Replies to the reviewers

The Reviewers' comments are highlighted in italic font, whereas Authors' replies are in normal text.

Ref: https://doi.org/10.5194/cp-2018-19-RC1, 2018
Title: Fire, vegetation and Holocene climate in the south-eastern Tibetan Plateau: a multi-biomarker reconstruction from Paru Co.
Journal: Clim. Past. Discuss.

Anonymous Referee #1

General comments

In this manuscript, A. Callegaro and colleagues present a multi-proxy investigation from a sediment core retrieved from a Lake on the Tibetan Plateau. They conducted relatively novel biomarker analysis to reconstruct past fire activity and vegetation in this area over the last 11 kyr.

The paper is relatively well written and structured, and addresses scientific questions relevant to the scope of Climate of the Past. However the presentation of the results (figure) and more importantly the discussion and argumentation need to be improved and strengthened. I will highlight several cases under my "specific comments – major issues" where the argumentation was to superficial. Several possibilities for interpretation or explaining the discrepancies are often presented (e.g. fire activity, different transport, different fire temperature), which is good. But in the end the authors need to clearly state which one they favor and why. Looking at the figures, I mostly see discrepancies between the proxies. It may well be the case as the authors are comparing quite different indicators sometimes only marginally influenced by the parameter they are investigating (e.g. the effect of fire on n-alkanes). I strongly advise the authors not to go into too many directions, especially too many comparison with other proxies or records that do not match. But rather make sure they have good arguments for their interpretation in the end. I would rather recommend showing a record of ISM intensity or temperature over the Holocene.

The data are always presented with moving averages, which makes it difficult for the reader to appreciate for himself the original data obtained.

A: We thank Anonymous Referee #1 for useful comments that helped us improving the quality of the work. We substantially revised and rewrote great part of the paper following your indications. We reply to specific comments below.

Specific comments Major issues:

Page 9 line 2: I have to admit that I am quite skeptical about the high fluxes observed in both MAs and PAHs. As you state a few lines down, these high fluxes are the result of the higher sedimentation rate observed in the bottom part of the core. There is only one age point that is causing this high sedimentation rate, and no errors on this radiocarbon age are provided. Could it be that the sediment was distorted (stretched) during the coring process, causing these higher sedimentation rate. After checking Bird et al. (2014), it appears they dismissed a date which was much older (17.7 cal kyr) at

388.6 cm, as it was bracketed in between two ages in stratigraphic order. It could also be that your last one is a contamination during coring or bioturbation and that instead sedimentation rates were much lower in the bottom part of the core. Alternatively, the very high sedimentation rates could reflect an erosive event in the catchment. The TOM shows lower values for these older sediments. Erosion of catchment soil could bring also older fire biomarkers into the lake. Looking at the fluxes on figure 2b (ignoring the high peak), the MAs and PAHs don't show such similar patterns, except for the first maximum around 10.5 kyr BP and the following decrease to 10-9 kyr BP. You do make this statement further down on lines 23-24. You never actually explain this difference.

A: We agree with you the fact that the high sedimentation rate found in the deepest part of the core could have been derived by a distortion during the coring process or bioturbation, causing these higher fluxes of biomarkers. Due to this high uncertainty, we decided to discuss our biomarker's dataset only until 10.78 cal ky BP, as we state in our revised paper (P.6 L.12-14): "Since the deepest part of the core shows much higher sedimentation rate that cannot be clearly explained, with the possibility of data distortion, the subsequent description and discussion of the results exclude the samples aging 10.784-10.937 cal ky BP, limiting the dataset interpretation to the period between 1.347 and 10.768 cal ky BP". Moreover, as suggested by Anonymous Referee #3 we reanalyzed the samples for PAHs, including new target ions. The obtained results are different from the discussion paper and are shown in figure 2, that strongly changed.

Page 10 Line 25-26: You also never explain what could cause the difference between the 2 ratios L/M and L/(M+G), and which ratio is the more trustworthy, or which one you use for reconstructing the vegetation.

A: The difference between the 2 ratios is the inclusion of galactosan in the calculation. However, due to the fact that galactosan seems to show a different degradation pattern, we decided to discuss only L/M, as we state in the new version of the paper (P.10 L.2-4): "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."

Page 11, GCD results: I don't quite follow your argumentation. Why did you compile charcoal records over such a vast area? Are you expecting similar climatic trends over the Holocene, over this entire area 1000s of km across? If yes, then you should make it clear why (monsoonal systems etc). If no, then it doesn't make much sense to compile all of these into one record. You should also discuss in more details the different temperature of production of charcoal and levoglucosan, what does it imply? What explains different fire temperature, and what fire would you then expect to explain your data.

A: We agree with your observation on the fact that an area of 1000 km could be too wide to be considered as reference for Paru Co. Due to the fact that also Anonymous Referee #2 commented on this topic, we decided to exclude this part of the work from the new version of the paper due to the fact that this confrontation with GCD was not improving the data interpretation.

Page 13: It would have been useful to show a record of changes in monsoon strength (e.g. precipitation) along your own records.

A: As suggested, we added dD per mill of C27 and C29 figure 4(a) and lithics% in figure 5(b), retrieved from Bird et al. (2014), which are used as Indian Summer Monsoon indicators. Figures 4 and 5 are now strongly different respect to the previous version of the paper.

Page 13: If you would rather not trust your PAH record as a fire record, then you should make it much clearer earlier on, and mention that you will then only discuss MAs. Given the high variability of your PAHs, it may be your best option.

A: Since we reanalyzed all the PAHs fractions, we found new interesting results and we were also able to calculate Ant/(Ant+Phe), IP/(IP+Bghi) and FluA/(FluA+Pyr) diagnostic ratios. Therefore, we improved the discussion and the comparison between PAHs and MAs (section 5, from P.10 of the new version of the paper).

Page 14 line 7: Are you now argumenting that the MAs peak at 5.6 is due to transport, and not fire activity? A bit earlier you were discussing the Bond Event 4 and monsoonal precipitation. This is somewhat confusing. If both could play a role, you should add a summary sentences stating that.

A: In the new version of the paper we removed the association between MAs and Bond Event 4. On the contrary, we improved a lot the discussion of long-range transport, with a new paragraph called “5.4 Atmospheric transport” in which we tried to explain how one of the most probable source of levoglucosan seems to be atmospheric transportation from the South.

Smaller issues:
Title: I recommend adding “Lake” to Paru Co

A: The new title is “Fire, vegetation and Holocene climate in a south-eastern Tibetan lake: a multi-biomarker reconstruction from Paru Co”.

Abstract, line 24: I would briefly explain why PAHs decreases but MAs remain high. What is the distinction to the intense biomass burning during the early Holocene where both were high?

The new data show that high molecular weight PAHs peak in the early Holocene, and we address this fact to intense local fires and elevated burning temperatures. We explain this fact in the discussion section (P.11 L.1-2): “The high concentrations of higher molecular weight PAHs during the early

Holocene could be explained with local fires of greater combustion temperatures, due to the fact that higher number of rings requires greater burning energy (Denis et al., 2012) [...]” .

Page 2, line 5: In the sentence just before you state that fire contribute to greenhouse gases, this contradicts the end of the sentence here.

A: We agree and revised accordingly, now the sentence sounds as: “The impacts of greenhouse gases and associated global climate change on the frequency, intensity, duration, and location of biomass burning **are not well understood** and the contribution of fire emissions to past and future atmospheric composition **are also unclear** (IPCC, 2014)”.

Page 2, line 6: Why do you use "therefore"? I don’t see a clear link into this last sentence. It makes sense that these sentences are in the introduction, but there is not much flow, or logical order to these sentences. They just seem to be put together. Please improve the argumentation.

A: We modified the sentences according to your suggestion. The new sentences sound as: “**However, a recent study found that the synthesized Holocene fire record in eastern monsoonal China strictly tracks global atmospheric CO2 concentration from Antarctica (Xue et al., 2018), but it is still not clarified which one between fire and CO2 triggered the other. Therefore, more studies would be needed to improve human knowledge about past and present biomass burning events, which need** to be characterized and accurately mapped in order to investigate interactions with weather, climate, and landscape dynamics over a range of spatiotemporal scales.”

Page 2, line 13: (and throughout the manuscript) List references in chronological order, the oldest one first, the most recent one last)

A: Thanks for the indication, however, Climate of the Past does not require chronological order for the in-text citations, as it is explained in the website https://www.climate-of-the-past.net/for_authors/manuscript_preparation.html: “In terms of in-text citations, the order can be based on relevance, as well as chronological or alphabetical listing, depending on the author's preference”. We have chosen the alphabetical order and we checked to be consistent throughout the manuscript.

Page 2 line 16: Can you specify what type of ecosystem processes?

A: We modified the end of the sentence, that now sounds as: “and other environmental processes **such as vegetation growth, detrital influx, volcanic eruptions**”.

