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Climate of the Past Discussions

Interactive comment on "Fire, vegetation and Holocene climate in the south-eastern Tibetan Plateau: a multi-biomarker reconstruction from Paru Co" *by* Alice Callegaro et al.

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Anonymous Referee #2 General Comments: In this paper Callergaro et al. present results from a biomarker multiproxy reconstruction of fire and vegetation from lake sediments Holocene on the Tibetan Plateau. The methodology used in this paper and the scientific aims of this study are will within the scope of this journal. This paper applies a clever approach where multiple lines of environmental evidence (i.e. fire, vegetation, human/animal habitation) can be reconstructed from the same samples using a relatively streamlined workflow. Additionally, I appreciate the authors' tactic of using data from the GCD to interpret their fire data within a regional framework. Despite these

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strengths, this paper could be improved by better presentation (figures) and clearer interpretation of the data. I found myself unable to follow the logic at times, and occasionally, the data and interpretations seemed at odds with each other. Adding more complete explanations of proxy interpretations (in both the text and the figures) may clear up some of this confusion. There were also times in the discussion where evidence from other studies was presented without being linked to the new data, and the new conclusions felt buried. Make sure to emphasize the novel contributions of your work and what it adds to the literature framework. I have outlined some more specific issues below. Making these improvements will greatly increase the readability of this paper and strengthen the arguments.

A: Thanks for your helpful review and your observations that are surely pivotal for improving our paper. We substantially revised and rewrote the paper following your indications. We have to say that we had some difficulties in replying to some of your observations, especially in the "technical corrections" part, due to the fact that the pages/lines that you indicated in the comments were not corresponding to the ones in the discussion paper file. We tried our best in finding the precise arguments in the paper and in responding to your questions. We reply to specific comments below.

Specific Comments: Page 8 Line 31: Are you using %BSi as a proxy for monsoon intensity? If so an added sentence explaining why would be helpful. Also did you measure %BSi or %TOM or is it from Bird et al 2014? Please specify.

A: We incorporated your suggestion in the new version of the paper where, however, BSi is no more used as indicator for ISM. Instead, we used lithics(%), whose data are inserted in figure 5(b). We also specified in the text that "more intense rainfall result in greater lithic deposition (Bird et al., 2014)". Moreover, we added dD per mill of C27 and C29, in figure 4(a), which are also used as Indian Summer Monsoon indicators. Figures 4 and 5 are now strongly different respect to the previous version of the paper. Finally, we specified, when necessary, that these data come from Bird et al. (2014).

Page 9 Lines 13: I don't know if concurrent increases between PAHs and TOM implies a specifically biogenic origin for PAHs, just that the total organics in the lake and the PAHs may have a similar source. Especially given that your aquatic/terrestrial indicators show an increase in terrestrial n-alkanes to the lake after 8 cal ky. Perhaps there is more windblown terrestrial material being added to the organic pool and that's why it's increasing? You touch on this in the next paragraph.

A: We agree with your observations. In the new version of the paper we dismissed the focus on PAHs and TOM, since, as suggested by Anonymous Referee #3, we reanalyzed all the samples for PAHs, including new target ions. The new obtained results are now shown in figure 2, that strongly changed. With the new PAHs data we calculated some diagnostic ratios such as Ant/(Ant+Phe), IP/(IP+Bghi) and FluA/(FluA+Pyr). Therefore, we improved the discussion and the comparison between PAHs and MAs, but the confrontation with TOM was considered to be pointless for the new paper. Moreover, we compared the aquatic/terrestrial indicators to the lake levels (from Bird et al., 2014) in a new figure 4(b,c,d).

Page 9 line 20: MAs are more water-soluble than PAHs, so this argument doesn't make a lot of sense

A: We agree with this point, indeed PAHs are more lipophilic than MAs. We talked about these differences in a new paragraph, that sounds as: "The explanation for the lack of levoglucosan and other MAs peaks during the period of the highest concentrations of PAHs (6.5-3 cal ky BP) may be due to: i) different burning temperatures and conditions, i.e. MAs are produced in smouldering and low temperature fires while flaming high temperature fires produce PAHs (Simoneit, 2002); ii) the lipophilic properties of PAHs, which have a low solubility in water (Haritash and Kaushik, 2009) while levoglucosan has a relatively higher water solubility, with an estimated half-life time of 5-8 days due to possible degradation from aquatic microorganisms who utilize the "free" form of levoglucosan (Norwood et al., 2013)".

