

## Reply to Anonymous Referee#1

We thank Anonymous Reviewer#1 for his/her constructive review. In the following we respond to all raised issues, and we hope that the comments and suggestions help to improve our MS.

### MAJOR ISSUES

*ISSUE 1. The authors make the following statements:*

*on lines 36-37 “the measured 2H leaf water enrichment as assessed by using the d2H<sub>prec</sub> and d2H<sub>n-alkane</sub> results and biosynthetic fractionation during n-alkane biosynthesis in leaves” and then on lines 321-324 “Given that n-alkane biomarkers are synthesised in leaves, they reflect the isotopic composition of leaf water (Kahmen et al., 2013), albeit with a systematic offsets of approximately -160‰ (Sachse et al., 2006; Sessions et al., 1999) referred to as biosynthetic fractionation.”*

*First, the d2H values of n-alkanes do not always reflect the isotopic composition of leaf water. They are certainly dependent on leaf water, but it is hardly the only control on their values. Differences in H isotope biosynthetic fractionation during water uptake can result in vastly different n-alkane d2H values among the plants that are very similar in their leaf water d2H values (see Eley et al., 2014, GCA v. 128, 13-28, but also the review by Sachse et al., 2012, Annu. Rev. Earth Pl. Sc., v. 40, 221-249).*

*Second, on line 126 the authors state that “Climatic factors and topography cause a pronounced vegetational zonation” in the study area. In order for the assumptions used by the authors to assess 2H-enrichment along the altitude gradient to be valid the authors need explain why they think that the changes in plant communities along the altitudinal gradient had no effect on the extent of biosynthetic fractionation between leaf water and n-alkanes among different plants species?*

*Can the authors exclude a possibility that the trend (labeled as “measured”) they show in Figure 6 is caused by changes in plant communities (i.e. changes in the apparent fractionation factor between leaf water and n-alkanes) rather than changes in relative humidity as they claim?*

Response:

→ We thank Reviewer#1 for raising this important issue. Indeed, we are aware that large interspecies variations in fractionation between leaf water and leaf waxes are reported (see ll. 87-89 and respective citations). This holds especially true for halophytic plants as investigated by Eley et al. (2014) and possible mechanisms for this salinity dependence are reviewed by Sachse et al. (2012). This information will be added to the MS.

→ We did not investigate different plant species along the altitudinal transect (only soils and precipitation). Therefore we cannot specifically evaluate/quantify plant-species effects. However, the general trend of the modelled  $\Delta\delta^2\text{H}$  values (depending primarily on relative humidity) captures pretty well the measured  $\Delta\delta^2\text{H}$  values (assuming no plant-species effect). This strongly suggests that at least for the here investigated transect the plant-species effect is not dominant. This information will be added to the MS.

*ISSUE 2. Because “a major re-interpretation of Peterse et al. (2009) data is required, taking into account previously neglected changes of climatically controlled 2H enrichment of leaf water in plants with altitude” (lines 103-105) is considered to be key, it would be useful to see a more detailed explanation in the introduction as to what was wrong with the previous interpretation and what the significance of the re-interpretation offered here is.*

Response: We are happy to explain more specifically in the introduction that Peterse et al. (2009) interpreted their leaf wax (alkane) data to reflect the isotopic composition of precipitation along the Kilimanjaro transect, whereas our data suggest that the leaf waxes actually need to be interpreted in terms of reflecting the isotopic composition of leaf water.

The respective implications for paleoclimate and paleoaltimetry studies are currently explained in the Conclusion chapter, but we can briefly describe them also in the introduction.

*MINOR ISSUES:*

Lines 20-21, “generally reflect  $d2H$  of precipitation”: This statement should be expanded to add the phrase “and/or changes in relative humidity”.

→ included

Lines 41-42, “Both in paleoaltimetry and in paleoclimate : : : values can completely mask : : :”: I suggest restricting this statement only to paleoaltimetry. Even though it is likely that the mechanisms investigated in this study are applicable to paleoclimate studies, the authors do not explain why it would be the case.

→ changed

Line 53, “and some others”: Either say what these “others” are or eliminate this phrase altogether.

→ deleted

Line 91, “seasonality of  $d2H$  of leaf wax  $n$ -alkanes was detected”: The authors need to provide more detail here as to what they mean by seasonality and what aspects of it were explored in the studies they cite.

→ Given that we do not discuss our data in terms of seasonality we prefer to maintain the sentence like it is and to refer our readers to the here cited literature.

