

Dear Anonymous Referee #2:

We appreciate your thoughtful review of our paper. Below we respond to each of your comments and indicate how we plan to revise the manuscript accordingly. For clarity, we have enumerated your comments and copied them in their entirety below in blue italics; we follow each with our response directly below in normal font.

1. *- Structurally: there are some paragraphs with only one sentence - I am not sure this is within the journal template, I recommend the authors to structure the manuscript with more concise paragraphs and better connections between paragraphs. it will be an easier read for everyone. Related to that, there are many sections in the methods and results and none in the discussion. For instance, section 2.2 and 2.3 could be combined. Accordingly, subsections in the discussion also would be better and easier to follow the flow of the discussion as in results.*

We place high value on readability and appreciate your specific suggestions for improving this. We are happy to add subsections to the discussion, focusing fractionation within leaves, potential as a soil-based proxy, and possible applications of the proxy. We will combine sections 2.2 and 2.3 into a single section called "Sample collection and preparation."

We are less clear on how to implement the suggestions on conciseness and flow. The sections on isotopic notation and results are the locations in our paper where we have paragraphs that either have only one sentence or are very short, and our instinct is that these are appropriate levels of conciseness. We hope that these changes, in addition to the other clarifications and rewriting we have described, will succeed in achieving an accessible and digestible manuscript. We note that our two referees both mentioned the ease of reading the current draft, but had opposite view points. We will appreciate any further comments on this front.

2. *- it will be probably corrected during the post-review process but still, do not forget to format the citation within the text ex: page 2, line 14 (e.g. (Drake...))*

We agree and will correct this in the revised manuscript.

3. *- I highly recommend authors to provide the data to databases where it is easily accessible upon publication. We should be supportive to open science and open data policies.*

We agree to do this and plan to upload our database to PANGAEA <https://www.pangaea.de/>.

4. *I am missing an introduction to compounds used. A nice introduction to pheophytin is only done in the discussion until I reached that point I did not really get why we are looking at pheo rather than chlorins (as the title say) and chl as it was introduced in the introduction. Overall the intro part gave a nice discussion on N dynamics in terrestrial environments, including the PNL where I was hoping to see this also in the discussion. how compound specific isotopic approaches would advance our understanding of N dynamics? what input ^{15}N pheo will provide in terms of all the ongoing discussion? these could be implemented to discussion part in accordance with the*

introduction. Otherwise, the introduction could be (maybe should be) more technical and focus on more in compounds and isotopic fractionation for instance.

We appreciate this critique of missing pieces from our introduction and discussion. We agree we should provide a better introduction to the compounds used in the introduction, and to expand our discussion of how compound-specific isotopic approaches would advance understanding of N dynamics.

In the introduction section, after introducing chlorins as degradation compounds of the chlorophyll molecule, we will name pheophytin as a key chlorin of interest due to its deriving from chlorophyll breakdown in the presence of oxygen and absence of high temperatures. We will note that pheophytin is the chlorin previously found in greater relative abundance than other chlorins in organic soils and litter (Sanger, 1971). Our study found, and did not assume from the outset, that pheo a would be present in sufficient quantities for compound-specific isotopic analysis, and so we will keep further discussion of this point in the discussion section.

In the discussion section, we will discuss how the $\delta^{15}\text{N}_{\text{pheo}}$ proxy could be applied. We see two key opportunities to advance understanding of N dynamics here. First, the ability to track changes in foliar ^{15}N over time gives insight into factors affecting $\delta^{15}\text{N}$ of plants, notably the availability of nitrogen. A time series of $\delta^{15}\text{N}_{\text{pheo}}$ covering periods of change in atmospheric pCO_2 could be obtained from an aggrading soil with dated, buried horizons such as in permafrost and used to evaluate the PNL hypothesis (and we have a paper on this in prep). Second, comparison of the compound-specific $\delta^{15}\text{N}_{\text{pheo}}$ value with other proxy $\delta^{15}\text{N}$ values over the same time period would provide information on processes that cause them to deviate. Common sources of $\delta^{15}\text{N}$ proxy values are subaqueous sediment deposits such as from lakes, ungulate tooth enamel, and bulk wood or soil. Deviations in records of $\delta^{15}\text{N}$ of pheo and tooth enamel at a single site would highlight changes in factors affecting dietary fractionation, such as animal growth rate. Aquatic signals could be distinguished from terrestrial signals by comparing $\delta^{15}\text{N}_{\text{pheo}}$ from soil with $\delta^{15}\text{N}_{\text{chl}}$ of lake sediment. $\delta^{15}\text{N}_{\text{pheo}}$ could validate a bulk proxy record such as obtained from wood or black carbon, or highlight diagenetic limitations of the record. In the case of bulk soil, combining records of $\delta^{15}\text{N}_{\text{pheo}}$ with $\delta^{15}\text{N}_{\text{bulk}}$ would provide information both on N availability to plants and dominant pathways of loss, hydrologic or gaseous, at a site, allowing for comparison of multiple N cycle dynamics over time.