Page 9 Lines 31-33: You need to explain and cite how you are interpreting these ratios and what is the difference between L/M and L/(M+G).

A: We incorporated your changes into the paragraph as the following: “Although MAs ratios cannot precisely point to the type of past burnt vegetation, these ratios can classify general vegetation types (Fabbri et al., 2009). However, due to the fact that galactosan presents a different biodegradation behaviour, the application of L/(M+G) ratio may be inadequate (Kirchgeorg, 2015). For this reason, we limited the discussion only to L/M ratio results. [...] In addition to the PAH ratios, L/M ratios can also help determine combustion sources. L/M emission ratios ranging between 0.6–13.8 may be due to softwood combustion, while ratios between 3.3–22 depict hardwood burning, and ratios 2.0–33.3 may be due to burning grasses (Fabbri et al., 2009 and references therein)”.

Page 10 Paragraph on Line 5: ACL and Paq represent indices for differential terrestrial/aquatic inputs I’m not sure how that directly relates to interpretations of fire and vegetation change. Make it clear if you are relating this to lake levels and climate because these proxies don’t explicitly address changes in terrestrial vegetation community.

A: Thanks for the request for clarification. We are using ACL and Paq in order to retrieve information on past vegetation changes in the lake catchment, since n-alkanes can record the organic input into and within the lake. We therefore clarified this point comparing these 2 ratios with lake level changes and climatic variations in the new figure 4 (a,b,c,d,e) where we parallel dD per mill of C27 and C29, ACL, Paq, lake level changes, summer insolation at 30° N.

Page 10 Line 19: Are you using this to say something about source area changes or vegetation community changes? This distinction is not clear.

A: If you refer to PAHs, in the new paper we discussed about their pyrogenic sources and no more about terrigenous sources (page 10 line 19). If you refer to sitostanol

(page 11 lines 12-13), due to the fact that human-related FeSts were below method detection limits, we suppose that its source have to be vegetation related.

Page 10 Line 23: Is there any evidence that sitostanol is correlated with grassy tissues? Citation?

A: We did not found these kind of correlation. However, derivation from vascular plants is reported (Vane et al., 2010).

Page 11 Lines 13-32: This paragraph is under “paleofire activity” but doesn’t mention fire at all, just gives some climate context. Perhaps having a climate section would be useful?

A: We agree with your observation. The paragraph to which you refer is actually on page 12, lines 3-24. Indeed, we removed this paragraph from the section 5.1 and we put it in a new section: “5.4 Atmospheric transport”.

Page 12 Lines 9-15: Throughout the manuscript, potential reasons for the differences between the MA and PAH records are mentioned, but no evidence is presented to support any of these interpretations over the other. Are there sedimentary changes (i.e. grain size) when these records diverge that indicate changes in transport to the lake? Are there changes in PAH ratios throughout the core that might indicate changes in transport/fire temperature? Perhaps there are inherent differences in the transport of MAs and PAHs due to their size/solubility differences? Why might the charcoal and PAH record correspond better than the charcoal and MA records? If these questions are explicitly addressed it will greatly strengthen interpretations of fire history. If you haven’t already, I’d suggest reading Denis et al. 2012 Organic Geochemistry, which has a good discussion considering transportation/degradation/fire temperature differences in lake fire proxy records.

A: Thanks for the questions. We have tried to address all of them in the new discussion sections. Due to the fact that new PAHs data were obtained, a new interpretation is ev-