Line 94, “degradation effects on  $d2H$  of leaf litter”: Using the phrase “alteration effects” instead would be preferable.

→ changed

Line 185, ““Each sample was analyzed in triplicate”: What was the uncertainties associated with these measurements? It would be useful to mention it here and to refer the reader to SI where this is given.

→ Standard deviations for all triplicate measurements are reported in Table S1. We included here that the mean standard deviation of all measurements is 3.1‰ and in the following sentence that the  $\delta^2H$  results are reported normalized to the VSMOW scale and in per mille according to the common  $\delta$ -nomenclature.

Line 191, “ $H3+$  factor was determined”: Over what range (in mV) was  $H3+$  determined? Does it cover peak intensities for  $nC27$  and  $nC29$  alkanes reported in SI?

→ This information will be added to the MS.

Lines 241-242, “It is well-known that the isotopic composition of transpired moisture is similar to that of the source water (see Section 3.4)”: It would be useful to cite here previous research that demonstrates that.

→ We will include respective citations in the MS.

Lines 258-259, “Decreasing condensation temperature (if occurs) can also contribute to the observe effect”: The role of decreasing condensation temperature needs to be clarified here. Wouldn't be the case that a decrease in condensation temperate results in more negative  $d2H$  values, i.e. something opposite to what is stated in the preceding statement on lines 256-258?

→ Sentence deleted.

Line 302, “thus corroborating the results of Peterse et al. (2009)”: This is a bit vague. Could the authors be more specific about what Peterse et al. (2009) have to say about the trend in this interval?

→ We will specify during revision that also the data of Peterse et al. (2009) show a trend towards slightly more positive  $\delta^2\text{H}$  values with increasing altitude above 2000 m.

*Lines 364-365, “The leaf temperatures were assumed to be equal ground-level air temperatures.”: Is this a valid assumption? Aren’t leaf temperatures generally several degrees C cooler than ambient air temperatures because of transpiration? If the authors maintain that their assumption is valid, they need to provide a reference to support it.*

→ We agree that leaf temperatures are not necessarily equal with air temperatures. However, given that the temperature effect on the modelling results is negligible compared to the effect caused by changing relative humidity, our assumption/simplification is valid. We will add this information in the revised MS.

*Lines 374-375, “corrected for biosynthetic fractionation equal to -160 per mil”: Is this use of this number justified? See the discussion in MAJOR ISSUES.*

→ First, there is no alternative to assuming a constant value for the biosynthetic fractionation factor, because species-specific fractionation factors have not been investigated for our research area. Note that also paleoclimate studies have to assume constant values (so far). As explained in our response to the major issue 1, we suggest that species effects are negligible along the here investigated transect, because our modelling results show the dominant role of relative humidity and evapotranspiration and are in good agreement with the measured data. Nevertheless we agree with Reviewer#1 that the robustness of the biosynthetic fractionation factor of -160 per mille needs to be further investigated.

*FIGURE 1 Line of longitude on the image, change 37 O to 37 E. Also, in the caption to the figure, mention what the numbers next to the markers mean.*

→ OK

*FIGURE 4 (A) It would be useful to show uncertainty associated with each data point.*

→ included

*FIGURE 5 I suggest removing the data points that are not reliable. They do not really contribute to the discussion. Also, add uncertainty associated with each data point like it is shown in the data from Peterse et al. (2009).*

→ We prefer (i) not to remove the excluded data points from Fig. 5 because they illustrate that care has to be taken not to over interpret  $\delta^2\text{H}_{\text{alkane}}$  results obtained for small alkane peaks in chromatograms (see discussion ll. 291-298) and (ii) not to add uncertainties to each data point because due to overlapping error bars the clarity of Figure 5 would be reduced. Standard deviations for all data points are provided in Table S1.

*FIGURE 6 The label “measured”, when talking about 2H enrichment of leaf water, is misleading. The data are estimated using an approach with several assumption that need to be clarified. Also, given the importance of comparing the “measured” data and model results it would be useful a) to show the errors associated with “measured” d2H data and b) display modelled results as “line+symbol” rather than continuous lines.*

→ We will put “measured” in quotation marks and clarify this better during revision and follow the suggestion of Reviewer#1 concerning error bars and “line+symbol”.

*TABLE S1 The authors give numbers for peak amplitudes in mV for the samples that were run in triplicate. What do these numbers represent? Is it a mean or all of the 3 runs resulted in exactly the same mV value for each sample (Which I find high improbable.)?*

→ These are the mean areas of all three runs; we will clarify this in the revised MS.