Page 2, line 18: Ice cores from where, also the TP?

A: In order to be more clear, we completely rewrote the sentence in this way: "Within the Tibetan Plateau (TP), only a few studies examine past biomass burning by using charcoal (Herrmann et al., 2010; Miao et al., 2017) or black carbon. Polycyclic aromatic hydrocarbons (PAHs) are reported in the lake sediments from the Tibetan Plateau (TP) spanning the last 2 centuries (Yang et al., 2016). Monosaccharide anhydrides (MAs), ammonia and black carbon in ice cores have been used as combustion proxies and indicators of fire on or influencing the Tibetan Plateau, but these records only cover the last century (Kaspari et al., 2011; Ming et al., 2008; Shugui et al., 2003; Xu et al., 2009; You et al., 2016b)".

Page 2 line 26: I don't see what you mean by specific environmental conditions?

A: The specific environmental conditions are explained after in the text, concurrently with the more detailed description of every class of biomarkers.

Page 2 line 31: I would clarify here that the following list of marker you are discussing are Mas

A: We agree and revised the sentence in this way: "**Within the listed biomarkers**, MAs are specific tracers of vegetation combustion".

Page 4 line 25-26: Please rephrase this sentence: the first part of the sentence is about difficult access to paleoclimate archives and then you mention few investigations into species diversity and plant communities. Where is the relationship?

A: We rephrased the sentence, that now sounds as: "**However, its remote nature restrains** access to possible paleoclimate **studies,** resulting in relatively few investigations **of** past species diversity and plant community changes".

Page 5 line 19: Define ecosystem functions or use another word, e.g. vegetation distribution?

A: We incorporated your change in the sentence as following: "More recently, human activities and related climate change have significantly altered the regional hydrology and **vegetation distribution** of the plateau, with degeneration of **plants** that led to desertification and frequent dust storms (Wang et al., 2008)".

Page 5 and Figure 1: It would have been more useful to have a more precise catchment map showing these features thant the large google map on figure 1 or the satellite picture showing only the lake.

A: In the new version of the paper we added a focus on atmospheric transport to the Tibetan Plateau. That is why in our opinion figure 1(a) is important to understand the continental position of the lake within the neighboring geographic areas. We have just zoomed in on the study region.

Page 6 Line 8: The 137Cs determination method is not cited here, whereas the radiocarbon is.

A: We added the expression “determined by direct gamma counting” in order to specify the 137Cs determination method.

Page 8 line 7: How did you obtain wet density?

A: We calculated wet density with this formula: dry density (g/cm^3) + (water content (%) * water density (g/cm^3)).

Page 8 line 11: for the other ratios you clearly state what they are useful for. You should do the same with the ACL, what can it tell you?

A: We briefly added the significance of ACL. The sentence now sounds as: “the average chain length (ACL), representing the composite of longer and shorter n-alkanes **between the chain length range of 21 to 33 and indicating the prevailing length**”.

Page 8 line 29: please provide these latitude an longitude ranges. How many records did you compile in total?

A: Since we removed the comparison with the GCD, this information can be neglected.

Page 9 line 7: or you had erosion of older compounds in the catchment (soil).

A: Thanks for the suggestion. As already specified earlier in the responses, in the new version of the paper we decided not to discuss the data of the deepest part of the core.

Page 9 line 30: Why didn’t you look at the correlation between BiSi and PAHs and TOM and PAHs?

A: The correlation between PAHs and TOM was indicated at page 10 line 14 in the discussion paper. However, due to the fact that the new PAHs data are different, we did not consider in the discussion these correlations.

Page 9 line 33: It would have been good to summarise here how the link ISM-BiSi works.

A: Thanks for the tip. In the new version of the paper, BSi is not used as indicator for ISM. Instead, we used lithics(%), inserted in figure 5(b), because more intense rainfall result in greater lithic deposition (Bird et al., 2014).

Page 10 line 1-9: These correlations should be presented when you first describe similar trends of both PAHs and MAs, at the beginning of the section!

A: We agree with the comment and, in the new version of the paper, we present the correlation values at the beginning of the result section.

Page 10 line 2: the (negative) correlation between MAs and TOM was larger than this (-0.54) and with a more significant p-value. Maybe there is something to discuss there, even though it is not positive as you expected.

A: Thanks for the comment. The correlation at page 10 line 2 was between PAHs and MAs. The correlation between TOM and MAs was at page 9 line 29. r = -0.54 was referred to the correlation between MAs and BSi, not to MAs and TOM. For this reason your question is not totally clear to us. By the way, we were not expecting positive correlation between MAs and TOM, because it would have been signified a relationship of MAs with organic matter.

Page 10 line 5: how does it vary? Does it vary with time, depending on the main climate?

A: That sentence was removed from the paper. However, an explanation about MAs catchment area is given in the new section 5.1 (paleofire activity) where we say that "MAs are **capable of travelling hundreds of kilometres** (Schüpbach et al., 2015; Zennaro et al., 2014)".

Page 10 lines 5-9: These last two sentences should probably be moved to the discussion section.

A: Thanks for the comment, we incorporated your changes in the new version of the paper. Now, section 4 is dedicated only to the mere description of the results.

Page 10 line 7: Unfortunately we don't see the original concentration in your figures, only the fluxes.

A: The new figures strongly changed due to the facts that we have some new data and that we decided to use concentrations instead of fluxes, because data discussion is focused between 1.347 and 10.768 cal ky BP, when sedimentation rate is constant.

Page 10 line 8: can you specify what you mean by "biogenic origin"? combustion of biomass is also biogenic for me

A: In the new version of the paper we the confrontation between PAHs and TOM was not considered for the data interpretation, so that sentence is no more present.

Page 10 Line 13-15: I don't fully understand this sentence, can you rephrase it to be more clear?

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 10 line 15: Is the statistic done only for this interval, or for the entire core? please specify

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 10 line 19: please provide some examples of these changes in the terrigenous environment.

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 10 line 28: It would be useful to either state the published range in the text, or in the figure. Are those ranges for L/M, or for L/(M+G)

A: As you suggested, we incorporated a better description of the L/M ranges in the new version of the paper, in the new section 5.2 “Combustion sources”, where we state, for example: “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”.

Page 11 line 1: the second part of this sentence is rather vague. n-alkanes do not record all organic input inot the lake, and they also record organic production within the lake.

A: We revised the sentence according to your suggestion: “Past vegetation changes can also be derived by variations in n-alkane ratios, as n-alkanes can record the organic input **into and within** the lake”.

Page 11 line 15: Here you should list the other FeSts, especially those that would have indicated the presence of humans.

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 11 Line 16-17: Are there other information (e.g. archeology) which could support this finding? No known settlement in this area, too high elevation, ...?

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 11 lines 19-23: All this first part should be in the method section.

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 11 line 31: I only counted 3 colour bars where the arrows go in the same direction, that's pretty bad as a similarity...

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 12 line 31: I wouldn't call this composite record regional, it's almost continental

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 13 line 5: How do these different burning temperature occur? You need to discuss this point in further details. Would we then have low or high temperature fire during this interval, and why, what caused this type of fire?

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and you would see that references to different combustion temperatures are made frequently throughout the text. For example: "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".

Page 13 line 14: The PAHs show a clear minimum at 8kyr. The Sum of MAs show a peak from 8 to 9 kyr BP. The 8.2 cal ky BP was a short and abrupt event, if the ISM was peaking then, I would not expect a 1000 year long dry interval.

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 15 line 4: that warm period would fall right into the 8.1 to 7.2 cold intervals (1-2 degrees cooling) you are mentioning a few lines up...that's contradictory

A: We agree and revised the sentence in order to be more clear. The new sentence sounds as: "**However, within this warm period, the climate had a sudden, intense change between 8.1 and**

7.2 cal ky BP with temperatures 1-2 °C below early and mid-Holocene levels and forests retreating downslope.”

Page 15 lines 6-7: please specify the time interval here. As you were just mentioning relatively young intervals (<2.7 kyr BP) where you don’t have fire records it is confusing. Do you mean for the early Holocene?

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 15 line 18-19: I wonder which one (forest or shrub) tend to have more fire? I also wonder if the presence of forest or shrub has an influence on the fire temperature?

In general, woody fires tend to have higher temperatures (350-550 °C) respect to grass fires (120-250 °C). Due to the fact that forest and shrubs are both woody plants, there is no much difference between them, but it depends mostly on the weather/climate. Fire temperatures are influenced by both quantity and quality of the fuel. In woody vegetation, backfires frequently burn longer and deeper but headfires are hotter.

Page 15 line 22-24: In this sentence is is hard to follow what the observations where, and what are the suppositions, could you reformulate more clearly what has been observed, and what is assumed?