page 3 line 13:... terrestrial d15Nleaf : leaf subscript

Agreed, thank you. We will make this change.

page 4 line 8: is climate a landscape effect? maybe precipitation is a better word?

As we are studying impact of position on a “climosequence” along which both precipitation and temperature vary, referring to climate effects seems more appropriate than precipitation effects. We will replace “landscape effects” with “environmental effects” to avoid confusion over whether the effects we are measuring are relevant to global climate change or only to the landscape scale (product of shading, aspect, precipitation patterns, etc.).

page 4 paragraph starting from line 15 needs reconstruction, it is not an easy or maybe not well written paragraph.

We will rewrite this paragraph to improve parallel structure and shorten the final sentence.

page 5 line 21: first sentence is a sampling strategy should be in the below section (2.2).

We agree. The first sentence will be removed and the detail that the pits were dug in open, grassy areas with minimal slope added to Section 2.2 on sample collection. The second sentence will be added to the preceding paragraph where grazing is discussed.

Page 5 lines 29-30: (and generally many more) can authors be more specific?

We will delete the parenthetical phrase "(and generally many more)" from this sentence.

page 6 line 1: what depth is the deepest soil sample from?

We will add a phrase to say that the deepest pit was dug to a depth of 65 cm.

page 7 lines 13 and 15: JAMSTEC acronym should change places. line 15 should be in line 13

We agree and will correct this in the revised manuscript.

page 9 line 31 d15Nptheo - o is missing

We agree and will correct this in the revised manuscript.

page 10 line 6: ...along the soil profile do (?) not deviate

We agree and will correct this in the revised manuscript.

page 12 lines 1-2: citation needed at this sentence where pheo is introduced.

We will provide citations for both the biosynthetic and degradation pathways of pheophytin synthesis.

page 12 paragraph starting with line 22: Can authors provide more info on the ages presented here? where are mentioned other sites here? close by? this paragraph and information given here can be improved.

This is a good point. We will expand this paragraph to clarify that radiocarbon dates of 4130 and 8030 yBP were taken on soil organic carbon (SOC) deep within the soil profiles at climosequence sites A and D, respectively (Chadwick et al., 2007), as located in Figure 1, making them most like site C of our studied sites. All soils along the climosequence have the Hawi volcanic flow as their parent material, which cooled around 150 ky ago, and so can be considered to be the same age, though differing climate and vegetation across the range of sites would be expected to result in different rates of organic matter production, decomposition, and preservation in soil.

References: please double check the format some references are all in caps lock

We agree and will correct this in the revised manuscript.

Figures: 1: would it be possible to indicate the vegetation somehow on these maps?

Yes: we will at least be able to indicate locations of vegetation zone transitions between the sites labelled on our map. We may also be able to indicate the shape and extent of vegetation zones. Four broad zones of vegetation have been mapped on the leeward side of Kohala Mountain: lowland dry scrubland and grassland; lowland dry and mesic forest, woodland and shrubland; and wet forest and woodland (Pratt and Gon, 1998). This reference is an atlas that we are currently trying to obtain.

2 & 5: y axis title is cut, missing some part of 15N

We agree and will correct this in the revised manuscript. The Y axis label will be shifted to the right to avoid shaving off the top of 15N.

Table 1: please add any other info on the sites below the letter like elevation or precipitation.

Agreed. Average precipitation values will be added below the site letters so that the key site differences are evident at a glance.

Table 3: I think the names should be written italics

We agree and will correct this in the revised manuscript.

References

Chadwick, O. A., Kelly, E. F., Hotchkiss, S. C., and Vitousek, P. M.: Precontact vegetation and soil nutrient status in the shadow of Kohala Volcano, Hawaii, *Geomorphology*, 89, 70-83, 10.1016/j.geomorph.2006.07.023, 2007.

Sanger, J. E.: Identification and Quantitative Measurement of Plant Pigments in Soil Humus Layers, *Ecology*, 52, 959-963, 1971.