idenced, explaining the divergence between PAHs and MAs as fire indicators. We also read Denis et al and used it for our interpretation. Are there sedimentary changes (i.e. grain size) when these records diverge that indicate changes in transport to the lake? We associated PAHs and lithics(%), evidencing that in some cases increased PAHs signal is concurrent with less lithics (drier periods). MAs, instead, are higher during the intense ISM of the early Holocene. Our explanation of this fact in the new paper sounds as: "Although PAHs are more of a local fire indicator than levoglucosan concentrations, PAHs are also affected by changes in atmospheric transport and associated precipitation. PAHs peak during periods of less intense ISM precipitation, as indicated by Paru Co lithics % in the periods 10.5-10.1, 7-5.8, 5.2-3.2 cal ky B (Figure 4). During these drier phases aridity could have increased regional fire activity (Section 5.1). However, this relationship between aridity and fire is not constant for the late Holocene Paru Co record as the increasing PAHs signal from 3 to 1.3 cal ky BP coincides with increasing lithic abundances that may be related to more ISM precipitation. Therefore, the 3-1.3 increasing PAHs could be related to a fire signal transported by ISM precipitation. Rainfall occurring together with or soon after with fire events scavenges PAHs particles from the atmosphere and increases deposition (Denis et al., 2012)". Are there changes in PAH ratios throughout the core that might indicate changes in transport/fire temperature? We calculated the ratios Ant/(Ant+Phe), IP/(IP+Bghi) and FluA/(FluA+Pyr) and we associated an absolute error to their values, due to the error propagation in the calculations. We plot these values in the new figure 2, in order to highlight that the use of PAH diagnostic ratios could be reconsidered due to high overlapping values and error propagation that may hinder the correct allocation of the source. Perhaps there are inherent differences in the transport of MAs and PAHs due to their size/solubility differences? Of course MAs and PAHs do not always record the same fire events due also to their size and solubility differences. PAHs, especially the heaviest ones, can record only local signal, whereas MAs can be both local and regional. In the new version of the paper, section 5.1 (paleofire activity) we reported that: "Higher molecular weight PAHs are more stable compounds compared to 3-4 rings PAHs. If we assume

that low molecular weight PAHs degrade at 500 °C, we have to assume that MAs may also degrade at this temperature, as maximum concentrations occur at burning temperatures centred around 250 °C (Zennaro et al., 2015 and references therein). In the Paru Co record levoglucosan concentrations are higher than PAHs during the early Holocene. Therefore, in order to explain this discrepancy, regional early Holocene fires must have been more frequent than local fires, producing high amounts of MAs, without excluding that atmospheric transport of levoglucosan to Paru Co was more efficient during the early Holocene. Therefore, this high abundance of levoglucosan may also be related to a regional signal, as MAs are capable of travelling hundreds of kilometres (Schüpbach et al., 2015; Zennaro et al., 2014)" and later, in section 5.4 (atmospheric transport) we said that "This monsoonal history may affect the transport of fire products to Paru Co. The difference between the Paru Co MAs and PAHs records may be influenced not only by the burning temperatures that produce the different products, as previously mentioned, but may also reflect changing atmospheric transport. MAs peak during the ISM maximum at Paru Co between 10 and 7 cal ky BP, which may reflect the long range transport of these fire aerosols associated with biomass burning on regional scales (Figure 4). MAs are generally considered as regional signals due to their ability to be transported longer distances than the more local PAHs, where this early Holocene levoglucosan peak may reflect either increased fire activity and/or changes in atmospheric transport. We may hypothesise that high levoglucosan concentrations during the early Holocene in Paru Co reflect the interplay between increasing influence of the ISM in the early Holocene resulting in wetter conditions and increase biomass on the southern TP (An et al., 2012) as well as increased Early Holocene winter monsoons causing a cold and dry climate on the north-eastern TP that is cited as a main driver for fire activity during this time period (Miao et al., 2017). Major transport to Paru Co could have come from the south via the ISM but, to best of our knowledge, no studies encompassing Holocene fire history exist from the possible southern source areas". Why might the charcoal and PAH record correspond better than the charcoal and MA records? We agree with your later comment on the fact that the comparison

with the GCD may no hold due to the too much wide continental area from which the sites were selected. Therefore, we decided to exclude this part of the work from the new version of the paper, due to the fact that this confrontation with the GCD was not helping with the data interpretation. Would be really interesting to compare our data with the GCD if this database would be richer in sites from the Tibetan Plateau and/or from the Indo-Gangetic Plain, Bay of Bengal and South Asia.

Page 12 Line 26-28: What particular climate effect would Bond events have on fire in Tibet? Link through a mechanism

A: Due to the substantial modifications in the results, thanks to the new analysis, section 4 and 5 of the new version of the paper are completely changed and the references to Bond events were considered to be pointless and removed.

Page 13 Lines 1-4: Like I said in the results, I don't think you can assume that the PAHs are biogenic. Not without some additional evidence. Have you tried to use some PAH degradation ratios? The two papers you cite here are good resources for these tools. I'd be interested to see if applying the appropriate ratios to your data provide evidence for degradation.