A: We clearly reformulated the sentences, that now sound as: “After 5.2 cal ky BP, lake levels decreased, probably causing opposite fluctuations in both ACL and Paq, suggesting diminished ISM rainfall, reduced clastic deposition, and lowered lake levels, which leading to an invasion of the littoral zone on the core site and an increase in sand deposition (Bird et al., 2014). The fluctuations in both ACL and Paq are consistent with these lake level changes (Figures 3 and 4).”

Page 15 line 32-33: I don’t follow your argumentation. How can you infer that this mechanism would also occur in the sediments?

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 16 line 1-2: leaf waxes can also be abraded and transported by the wind, as well as in streams in suspended sediments, leaves are not necessarily requested for their transport and deposition

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 16 line 6: I don't understand what you insinuate here? What details would you look into? Please be more specific and/or provide examples.

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 16 line 25: No, they can also be transported quite far by winds.

How much do you mean with "quite far"?
In the new version of the paper we found that CPI and L/M have a slight positive correlation (r = 0.31, p-value = 0.03) suggesting that both local and regional sources are possible.

Page 17 line 1: what do you mean by paleoreconstruction information, this is too vague, could be paleotemperature, paleoprecipitation...

A: Due to the substantial modifications in the results, section 4 and 5 of the new version of the paper are completely changed and this sentence is no more present.

Page 17 line 27: You should not discuss something for the first time in the conclusion. This expansion of Bronze Age civilization should have been mentioned earlier. In the text, you mention 4.2k as the collapse of Chinese Neolithic cultures

A: Due to the substantial modifications in the results, also section 6 of the new version of the paper is completely changed and this sentence is no more present.

Page 31 – figure 5: Your lake is located at 30N, why do you use the insolation at 40N?

A: All the figures from 2 to 5 have strongly changed. Now we used insolation a 30° N in figure 4(e).

Page 31 – figure 5: There are dots in between the Paq and the ACL graphs, to which graph do they belong to?

A: All the figures from 2 to 5 have strongly changed. Paq and ACL are now shown in figure 4(b,c).

Page 31 – figure 5: I wonder what signal you would obtain if you were to use the same 5pt moving average on the Paq and the ACL. It seems to me that the Paq data without moving average show a signal similar to the sitostanol

A: All the figures from 2 to 5 have strongly changed. Paq and ACL are now shown in figure 4(b,c) and compared to lake level changes from Bird et al. (2014).

Page 31 – figure 5. I am not quite convinced by the insolation driven trend. Or at least I don't think this is the signal you should be looking for in your n-alkanes data over the Holocene. I am also not convinced by your green bars highlighting similar oscillations in Norm 31 and sitostanol. The youngest peak is relatively coeval, the one before is already almost opposite (sitostanol peak is closer to the Norm31 minimum than to its maximum). The third Norm31 peak is not coeval with any peak in Sitostanol. And the 4th peaks are again quite offset.

A: All the figures from 2 to 5 have strongly changed. The comparison between Norm31 and Sitostanol was removed, since it was not considered significant within the new data interpretation.

Page 32 Figure 6: The figure is of poor resolution. The blue dots on the map in panel a are not described/explained. If not used these should be removed. The oscillations in panel b (charcoal) can barely be seen. The curve and its envelope (dotted curves) are not explained. I suggest deleting "resulted" and "analysis" from the figure caption. The figure caption should also describe what the blue green and red arrows are for.

A: Figure 6 was removed, since the GCD comparison was no more informative within the new data interpretation.

Technical corrections
Page 2, line 11: delete provided

A: We agree and revised accordingly, the new sentences sounds as: "Lake sediments archive high-resolution histories of sediment flux, as well as climatic, hydrological and ecological changes, as long as the lakes preserve sediments through time"

Pag 2 line 28: delete "in buried sediments"

A: We agree and revised accordingly, the new sentences sounds as: "Significant concentrations of these compounds are present in soil and sedimentary archives with ages older than 10 cal ky BP".

Page 2 line 31: You could add "and longer timescales" after "the Holocene"

A: We agree and revised accordingly, the new sentences sounds as: "suggesting that degradation, if happening, is a low-kinetic process (Battistel et al., 2016) and that these compounds resist over the Holocene **and longer timescales**"

Page 3 line 17: I would here mention "diverse distribution of chain length"

A: We agree and revised accordingly, the new sentences sounds as: “Different types of plants have **diverse distribution of n-alkanes chain-lengths**”.

Page 4 line 1: replace “anthropological” with “archeological”?

A: With “anthropological” we mean a wider evidence of human presence (FeSts, human-related pollens, …) respect to “archeological”, that is only related to archeological findings.

Pag 4 line 2: replace “quantification” with “determination” (I wouldn’t say that we can truly quantify the presence of humans. We are not there yet.

A: We agree and revised accordingly, the new sentences sounds as: “Revealing human presence in lake catchments often relies on anthropological evidence, but advances in proxy development during the past two decades now allows **determining** of the presence of humans or pastoralism through steroid fecal biomarker concentrations”.

Page 5 line 5: You could mention the Younger Dryas & Bolling Alerod in your text.

A: We agree and we added “in the context of Bølling–Allerød and Younger Dryas events in the region” to the sentence.

Page 5 line 12: You could maybe use "superimposed on these oscillations" instead of “even with these oscillations”.

A: We agree and revised accordingly, the new sentences sounds as: “**Superimposed on these oscillations**, the general temperature trends affecting the TP include warm and humid climate in the early to mid-Holocene, as registered in sediments and dust deposits”.

Page 8 line 3: I would rather use data “analysis” instead of “elaboration”

A: We agree and revised accordingly. The title of the section is now “3.4 Data **Analysis**”

Page 8 line 9: Replace “significative” with “significant”

A: We agree and revised accordingly, the new sentence sounds as: “In order to help data interpretation, 2-tailed Pearson’s correlations were calculated in R with a 95% confidence interval (Supplement S3) with statistically **significant** results when p-values are < 0.05”.

Page 8 line 11: what do you mean with "useful for work"? I would rather say "useful for our study, or for our interpretation"

A: We agree and revised accordingly, the new sentence sounds as: “*N*-alkanes ratios **useful for our study** include [...]”.

Page 8 line 30: Here you should refer to the figure presenting this data

A: Due to the substantial modification of the paper, this part is no more included in the new version.

Page 9 line 17: use "up to" instead of "touching values till"

A: Due to the substantial modification of the paper, this part is no more included in the new version.

Page 9 line 21: remove the comma

A: Due to the substantial modification of the paper, this part is no more included in the new version.

Page 13 line 27: delete the second "not"

A: Due to the substantial modification of the paper, this part is no more included in the new version.

Page 15 line 2: Is there a word missing? (“a limited abrupt to”???)

A: Due to the substantial modification of the paper, this part is no more included in the new version.

Page 15 line 13: I would rather use another verb, for instance "place the Paru Co..."

A: Due to the substantial modification of the paper, this part is no more included in the new version.

Page 15 line 15: specify the time interval considered

A: Due to the substantial modification of the paper, this part is no more included in the new version.

Page 15 line 26: I would rather call this "long term trend" than "millenial scale".

A: Due to the substantial modification of the paper, this part is no more included in the new version.

Page 15 line 29: can you indicate by how much on average?

A: Due to the substantial modification of the paper, this part is no more included in the new version.

Page 15 line 30: same here, by how much? is it significant?

A: Due to the substantial modification of the paper, this part is no more included in the new version.

Page 16 line 4: “it seems that”

A: Due to the substantial modification of the paper, this part is no more included in the new version.

Page 16 line 11: slash and burn -> at which time?

A: We added “over the past 4600 years (Miehe et al., 2006)” in the sentence.

Page 16 line 11: Please indicate Lhasa on Figure 1.

A: We agree and revised accordingly.

Page 16 line 12: where exactly? Please show on the map.

A: We agree and revised accordingly.

Page 16 line 16: compared to (or "with respect to")

A: Due to the substantial modification of the paper, this sentence is no more included in the new version.

Page 16 line 32: Comparing data that resulted from (or originated from)

A: Due to the substantial modification of the paper, this sentence is no more included in the new version.

Page 16 line 33:“processes that happened”

A: Due to the substantial modification of the paper, this sentence is no more included in the new version.

Page 19 line 6: The journal and pages are missing.

A: We checked and added the correct missing details.

Page 29: Specify in the figure caption that the BSi axis is inversed

A: Thanks for the observation, but now BSi in no more used in our graphs.

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 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 evidenced, 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 C_{23} to C_{35}, 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

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**”

Anonymous Referee #3

General Comments:

In this manuscript, Callergaro et al. demonstrate the usefulness of a multi-proxy approach to reconstruct fire and vegetation change throughout the Holocene from a lake sediment record in the Tibetan Plateau. The research objectives and methodology used in this study are within this journal's scope. This manuscript attempts to reconstruct fire, vegetation change, and human presence nearby the lake using biomarkers, and discuss how the results and other regional analyses of fire and climate compare. This manuscript presents a unique and novel record for the TP region. However, this paper could be improved with better interpretation of data, as well as better figures and presentation of data. The data and the author's interpretation seemed to conflict with eachother, particularly with the fire records presented. Adding more background information, discussion, and analysis of some of the records (detailed below) could drastically improve this paper and the discussion of results. I have presented some more specific issues below, listed from "Major Comments" to "Minor Comments" to "Figure Comments". Making these improvements will greatly increase the readability of this paper and strengthen the arguments.

