and therefore the modelled Rh is independent of potential influences from local vegetation changes during this period. However, I must acknowledge that so far the mid-YD transition has been inferred only using qualitative or indirect hydro-climate proxies and thus a net shift from dry to wet conditions in Europe still requires conclusive evidence. Perhaps these issues can be briefly addressed in an apposite YD section of the paper (please see main comment on YD background discussion).

Answer from authors: We are aware that εterr-aq and also ΔrH reconstruction do show different mean values before and after 12.100 BP, i.e. we do reconstruct a slightly higher rH after 12.100 BP for all model runs (on average 3% higher). This 2-phased YD is also reflected in a variety of other proxies at MFM, for example the decrease of Artemisia pollen after 12.100 BP. Since the change in ΔrH is within our model uncertainty, we refrained from including this observation.

L491-494: How does the percentage of shrub pollen vary with respect to the percentage of tree and herb. If there is a strong covariance between shrubs, trees and herbs then it is not surprising that both the vegetation-corrected (using grass and tree+grass, respectively) Rh reconstructions correlate with the Artemisia pollen percentage (i.e. included in the shrubs pollen record) better than the uncorrected Rh. I would therefore like to know the level of covariance between the distributions of trees, grasses and shrubs at MFM. In addition, I would recommend that the authors include the relative shrub pollen percentages in Figure 3 for reference (please note that in the same figure either tree or herb distributions have not been plotted). I also wonder whether it is possible that the improved fit between the corrected Rh and Artemisia data (Figure 4) merely stems from subduing the Rh series variability when applying the vegetation correction. I believe that the paper would benefit from including some selected pollen diagrams (as supplementary material for example). Analogously, I think the original δD and $\Delta\delta$Dterr-aq records should also be presented in Figure 2 or 3. I also recommend that the author consider to include as Figure 1 their conceptual overview model of the hydrogen-isotopic relationship between source water and sedimentary lipids (Figure 6 in Sachse et al. (2012) and Figure S6 in Rach et al. (2014)) to illustrate the initial formulation steps of the DUB model.

Answer from authors: As mentioned in our manuscript (line 462-466) we assume Betula and Salix as the most dominant tree vegetation during the YD in the catchment of MFM from available pollen data. Only these two species were included in the calculation of the tree fraction (ftree) (see line 464). We did not use any “shrub” classification for any of our calculations (there is also no such classification existing for MFM in the literature), but only a “grass” classification for quantifying the fraction of grasses (fgrass). Since Artemisia does also not belong to grass classification (i.e. is not included in fgrass) the Artemisia record represents an independent proxy which therefore has no direct or indirect influence on the vegetation corrections.

Furthermore, we don’t see any necessities to include to the original δ2H data in our manuscript since these data are already published and extensively discussed previously (Rach et al., 2014). We would include a plot of εterr-aq as our main model input parameter. We also think that the manuscript would not benefit from a plot of the conceptual overview as suggested by the referee since these plots are already published several times (as mentioned by the reviewer). To better illustrate the model input parameters, we will include a conceptual overview on our modelling approach (similar to Figure 1 in Kahmen et al. 2011) in a revised version of our paper.

Line-by-line comments

Answer from authors: Issues below will be fixed in a revised version of the manuscruipt.

L77-78 and 87-88: In equations (1) and (2) please specify that the terms εbio refer to terrestrial and aquatic components, respectively (i.e. εbio (terr) versus εbio (aq)).

L159: The term esat "Saturation vapour pressure" should be introduced at line 148.

L172: Missing "of" after "function".

L243-244: Please provide reference for this statement.

L392: The line in brackets should start with small letters.

L416: "... low humidity treatment": how much?

L462: "...provides are more ... " should read " ... provides a more ... "

Figure 1: The data plotted in the upper-right panel are not in scale with the data presented in the upper-left panel. Please adjust.

Figure 3: Either the tree or shrub relative distribution is missing from the figure.

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