A: As we already said, due to the new data and interpretation, we did not assumed the PAHs are biogenic. We also calculated Ant/(Ant+Phe), IP/(IP+Bghi) and FluA/(FluA+Pyr) diagnostic ratios, that helped in the interpretation. A new paragraph describing the ratios sounds as: "PAH diagnostic ratios used in this study are Ant/(Ant+Phe), IP/(IP+Bghi) and FluA/(FluA+Pyr). Ant/(Ant+Phe) values generally discriminate between petroleum (< 0.10) and combustion (> 0.10) sources; IP/(IP+Bghi) distinguishes between different combustion sources, with values > 0.5 for grass, wood or coal combustion, values between 0.2 and 0.5 for liquid fossil fuel combustion and values < 0.2 for petroleum sources; FluA/(FluA+Pyr) is used to define the transition point (0.5) between petroleum and combustion (Denis et al., 2012; Yunker et al., 2002a; Yunker et al., 2002b; Yunker et al., 2015; Zakir Hossain et al., 2013). In Paru Co

these ratios are plotted with absolute error bars (Fig. 2g, 2h, 2i), in order to highlight that the influence of error propagation from the original analysis to the ratio values should be carefully investigated when assigning sources from the ratios. Considering the error bars, the three ratios shows values > 0.10 for Ant/(Ant+Phe), > 0.5 for IP/(IP+Bghi) and > 0.5 for FluA/(FluA+Pyr) for the majority of the analysed samples". As you suggested, we also calculated LPAH/HPAH as index of degradation (Stogiannidis and Laane, 2015), obtaining all values >1, that would signify no degradation and petrogenic sources. However, the cited paper is more focused on urban areas, so we think that applying this to our Holocene samples may be pointless, since the resulted values >1 do not make sense in our interpretation as pyrogenic PAHs.

Page 13 Line 5-15: I appreciate bringing in this discussion of transport: : :but I feel like how this relates specifically to your data gets lost. Does the peak at 5.6 represent increased wind, fire or both?

A: Due to the substantial modifications in the results, thanks to the new analysis, section 4 and 5 of the new version of the paper are completely changed and this reference to 5.6 wind event was removed. In the new version of the paper we talk about atmospheric transport from a regional point of view, listing the air masses movements that possibly brought fire signal to Paru Co.

Page 13 Lines 14-15: Are there dust layers recorded at Paru Co?

A: Thanks for the question. There are no dust layers recorded at Paru Co. We removed this paragraph from the new version of the paper.

Page 14 Lines 4-13: The times you highlight as having higher vegetation density are not all correlated with your fire records. Make sure you state when this mechanism applies and when it does not, and why that may be.

A: Thanks for the comment. We incorporated your suggestion in the new version of the paper, checking to be consistent and not to contradict the interpretation of the different

proxies. For example: “PAHs values are low in the early Holocene where, instead, tree pollen values are quite high. However, in the mid-Holocene PAHs contain higher concentrations from 6.5 cal ky BP, concurrent with a peak in the percentage of tree pollen. The subsequent decreasing trend in tree pollen, from 4.7 cal ky BP onward, is associated with a drying and cooling climate that may have intensified fire as recorded by PAHs in Paru Co, creating a positive feedback resulting in even more decreasing tree coverage. This decreasing trend in tree pollen reaches its lowest values after 3 cal ky BP. The regional wetter climatic conditions during the early and mid-Holocene may have favoured forest expansion, where this biomass became available for successive burning during the more arid climate of the late Holocene, when PAHs show indeed an increasing trend”.

Page 15: I think that you need to address explicitly how the vegetation changes (i.e. shrubs versus trees) that are observed in the record are (or aren’t) tied to your record of fire activity.