A: We thank you for your kind revision and useful suggestions that helped us improving data interpretation, figures and the whole paper. We reply to the specific comments below.

Major Comments:

I'm not sure I follow the PAH argument. In the manuscript, you argue that PAH track local and regional fire activity, along with MAs, in the early portion of the record (10.7- 8.7), but may switch to having biogenic origins after 8.7, just due to the fact that they correlate slightly with TOM. Looking at Fig 3, it seems as though the PAHs are actually making sense as a fire proxy more-so than MAs- lower values during the times of increased ISM rainfall, higher values during the times of decreased ISM rainfall. Furthermore, the "noise" in the PAH record looks like millennial scale fluctuations in the fire activity, which you aren't capturing in the MA records. I would suggest more discussion on the PAHs as potentially tracking fire activity, instead of just writing them off as being biogenic in nature. There are many different ratios of PAHs that studies have shown to prove useful in determining PAH source (i.e. biomass burning vs fossil fuel burning, biogenic vs burning, etc: : :). Possibly look into some of these ratios as well to see if you can determine a ratio that is suitable for developing your story. Some papers that use ratios include:

Denis et al. (2012). PAHs in lake sediments record historic fire events: Validation using HPLC-fluorescence detection. Org Geochem.

Miller et al. (2017). Local and Regional Wildfire Activity in Central Maine (USA) during the past 900 years. Journal of Paleolimnology

Yunker et al. (2002). Sources and significance of alkane and PAH hydrocarbons in Canadian arctic rivers. Estuar Coast Shelf Sci

A: We agree with all your comments. We checked the papers you suggested and used them for our interpretations. As you suggested in a comment below, we also checked the target ions for each compounds and repeated the GC analysis for PAHs fractions, with new interesting results. Therefore, we were able to calculate some diagnostic ratios, such as Ant/(Ant+Phe), IP/(IP+Bghi) and FluA/(FluA+Pyr). For example, in the new version of the paper, the description of the diagnostic 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 [MOU1] 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”.

I’d like to note that just through comparing Figures 2 and 6 by eye, it seems like the PAH record tracks the GCD regional composite record fairly well (at least way better than the MA record does). I would advise plotting the PAH curve on figure 6 – that way we can visualize how the PAH record tracks regional fire activity.

A: We performed a deep revision to the paper and, due to the fact that both referees #1 and #2 both commented on the difficulty of interpretation of the charcoal composite record within our Paru Co record, we decided to exclude this part of the work. We totally removed the confrontation with GCD, that was not improving the data interpretation. Moreover, as we responded to referee #1, an area of 1000 km or more could be too wide to be considered as reference for Paru Co.

Minor Comments:
Page 13, line 23-24: “In general, fire history shows a decreasing trend from 8 cal ky BP to the present” – this isn’t apparent based on the figures you show. The MAs decrease, but the PAHs steadily increase. Distinguish between the two instead of saying “the Paru Co fire history”

A: We agree with your comment, indeed, in the revised paper, we paid attention on keeping distinguished the fire recorded by PAHs from the MAs one. The sentence to which you refer was completely removed from the discussion, due to the new results that implied a deep rewriting and new interpretative points of view. For example, a new part of the discussion sounds as: “During the early Holocene (10.8-8.5 ky BP), levoglucosan, galactosan, mannosan and 5-6ring-PAHs show similar trends, with a general decreasing pattern and some higher peaks at 10.5-10, 9.2 and 8.5 cal ky BP. Both levoglucosan and PAHs record fires during the middle Holocene between 6.5-4 cal ky BP. PAHs increase during the late Holocene from 3 to 1.3 ky BP, while levoglucosan also contains peaks during this time period (Figure 2). The peaks in higher molecular weight PAHs during the early Holocene (Figure 2d) may be explained by local fires with higher combustion temperatures, due to the fact that the higher number of rings requires greater burning energy (Denis et al., 2012). High percentages of 4 -6 ring PAHs generally suggest the contribution of local high-temperature combustion origins (Yang et al., 2016), where such combustion may be the source of BePyr, the congener with the second highest concentration in Paru Co, but also of IP and Bghi, which are high temperature markers (Zakir Hossain et al., 2013). When fuel sources are uniform, hotter fires (at and above 500 °C) commonly produce high concentrations of BePyr, IP, Bghi (Zakir Hossain et al., 2013; Mcgrath et al., 2003). The lower, but not lacking, presence of 3ring and 4ring-PAHs could be due to the fact that lower molecular weight PAHs are more depleted due to weathering processes (Zakir Hossain et al., 2013). Their lower concentrations may also be due to the fact that the 3ring and 4ring-PAHs could have travelled farther as they are more volatile than the 5-6 ring PAHs. In addition, the 3ring and 4ring-PAHs may have photochemically degraded in the gas phase after emission to the atmosphere (Wang et al., 2010). 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). MAs continue to decrease from 8.5 ky BP to 1.5 ky BP whereas 3, 4, and 5ring-PAHs start increasing before reaching their greatest values between 6.5 and 4 ky BP (Figure 2-a,b,c,d,e,f). [...]".

Page 17, line 7: this should be labeled the MA fire history, not the overall fire history from your record. The PAH fire history shows the opposite of this – with lowest values at 8 cal kyr BP, and then a long term increasing trend. It could be beneficial to include coring location in lake and a bathymetric profile of the lake. Looking at lake bathymetry could give insight or possibly explain some of the trends seen in the data, and could manipulate the age-to-depth model so that it isn'tlinear in reality.

A: Thanks for the comment. As we answered previously, we kept separated MAs and PAHs fire histories in the new version of the paper. Regarding the bathymetric profile, we did not introduced it in the figure, but we added a phrase in section 2 (study area) as following: "The lake's watershed is 2.97 km2 and consists of a sloping glacial valley measuring 0.5 to 2.0 km in length with lateral mountain crests higher than 5000 m asl. **The maximum water depth of the modern lake is 1.2 m, with gently sloping sides, but may tolerate a total water level of about 3 m.** A central ephemeral stream channel and a second incised channel drain the lake's watershed and feed Paru Co with runoff. Outflow from the lake drains via a small stream channel located approximately 430 m west of the primary outlet (Bird et al., 2014)", and we also considered data for the Holocene lake level changes in our interpretation (from Bird et al., 2014), plotting them in the new figure 4, together with dD n-alkanes (Bird et al), ACL, Paq, summer insolation at 30° N (Berger and Loutre, 1991).

Please check target ions for each compound – for example, many studies that look at retene have a target ion of 219 instead of 234 (the compound's molecular weight). The mass spectra can be found here: https://webbook.nist.gov/cgi/cbook.cgi?ID=C483658&Units=SI&Mask=2380#IR-Spec. Using 234 may be adequate, but in some cases using non-major ions may "hide" compounds, particularly when running in SIM mode on a GC-MS. In your case, it seems that this may in fact be occurring, since you report retene was undetectable in most/all samples. Given the fact that retene is produced by combustion of coniferous trees, its surprising that retene is not found, given the fact that you mention a coniferous forest near the lake (line 24, page 5).

A: As you suggested, we checked again the target ions and ran again the samples on the GC-MS. Retene was detected and considered in the interpretation of the data, as we stated in the new version of the paper: "MAs continue to decrease from 8.5 ky BP to 1.5 ky BP whereas 3, 4, and 5ring-PAHs start increasing before reaching their greatest values between 6.5 and 4 ky BP (Figure 2-a,b,c,d,e,f). This difference may due to higher percentages of lignin burning (evidenced by retene peaks – Supplement S5) with respect to cellulose burning (represented by MA concentrations). Pollen profiles (Zhao et al) indicate an increased presence of trees between 7 and 3 ky BP, coincident with major peaks of retene (Supplement S5). The combination of low concentrations of the 5-6ring PAHs but abundant FluA, Pyr and BePyr suggests relatively geographically small , but more frequent wildfires (Zakir Hossain et al., 2013). This is what probably happened near Paru Co between 6.5 and 4 cal ky BP".

The spikes seen in all data at the beginning of the record has me skeptical of whether or not it is a true signal of some climate/environmental variability. Adding in some discussion (1-2 paragraphs) on other, more plausible causes of this (i.e. an event in catchment that was preserved in the sediment record, a coring artifact, etc: : :) could give your arguments more validity throughout the manuscript.

A: We agree with you the fact that the high sedimentation rate found in the deepest part of the core could have been derived by a distortion during the coring process or bioturbation, causing the higher fluxes of biomarkers. Due to this high uncertainty, we decided to discuss our biomarker's dataset only until 10.78 cal ky BP, as we state in our revised paper (P.6 L.12-14): "Since the deepest part of the core shows much higher sedimentation rate that cannot be clearly explained, with the possibility of data distortion, the subsequent description and discussion of the results exclude the samples aging 10.784-10.937 cal ky BP, limiting the dataset interpretation to the period between 1.347 and 10.768 cal ky BP".

Figure Comments:

Fig 1 a) the map seems a more complex than is necessary. The surrounding areas may not be as important to this study as the TP, so one option could be zooming in on the study region. One option could be to make it similar to the map in the supplement – that map is simpler and much easier to read, and having a map similar to that could more easily highlight the study areas in this figure. Also, you might *want to confirm with google about publishing google map images in academic journals – I'm unsure if there are any special permissions needed from Google, but it could be a good thing to check.*

A: In the new version of the paper we added a focus on atmospheric transport to the Tibetan Plateau. That is why in our opinion figure 1(a) is important to understand the continental position of the lake within the neighboring geographic areas. As you suggested, we have zoomed in on the study region. We had already checked on Google's permission's rules (https://www.google.com/permissions/geoguidelines.html) and the use in journals is allowed citing the sources.

Fig 2) do not overlap a) and b). This makes it seem as though there is a peak in values occurring at 4 cal kyr BP. There are multiple ways to fix this – you can either separate them so they don't overlap, or possibly highlight/box the areas in a) that are being zoomed in on in fig b).

A: All the figures from 2 to 5 have strongly changed. Now figure 2 shows sum of PAHs, 3-4-5 ring PAHs, levoglucosan, mannosan, PAHs diagnostic ratios.

Fig 3) try moving a) and c) y axes over to the right side – that way the axes are not overlapping or too close together.

A: All the figures from 2 to 5 have strongly changed. Now figure 3 is much more readable and alternating axis left/right are used in the figures.

Figs 2 and 5) use the same color between these two plots for similar things. For example, in figure 5 you use a gold line to separate ISM changes, while in figure 2 it is a blue line. Try to stay consistent in color schemes for the reader.

A: All the figures from 2 to 5 have strongly changed. We paid attention on using the same colours to indicate similar things.

Fig 6) needs to be higher resolution. On figures b and c, you can barely see the lines. Making the lines bolded/bigger, as well as saving a high resolution image, would help fix this issue. Furthermore, adding the PAH record, not just the MA record, would be very beneficial, as the PAH and GCD records seem to track eachother.

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

J. L. Toney (Referee)

The manuscript "Fire, vegetation and Holocene climate in the south-eastern Tibetan Plateau: a multi-biomarker reconstruction from Paru Co" uses a suite of biomarkers to assess vegetation and fire change during the Holocene from a sedimentary record of a small lake on the Tibetan Plateau. This study presents original data with potentially interesting new results on fire history that has not been widely studied on the Tibetan Plateau.

The manuscript is well written and methodologies are robust. Given that this is a relatively new field of research, mainly the application of fire-related biomarkers to paleoclimate records, there are additional aspects that the authors should consider.

A: We are grateful to Dr. Toney for the useful review and the kind suggestions. We improved the paper according to the suggestions and we incorporated most of the indicated references. We reply to the specific comments below.

PAHs: For instance, although they suggest that there are only a few studies of PAHs as tracers of biomass burning and only cite two (Page 3, Line 12), there are others out there that may help with their interpretations, for instance: Page et al. 1999, Marine Pollution Bulletin; Yunker et al. 2002, Organic Geochemistry; Denis et al. 2012, Organic Geochemistry; Yan et al. 2014, Environmental Toxicity and Chemistry; Yunker et al. 2015, Organic Geochemistry; and Denis et al. 2017, Organic Geochemistry. In particular, not all PAHs result from biomass burning, so using the sum of PAHs, for example, may not be as useful as targeting the pyrogenic PAHs (examples in Page et al. - fluoranthene, phenanthrene, benzo(e)pyrene). Denis et al. 2012 suggest that there are differences in high molecular weight PAHs representing the intensity of the fire (also see McGrath et al. 2003, Journal of Analytical and Applied Pyrolysis), whereas, the low molecular weight PAHs more consistently record local fire events. These considerations may or may not be applicable, but could be tested without acquiring more data. This analysis may help to resolve differences in between the MAs and the PAHs.

A: Thanks for the useful guideline. We incorporated your changes in the sentence (page 3 lines 5-10 of the new version) as following: “PAHs are semi-volatile, persistent, and ubiquitous in the environment with multiple possible sources, and therefore commonly detected in soil, air, and water (Abdel-shafy and Mansour, 2016; Johnsen et al., 2005). **The investigation on PAHs as tracers of biomass burning in past climate archives such as sediments (Jiang et al., 1998) and ice (Gabrieli et al., 2010) increased in the last decades (Yan et al., 2014; Page et al., 1999)**”.

Following also the indication from referee#3, we reanalysed all the PAHs fractions, obtaining new interesting results which also include the application of some diagnostic ratios: Ant/(Ant+Phe), IP/(IP+Bghi) and FluA/(FluA+Pyr). We also looked at the signal from different group of PAHs according to their molecular weight. The paper went, therefore, through a deep revision in sections 4 and 5 (Results and Discussion). For example, a new paragraph describing PAHs results sounds as: “The lowest PAH value is 0.2 ng g-1 of benzo[b]fluoranthene (BbFl) while the highest PAH concentration is 310.3 ng g-1, of naphthalene (Naph). Phenanthrene (Phe), benzo[e]pyrene (BePyr) and Naph respectively represent

20.9%, 18.9% and 17.5% of the total PAH signal in Paru Co. The total sum of PAHs (∑PAHs) shows higher valuesin the middle Holocene, with major peaks at 6.3, 5.8, 5.2, 4.8, 3.9 and 3.5-3.3 cal ky BP. The general trend shows increases from 2.2 to 1.3 cal ky BP. The molecular weight and/ or number of aromatic rings of PAHs allows investigating the influence of different PAH types through time. The group of 3ring-PAHs includes Phe, anthracene (Ant) and fluoranthene (FluA), demonstrating a similar pattern to the ∑PAHs. The group of 4ring-PAHs encompasses pyrene (Pyr), benzo[a]anthracene (BaAnt), chrysene (Chr), retene (Ret), benzo[b]fluoranthene (BbFl) and benzo[k]fluoranthene (Bkfl), which also has higher values during the middle Holocene and then an increasing trend towards 1.3 cal ky BP. The group of 5-6ring-PAHs is composed by benzo[a]pyrene (BaPyr), BePyr, benzo[ghi]perylene (Bghi), Indeno[1,2,3,-c,d]pyrene (IP) and dibenzo[a,h]anthracene (DBahAnt), with a more noisy trend and dissimilar behaviour from the rest of the groups. 5-6ring-PAHs are high in the early Holocene, peaking at 10.3-9.9 cal ky BP, and then have separate high concentrations at 9.3, 8.6, 7.2, 5.2, 3.9, 3.5, 2 and 1.3 cal ky BP".

Another paragraph in which we discuss the PAHs ratios sounds as: "The diagnostic ratios and associated error propagation (Figure 2g, 2h, 2i) do not allow quantitatively assigning PAH sources. IP/(IP+Bghi) contains values above the 0.5 threshold for combustion of wood, wood soot and/or grasses, creosote, as well as almost all wood and coal combustion aerosols and bush fire (Yunker et al., 2002b). The FluA/(FluA+Pyr) ratio, with values above 0.5 for the majority of the samples, indicates the combustion of grass, wood or coal, although this threshold is not definitive (Yunker et al., 2002b). The Ant/(Ant+Phe) ratio with values > 0.10 is generally related to pyrogenic PAH sources, but overlapping values between petroleum and combustion sources are reported (Yunker et al., 2002b). In Paru Co, when including the error propagation, the majority of samples show values of Ant/(Ant+Phe) > 0.10. Due to the improbability that petroleum sources were burned near Paru Co during the geological time period covered by the analysed core, the obtained values for the ratio Ant/(Ant+Phe) must be related to vegetation combustion. In general, Ant undergoes more rapid photochemical reaction in the atmosphere than Phe. In contrast, FluA/Pyr and IP/Bghi isomer pairs degrade at comparable rates and the original composition information is preserved during atmospheric transport (Yunker et al., 2002b) suggesting that their ratios may be more reliable compared to the Ant/(Ant+Phe) ratio. Given these considerations, we confirm that diagnostic ratios are important tools for the source assignment but cannot be completely trusted due to overlapping values and error propagation that may hinder the correct allocation of the signal origin. However, PAHs in Paru Co can function as pyrogenic markers as we did not find any evidence of other sources (e.g. volcanic eruptions, anthropogenic emissions)".