A: We agree and revised the paper according to your suggestion. Section 5.2 (combustion sources) and 5.3 (vegetation in the lake catchment) are now more focused on the relationship between fire and vegetation. For example, in the new version of the paper, we describe L/M comparing it to CPI: “In order to obtain more information from the burning conditions, we compared CPI values to L/M and PAHs. Considering that PAHs and n-alkanes are both local indicators, variations in CPI corresponding to spikes in local fire markers may link combustion and vegetation types demonstrated by n-alkane abundances. While no correlation exists between PAHs and CPI, the CPI and L/M have a slight positive correlation (r = 0.31, p-value = 0.03). Medeiros and Simoneit (2008) found that the n-alkanes in green vegetation smoke contained distributions ranging from C23 to C35, with strong odd-to-even carbon number predominance evidenced by CPI ranging from 9 to 58. MAs are better at recording smouldering fires than are PAHs, which may in part explain the similarity between MA and CPI variability through time. The Paru Co CPI values peak around 10 cal ky BP, in the period between 7.8 and 3.5

cal ky BP, and at 2.3 cal ky BP, with values up to 41.2, similar to the peak distributions of L/M. Another argument for relationship between CPI and MAs fire is the fact that lower temperature fires (MAs) essentially steam-distil the vascular plant lipids into the smoke, while high-temperature fires (PAHs) can result in decrease of the CPI[MOU1] (Schefuss et al., 2003 and references therein). In addition, the distance from the vegetation to the sediments may influence the CPI record as plants that are in or near the water pools contain shorter carbon chains, whereas more distant plants have higher CPI values (García-Alix et al., 2017). From these considerations can be assumed that, when CPI parallels L/M, fire from the surrounding areas, and not only near the lake catchment, could have been recorded".

Page 15 Lines 2-6: Leaves deposited into the lake are not the only source of terrestrial alkanes, they can also be ablated off leaves and transported by wind.

A: Due to the substantial modifications in the results, thanks to the new analysis, section 4 and 5 of the new version of the paper are completely changed and this part was considered to be pointless and removed.

Figure 3. Instead of color boxes interpreting the ISM instead show the original proxy record you are using for those interpretations. The Dalkane record from this same core from Bird et al. 2014 would be perfect to show plotted against proxies that you argue are influenced by ISM variations.

A: Thanks for the suggestion, we incorporated your changes in the new figures, which all strongly changed, where figure 2 compares PAHs, MAS, and PAHs ratios; figure 3 shows L/M, CPI, tree pollen, and PAHs; figure 4 shows Dalkane, ACL, Paq, lake levels, and insolation; figure 5 shows PAHs, lithics, MAs.

Figure 4. A and B could use some interpretive annotations. What to high/low MA ratios mean in terms of vegetation community? This is something that is unclear throughout the entire manuscript. How are these ratios interpreted and why do they differ? Adding some interpretive lines (like in 4C) would be very helpful. 4C. What is meant by arbitrary

units? Explain this in the caption.

A: Thanks for the suggestion, however it would be very confusionary to put interpretative annotations near to L/M plot, due to the intense overlap between ratio values. Moreover, we dismiss the use of Tang's data, preferring the dataset from Zhao et al., 2011 (http://apps.neotomadb.org/Explorer/?datasetid=14619)

Figure 5. I think plotting these records with a moving average obscures some important variability. It almost seems that the ACL and Paq have millennial cycles. Looking back at Bird et al. 2014, this seems to vary with reconstructed lake levels, rather than insolation as you argue. I think plotting that data in the same figure as these would really strengthen the interpretability of these proxies. Also, ACL and Paq and Norm31 and Sitostanol are really proxies for different things (aquatic v. terrestrial vegetation and changes in terrestrial community). It would be much clearer to separate these proxies into different figures. Perhaps, one figure with lake levels, insolation, ACL and Paq, and then add Norm31 and Sitostanol to Figure 4, which is your terrestrial vegetation figure.

A: We agree with your comment and we created the new figures according to your suggestion, except for Norm31, which was reputed not significant within the new data discussion.

Figure 6: Pg. 10 line 33 it is stated that the charcoal data from the GCD was drawn from a 1000 km radius from the Paru Co site: : :However, after a quick check on Google maps, I found all of the red dots on the Fig 6a map are actually more that 1000 km away from the site. Please address this contradiction and correct how the GCD sites were chosen in the text. Additionally, this large area of integrated charcoal records is potentially problematic because this data is drawn from a large continental area and the assumption that these sites are subject to the same climate conditions as the Paru Co site may no longer hold.

A: As already stated, the comparison with the GCD was removed, and consequently also figure 6 was deleted.