Finally, with respect to the PAHs (Page 13, Line 28), if there is a change in the biogenic/ diagenic signal of the PAHs, then it would likely manifest specifically in the PAHs like perylene - it would be worth having a look at how the individual PAH profiles change when this signal becomes prominent. Degradation, if it is of the overall organic material should also manifest in changes in the carbon preference index (CPI) values, but these data are not plotted. If a low CPI is seen during times of highly charred material, this index could help support the argument made on Page 16, Line 3.

A: As we answered to the previous point, we looked at the at the signal from different group of PAHs according to their molecular weight. Due to the fact that the new interpretation addressed PAHs as pyrogenic derived, we no more discussed the biogenic/diagenic sources.
Regarding CPI, we calculated the ratio in range of chain length of 21-33 and we compared it to the fire signals, in the new figure 3. Our interpretation of these results in the new discussion section 5.2 “combustion sources” sounds as: “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 ($r = 0.31$, $p\text{-value} = 0.03$). 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 (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”.

n-Alkanes: It is worth applying some caution in the use of the Paq from Ficken et al. 2000, which was derived in from Mt. Kenya in Africa. The organic geochemistry community is finding that a site-specific approach may be needed and while the assertions about long-chain and short-chain n-alkanes generally hold true, in some environments the relationship is slightly more complex. For example, in Garcia-Alix et al. 2018, Scientific Reports, the supplemental information shows how this index and the ACL vary with distance from water source. Because grasses are prominent during more humid conditions in the arid Sierra Nevada region, the C31 shows aquatic rather than terrestrial-type vegetation changes. This may apply to similar high-elevation sites on the Tibetan Plateau and should be discussed. It is not a fault of the authors, just a really recent paper that might change the interpretations made here. This could help explain why the n-alkanes are showing a different pattern of change than the MAs and the grass/wood prevalence of pollen data (Page 16, Line 27). Overall, this is a very interesting and well thought out study, but further analysis given the above comments could help with discussion.

A: Thanks for your comments. We checked both Garcia-Alix et al. 2017, Scientific Reports and Garcia-Alix et al. 2018, Scientific Data, finding your useful consideration, as well as other papers. We focused our discussion on Paq and ACL in relation to lake level changes (in the new figure 4) and we found increasing ACL (and diminished Paq) when lake levels are lower and vice versa. Part of the new paper in which we explain this fact sounds as: “Fluctuations in lake levels (Fig. 4d) can be associated with fluctuations in Paq, suggesting a general relationship between higher lake levels and the prevalence of submerged plants between 10 and 5 cal ky BP. The opposite situation occurs between 5 and 1.3 cal ky BP, when a decreasing trend in lake level corresponds to diminishing Paq values. ACL confirms this trend with where the majority of values near 25 occur during higher lake levels (10-5 cal ky BP) and majority of values around 27 occur from 5 to 1.3 cal ky BP. The se high lake levels are consistent with wet conditions from a more intense ISM prevailing until ~ 6 cal ky BP, as evidenced by dD wax and pollen records (Figures 3 and 4).”

Thanks again for your comments that really improved the quality of our paper.

PAGES Data Review Team

The PAGES Data Stewardship Integrative Activity seeks to advance best practices for sharing the data generated and assembled as part of all PAGES-related activities. The CP Special Issue, "PAGES Young Scientists Meeting 2017" is part of this PAGES activity.

The co-editors of the Special Issue are reviewing the data availability within each of the CP-Discussion papers in relation to the CP data policy (https://www.climate-of-thepast. net/about/data_policy.html) and current best practices. The editor team is making recommendations for each paper, with the goal of achieving a high and consistent level of data stewardship across the Special Issue. We recognize that an additional effort will likely be required to meet the high level of data stewardship envisaged, and we appreciate the dedication and contribution of the authors. This includes the use of Data Citations (see example below). Authors are also strongly encouraged to deposit significant code into a suitable repository and to cite it using a Data Citation.

We ask authors to respond to our comments as part of the regular open interactive discussion. If you have any questions about PAGES Data Stewardship principles, please contact any of us directly.

Best wishes for the success of your paper.

YSM Special Issue editor team (E. Dearing Crampton-Flood, D.S. Kaufman, R. Barnett, M.F. Loutre, M.N. Evans, S.C. Fritz, C. Tabor, Y. Zhang, E. Razanatsoa, and H. Plumpton)

For this paper:

All papers submitted to Climate of the Past must include a Data Availability section that details the location of the data that were used as input to the study, including previously published data that were used for comparison purposes, and the data that were generated by the study.

(1) Research input data- The paper makes use of the Global Charcoal Database (GCD). In order to adhere to the Data Policy for Climate of the Past, persistent identifiers (doi or URL from NOAA Paleoclimatology), or full data citations to the primary data must be included in the Data Availability section.

A: We thank the PAGES Data Review Team for the comments and suggestions.

Fig. 1 (c) includes data from the TRMM dataset, which is not in the reference section. Please add the appropriate data citation to the paper.

A: We have added the appropriate data citation in figure 1(c).

Fig. 3 includes proxy data from Bird et al. (2014). We were delighted to discover that the data are already available in a public repository at https://www.ncdc.noaa.gov/paleo/study/16399. Add a data citation for this dataset to the figure caption and to the Data Availability section.

A: Now figure 3 is different and it includes data from Zhao et al. (2011) that we appropriately cited, adding the data citation as well:http://apps.neotomadb.org/Explorer/?datasetid=14619. Data from Bird et al.

(2014) are now included in the figures 4(a,d) and 5(b). We inserted the appropriate data citation to the figures' captions and to the Data Availability section.

Fig. 4 (c) includes a summary of pollen data from Tang et al. (2000) that was digitized from the original publication. Digitizing data from previous publications is a legitimate practice, but has important disadvantages. The accuracy is often degraded by the digitizing, and without essential metadata, the data are not available for reuse. Instead of digitizing previously published data, we encourage authors of this special issue to serve as data stewards by working with data generators to rescue and properly curate important datasets that are used in their paper. Please contact Tang to explain the reuse of the data and to offer to facilitate the transfer of the dataset to a data repository. Once deposited in a repository, the data can be cited with a data citation.

A: We agree with the fact that digitazing can reduce accuracy. However, we were not able to came in contact with Tang. We therefore decided to remove the comparison with Tang's dataset from figure 4 and we choose to compare our data with pollen dataset from Zhao et al. (2011), which is uploaded in Neotoma Data Publisher public repository (http://apps.neotomadb.org/Explorer/?datasetid=14619), displaying this data in figure 3(c).

Fig. 6. Include a data citation to the GCD in this figure caption and in the Data Availability section.

A: Due to the revision process we changed big part of the paper, dismissing the use of the GCD and therefore removing figure 6.

(2) Research output data – Biomarker data plotted in Figs 4a-b, 5b-e, 6c: Monosaccharide anhydrides (MAs), polycyclic aromatic hydrocarbons (PAHs), n-alkanes, fecal sterols and stanols (FeSts) – This paper presents new and valuable biomarker data for the south-eastern Tibetan Plateau during the Holocene. These new data must be uploaded to a long-standing online data repository, and a data citation or URL link from NOAA Paleoclimatology for access to these data must be provided in the Data Availability section of the paper. Charcoal index plotted in Fig. 6b: Composite of previously published data from 43 sites – This is an important new summary of the regional fire history based on available charcoal records. The outcome of the synthesis (the time series with uncertainties) should be transferred to a repository along with a table of metadata that includes the name and location and reference for each of the 43 sites. This product could be included on the same landing page, with the same doi/NOAA URL along with the biomarker data.

A: All the research output data, except for charcoal index that was removed from the paper, are now available at this link: https://www.ncdc.noaa.gov/paleo-search/study/24410.

LIST OF ALL RELEVANT CHANGES

The new manuscript include all the listed relevant changes, as can be seen from the marked up version of the paper, that follows from the next page.

1) The title of the paper is now “Fire, vegetation and Holocene climate in a south-eastern Tibetan lake: a multi-biomarker reconstruction from Paru Co”.
2) Due to the fact that new substantial work has been done for improving the paper, a new author is included within the co-authors, since her contribute was fundamental: Maria del Carmen Villoslada Hidalgo. The order of the co-authors names slightly changed.
3) Figure 1 is improved both in clearness and resolution.
4) Figures 2 to 5 are completely changed and improved in resolution.
5) Figure 6 was removed.
6) “Abstract” section substantially changed.
7) “1 Introduction” section substantially changed.
8) Section 2 is now called “2 Study Area, Modern Climate, and Holocene Climate History”.
9) “3 Methods” section substantially changed.
10) “4 Results” section completely changed.
11) “5 Discussion” section completely changed.
12) “6 Conclusions” section substantially changed.

Fire, vegetation and Holocene climate in ~~the~~a south-eastern Tibetan ~~Plateau~~lake: a multi-biomarker reconstruction from Paru Co

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Abstract. The fire history of the Tibetan Plateau over centennial to millennial timescales is ~~still unknown.~~not well known. Recent ice core studies reconstruct fire history over the past few decades but do not extend through the Holocene. Lacustrine sedimentary cores, however, can provide continuous records of ~~large-scale and~~ local environmental ~~modifications due to their~~change on millennial scales during the Holocene through the accumulation and preservation of specific organic molecular ~~markers throughout the past millennia. In order to~~biomarkers. To reconstruct Holocene fire events and vegetation changes occurring on the south-eastern Tibetan Plateau and the surrounding areas, we ~~improved and integrated previous analytical methods. The~~used a multi-proxy ~~procedure was applied to~~approach, investigating multiple biomarkers preserved in core sediment samples retrieved from Paru Co, a small lake located in the Nyainqentanglha Mountains (29°47'45.6" N; 92°21'07.2" E; 4845 m a.s.l.). ~~The investigated biomarkers~~Biomarkers include *n*-alkanes as indicators of vegetation, polycyclic aromatic hydrocarbons (PAHs) as combustion proxies, fecal sterols and stanols (FeSts) as indicators of the presence of humans or grazing animals and finally monosaccharide anhydrides (MAs) as specific markers of vegetation burning processes. ~~Relatively high concentrations of both MAs and PAHs demonstrate intense local biomass burning activity during the early Holocene (10.9-10.7 cal ky BP), which correspond to a drier climate following deglaciation. High concentrations of MAs but not PAHs between 10.7-9 cal ky BP suggest a period of regional biomass burning followed by a decreasing fire trend through the mid-late Holocene. This fire history is consistent with local vegetation changes reconstructed from both *n*-alkanes and regional pollen records, where vegetation types depend on the centennial-scale intensity of monsoon precipitation. FeSts were below detection limits for most of the samples, suggesting limited direct human influences on fire regime and vegetation changes in the lake's catchment. Climate is the main influence on fire activity recorded in Paru Co over millennial timescales, where biomass burning fluctuates in response to alternating warm/humid and cool/dry periods~~Insolation changes and the associated influence on the Indian Summer Monsoon (ISM) affect the vegetation distribution and fire types recorded in Paru Co throughout the Holocene. Early Holocene (10.7 – 7.5 cal ky BP) n-alkane ratios demonstrate oscillations between grass and conifer communities, resulting in respective smouldering fires represented by levoglucosan peaks, and high temperature fires represented by high-molecular weight PAHs. Forest cover increases with a strengthened ISM, where coincident high L/M ratios are consistent with conifer burning. The decrease in the ISM at 4.2 cal ky BP corresponds with the expansion of regional civilizations, although the lack of human FeSts above the method detection limits excludes local anthropogenic influence on fire and vegetation changes. The late Holocene is characterized by a relatively shallow lake surrounded by grassland, where all biomarkers other than PAHs display only minor variations. The sum of PAHs steadily increases throughout the late Holocene, suggesting a net increase in local to regional combustion that is separate from vegetation and climate change.

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1 Introduction

~~Terrestrial vegetation is the primary source of biomass burning (Simoneit et al., 1999), where this combustion is due to both natural processes and anthropogenic activities. Fire-related forest clearance contributes to the global burden of greenhouse gases and causes associated global warming (Bowman et al., 2009). The impacts of global climate change on the frequency, intensity, duration, and location of biomass burning are not well known and the contribution of fire emissions to past and future atmospheric composition are not clear (IPCC, 2014). Therefore, past and present biomass burning needs to be characterized and accurately mapped in order to investigate interactions with weather, climate and landscape dynamics over a range of spatiotemporal scales.~~

~~Lake sediments archive high-resolution histories of sediment flux, as well as hydrological and ecological modifications, provided as long as the lakes persist and preserve sediments through time (Yan and Wünnemann, 2014). Numerous recent studies demonstrate climatic variations throughout China and surrounding areas during the Holocene using lacustrine sediment as archives of the past climate (Bird et al., 2017; Dietze et al., 2013; Liu et al., 2009; Opitz et al., 2012; Saini et al., 2017; Yanhong et al., 2006). The paleoclimate proxies used in these studies including carbonate percentages, mineralogy, grain-size distribution, elemental geochemistry, stable isotope composition, leaf wax long-chain *n*-alkanes, aquatic diatoms and terrestrial pollens, collectively record changes in hydroclimate and ecosystem processes. Only a few studies examine past biomass burning by using charcoal in Tibetan sediments (Herrmann et al., 2010; Miao et al., 2017) or black carbon, levoglucosan and ammonia as combustion proxies in ice cores (Kaspari et al., 2011; Ming et al., 2008; Shugui et al., 2003; Xu et al., 2009; You et al., 2016b). However, these studies only investigate the fire history of the last century. Polycyclic aromatic hydrocarbons (PAHs) are reported in the lake sediments from the Tibetan Plateau (TP) spanning the last two centuries (Yang et al., 2016). To the best of our knowledge, however, no studies examine PAHs or monosaccharide anhydrides in sediments from the TP for the entire Holocene.~~

~~The combination of innovative molecular markers helps infer past fires, vegetation and human interactions, as highlighted in sediment cores analysis from Guatemala (Schüpbach et al., 2015) and East Africa (Battistel et al., 2016). Here, we use~~

~~biomarkers that are produced in specific environmental conditions and then transported, accumulated, and stored in lacustrine sediments: monosaccharide anhydrides (MAs); fecal sterols and stanols (FeSts); polycyclic aromatic hydrocarbons (PAHs); and normal(*n*)-alkanes. Significant concentrations of these compounds in buried sediments are present in soil and sedimentary archives with ages even older than 10 cal ky BP (D'Anjou et al., 2012; Johnsen et al., 2005; Schüpbach et al., 2015), thus suggesting that degradation, if happening, is a low-kinetic process (Battistel et al., 2016) and that these compounds resist over the Holocene. MAs are specific tracers of vegetation combustion (Simoneit, 2002; Zangrando et al., 2013). Cellulose pyrolysis creates the molecular marker levoglucosan (1,6-anhydro-β-D-glucopyranose) (Simoneit et al., 1999), while hemicellulose combustion produces the isomers mannosan (1,6-anhydro-β-D-mannopyranose) and galactosan (1,6-anhydro-β-D-galactopyranose) (Kuo et al., 2011). Several studies examine levoglucosan (L), mannosan (M) and galactosan (G) in aerosols and ice cores (Kehrwald, 2012; Simoneit, 2002; Yao et al., 2013; Zennaro et al., 2014; Zhang et al., 2008), as well as in sediment cores (Battistel et al., 2016; Kirchgeorg et al., 2014; Schüpbach et al., 2015), demonstrating the suitability of MAs as paleofire proxies. PAHs are a wide group of organic compounds made up of two or more benzene rings combined together in linear, angular, or clustered arrangements (Zakir Hossain et al., 2013). The physical properties of PAHs, such as low aqueous solubility and high lipophilicity, prevent microbial utilization and promote their accumulation in the particulates in terrestrial environments (Johnsen et al., 2005). This class of molecules is produced by incomplete combustion during a wide range of natural and anthropogenic processes, such as volcanic eruptions, vegetation burning, fossil fuels, garbage and cigarette or car emissions (Abdel-shafy and Mansour, 2016; Kim et al., 2013; Lima et al., 2005). PAHs are semi-volatile, persistent and ubiquitous in the environment, and therefore are commonly detected in soil, air, and water (Abdel-shafy and Mansour, 2016; Johnsen et al., 2005). However, due to their multiple possible sources, only a few studies consider PAHs as tracers of biomass burning in past climate archives such as sediments (Jiang et al., 1998) and ice (Gabrieli et al., 2010).~~

~~Leaf waxes are preserved in sediments and can help determine past vegetation in a lake catchment. The cuticular wax layer of terrestrial plants consists predominantly of long-chain hydrocarbons and creates a protective barrier that helps maintain the plant's integrity within an intrinsically hostile environment (Sheperd and Griffiths, 2006). The leaf wax of higher plants is difficult to degrade during transport, deposition and burial (Cui et al., 2008). Different types of plants have diverse chain-lengths of *n*-alkanes (Diefendorf and Freimuth, 2017). Angiosperms generally produce more *n*-alkanes than gymnosperms; however, chain-length distributions are highly variable within plant groups, and especially for conifers where the Cupressaceae group tends to have long chain *n*-alkanes, while the Pinaceae group tends to have relatively short chain *n*-alkanes (Diefendorf and Freimuth, 2017; Diefendorf et al., 2015). *Sphagnum* mosses are among the few plants that provide a characteristic signal as these mosses are marked by the predominance of C_{23} and C_{25} (Bush and McInerney, 2013). Long chain *n*-alkanes (C_{27}–C_{33}) with a strong odd/even predominance are usually interpreted to originate from terrestrial plants; mid-chain *n*-alkanes (C_{20}–C_{25}) are mainly present in aquatic macrophytes; bacteria, algae and fungi primarily produce short chain *n*-alkanes in the range C_{14}–C_{22}, while n-C_{17} is an indicator for algae and photosynthetic bacteria (Aichner et al., 2010; Ficken et al., 1998; Grimalt and Albaigés, 1987; Han and Calvin, 1969). Due to the large range of possible chain lengths present within sediments, ratios of *n*-~~