Figure 6 B and C: The shaded regions and arrows are not explained in the caption. And if they are showing correspondence then it almost seems that these records hardly reflect each other (which is discussed a bit in the text). Why is the PAH record not shown? I know it is highly variable, but it actually may correspond better to the charcoal record: : :if this is the case, then what are the implications for your data?

A: As already stated, the comparison with the GCD was removed, and consequently also figure 6 was deleted.

Technical Corrections:

A: We are sorry in saying that it was in some cases impossible to find where to address the requested changes, since we think that the indicated page/line references are wrong. We indicated this cases with “n.a.”

Page 2 Line 1: The dependent clause of this sentence is unclear

A: n.a.

Page 2 line 13-15: perhaps an i.e. style list of just three or so methods

A: n.a.

Page 2 Line : move “in the last Century” to after “biomass burning”

A: We are not sure if you refer to “biomass burning” of page 2 line 1 or page 2 line 5.

Page 4 Line 6: Perhaps this would be a good place to introduce Neolithic/bronze age societies you talk about in the discussion.

A: n.a.

Page 4 Line 13: cite Figure 1a

A: n.a.

Page 5 Line 30: cite Figure 1d

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A: n.a.

Page 6 Line 18: new paragraph at “Each Sample”

A: We revised accordingly. (found at page 6 line 26)

Page 7 Lines 18-19: What company did you obtain your standards from?

A: We added the companies and the sentence now sounds as: “We created response factors containing all of the target compounds as well as internal standards molecules (13C labelled acenaphthylene, phenanthrene and benzo[a]pyrene - Cambridge Isotope Laboratories, Inc; hexatriacontane and cholesterol-3,4-13C2 - Sigma Aldrich)”.

Page 8 Line 11-12: This topic sentence does not fit the content of the paragraph. Additionally, I think you actually end up arguing that these records don’t agree?

A: We suppose you refer to MAs and PAHs initial interpretation (page 9 lines 22-23). Section 4 was completely rewritten and the sentence was removed.

Page 9 Lines 10-13: This sentence is confusing and potentially unnecessary since you elaborate on it in the results.

A: n.a.

Page 10-11 Section 4.3: This reads like a methods section rather than results. Move this to methods and instead describe the trends you see in your analysis in the results.

A: We revised the paper removing section 4.3 (GCD results).

Page 11 Line 29: delete “a” before decreasing

A: The sentence was completely rewritten in this way: “The cooling trend after the Holocene Climatic Optimum (6.5-4.7 cal ky BP) correlates with decreasing solar insolation (Zhao et al., 2011), resulting in the decreasing strength of the Asian monsoon systems and in a drier climate across much of the TP”.

Page 12 Lines 15-19: This is a rambling sentence; perhaps splitting it into two would

make it clearer?

A: n.a.

Page 15 Line 6: you seem to be missing a word after sedimentological

A: The sentence containing “sedimentological” was removed due to the intense rewriting process.

Page 15 Line 8: delete “seems that and add “also” between “could” and “help”

A: The sentence to which you refer was removed due to the intense rewriting process.

Page 15 Line 26: I’m confused what you mean with “except when associated to PAHs”

A: The sentence to which you refer was removed due to the intense rewriting process. By the way, we meant that when MAs parallel PAHs the most probable interpretation would be local fires; instead, when these 2 fire proxies show different behaviour, the MAs signal could be related to regional/continental fires.

Page 15 Line 28: Citation needed Bush and McInerney (2013) GCA and/or Diefendorf et al (2011) GCA.

A: The sentence to which you refer was removed due to the intense rewriting process.

Page 15 Line 27: Be consistent with the use of pollens or pollen, don’t switch off

A: We revised the paper in order to be consistent with the use of pollen data / pollen records / pollens.

Page 16 Line 28: I think this is the first time you mention Bronze Age civilizations. You should elaborate on this earlier in the paper.

A: The sentence to which you refer was removed due to the intense rewriting process.

Page 16 Line 32: This is an abrupt way to end. Perhaps add a sentence of significance or implications?

A: The sentence of significance/implication were put at the beginning of the conclusions, which, in general, have been strongly modified due to the major revision process.

Figure 1. Include what the dates in 1D are based on (14C?).

A: We revised the caption incorporating your suggestion, as following: “(d) Plot of the age/depth model for Paru Co according to Bird et al. (2014) based on AMS 14C”

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