~~alkanes are often used to determine the vegetation distribution. Commonly used ratios are the average chain length (ACL) (Poynter and Eglinton, 1990), the carbon preference index (CPI) (Bray and Evans, 1961), the submerged versus emergent aquatic plants predominance ratio (P_{aq}) (Ficken et al., 2000), and the vegetation change ratio (Norm31) (Carr et al., 2014). Nevertheless, it is still unclear to what extent variations in leaf wax composition within paleoenvironmental archives can be explained in terms of changes in the relative proportions of different plant species and/or the reaction of a plant community to environmental conditions (Carr et al., 2014; Diefendorf and Freimuth, 2017).~~

~~Determination of human presence in lake catchments often relies on anthropological evidence, but advances in proxy development during the past two decades now allow quantification of the presence of humans or pastoralism through steroid fecal biomarker concentrations (Bull et al., 2002). FeSts, such as stanols and bile acids, in lake sediments reflect grazing in a lacustrine catchment (D'Anjou et al., 2012). Specific FeSts such as 5β-stanols are organic compounds produced by the microbially mediated alteration of cholesterol in the intestinal tracts of most mammals, making them ideal fecal biomarkers (Dubois and Jacob, 2016). Coprostanol and stigmastanol derive from hydrogenation of cholesterol and stigmasterol by bacteria present in the intestines of humans or animals and can indicate human presence and animal husbandry, respectively (Daughton, 2012; Vane et al., 2010). These molecules are also used as chemical indicators of fecal pollution of lakes, rivers, and drinking water (Daughton, 2012; Vane et al., 2010; Wu et al., 2009). In addition, FeSts can originate from vegetation, e.g. β-sitosterol is synthesised by higher vascular plants (Nishimura and Koyama, 1977; Vane et al., 2010) and its derivative β-sitostanol is generated from a reduction reaction in sediments (Martins et al., 2007).~~

The combustion of terrestrial vegetation by natural processes and anthropogenic activities are the primary sources of biomass burning (Simoneit et al., 1999). Human add to the global burden of greenhouse gases (Bowman et al., 2009) through fire-related forest clearance. The impacts of greenhouse gases and associated global climate change on the frequency, intensity, duration, and location of biomass burning are not well understood and the contribution of fire emissions to past and future atmospheric composition are also unclear (IPCC, 2014). However, a recent study found that the synthesized Holocene fire record in eastern monsoonal China strictly tracks global atmospheric CO_2 concentration from Antarctica (Xue et al., 2018), but it is still not clear if fire and CO_2 triggered the rise in the other component or vice versa. Therefore, more studies are needed to investigate interactions with weather, climate, and landscape dynamics over a range of spatiotemporal scales.

Lake sediments archive high-resolution histories of sediment flux, as well as climatic, hydrological and ecological changes, for as long as the lakes preserve sediments through time (Yan and Wünnemann, 2014). Numerous recent studies demonstrate climatic variations throughout China and surrounding areas during the Holocene using lacustrine sedimentary records (Bird et al., 2017; Dietze et al., 2013; Liu et al., 2009; Opitz et al., 2012; Saini et al., 2017; Yanhong et al., 2006). The paleoclimate proxies used in these studies including carbonate percentages, mineralogy, grain-size distribution, elemental geochemistry, stable isotope composition, leaf wax long-chain *n*-alkanes, aquatic diatoms and terrestrial pollen, collectively record changes in hydroclimate and other environmental processes such as vegetation growth, detrital influx, volcanic eruptions. Within the Tibetan Plateau (TP), only a few studies examine past biomass burning by using charcoal (Herrmann et al., 2010; Miao et al., 2017) or black carbon. Polycyclic aromatic hydrocarbons (PAHs) are reported in the lake sediments from the TP spanning the last 2 centuries (Yang et al., 2016). Monosaccharide anhydrides (MAs), ammonia and black carbon in ice cores have been used as combustion proxies and indicators of fire on or influencing the Tibetan Plateau, but these records mainly cover the last century (Kaspari et al., 2011; Ming et al., 2008; Shugui et al., 2003; Xu et al., 2009; You et al., 2016b). To the best of our knowledge, no studies examine PAHs or MAs in sediments from the TP during the entire Holocene.

A combination of innovative molecular markers were used to infer past fires, vegetation, and human interactions,in sediment cores analysed from Guatemala (Schüpbach et al., 2015) and East Africa (Battistel et al., 2016).Using a similar approach, we use biomarkers that are produced under specific environmental conditions and then transported, accumulated, and stored in lacustrine sediments: monosaccharide anhydrides (MAs); fecal sterols and stanols (FeSts); polycyclic aromatic hydrocarbons (PAHs); and normal(*n*)-alkanes. Significant concentrations of these compounds are present in soil and sedimentary archives with ages older than 10 cal ky BP (D'Anjou et al., 2012; Johnsen et al., 2005; Schüpbach et al., 2015), suggesting that degradation, if happening, is a low-kinetic process (Battistel et al., 2016) and that these compounds resist over the Holocene or longer timescales. Within the listed biomarkers, MAs are specific tracers of vegetation combustion (Simoneit, 2002; Zangrando et al., 2013). Cellulose pyrolysis creates the molecular marker levoglucosan (1,6-anhydro-β-D-glucopyranose) (Simoneit et al., 1999), while hemicellulose combustion produces the isomers mannosan (1,6-anhydro-β-D-mannopyranose) and galactosan (1,6-anhydro-β-D-galactopyranose) (Kuo et al., 2011). Several studies examine levoglucosan (L), mannosan (M) and galactosan (G) in aerosols and ice cores (Kehrwald, 2012; Simoneit, 2002; Yao et al., 2013; Zennaro et al., 2014; Zhang et al., 2008), as well as in sediment cores (Battistel et al., 2016; Kirchgeorg et al., 2014; Schüpbach et al., 2015), demonstrating the suitability of MAs as paleofire proxies. PAHs are a wide group of organic compounds made up of two or more benzene rings combined together in linear, angular, or clustered arrangements (Zakir Hossain et al., 2013). The physical properties of PAHs, such as low aqueous solubility and high lipophilicity, prevent microbial utilization and promote their accumulation as particulates in terrestrial environments (Johnsen et al., 2005). This class of molecules is produced by incomplete combustion during a wide range of natural and anthropogenic processes, such as volcanic eruptions, vegetation and/or garbage burning, fossil fuels, and cigarette or car emissions (Abdel-Shafy and Mansour, 2016; Kim et al., 2013; Lima et al., 2005). PAHs are semi-volatile, persistent, and ubiquitous in the environment with multiple possible sources, and therefore commonly detected in soil, air, and water (Abdel-Shafy and Mansour, 2016; Johnsen et al., 2005). The investigation of PAHs as tracers of biomass burning in past climate archives such as sediments (Jiang et al., 1998) and ice (Gabrieli et al., 2010) is increasing in the last decades (Yan et al., 2014; Page et al., 1999).

Leaf waxes are preserved in sediments and can help determine past vegetation in a lake catchment. The cuticular wax layer of terrestrial plants consists predominantly of long-chain hydrocarbons (*n*-alkanes) and creates a protective barrier that helps maintain the plant's integrity within an intrinsically hostile environment (Sheperd and Griffiths, 2006). The leaf wax of higher plants is assumed to be stable, and is difficult to degrade during transport, deposition, and burial (Cui et al., 2008). Different types of plants have diverse distribution of *n*-alkanes chain-lengths (Diefendorf and Freimuth, 2017). Angiosperms generally produce more *n*-alkanes than gymnosperms; however, chain-length distributions are highly variable within plant groups, and especially for conifers where the Cupressaceae group tends to have long chain *n*-alkanes, while the Pinaceae group tends to have relatively short chain *n*-alkanes (Diefendorf and Freimuth, 2017; Diefendorf et al., 2015). *Sphagnum* mosses are among the few plants that provide a characteristic signal as these mosses are marked by the predominance of C_{23} and C_{25} (Bush and McInerney, 2013). Long chain *n*-alkanes (C_{27}–C_{33}) with a strong odd/even predominance are usually interpreted to be of terrestrial origin; mid-chain *n*-alkanes (C_{20}–C_{25}) are mainly present in aquatic macrophytes; bacteria, algae and fungi primarily produce short chain *n*-alkanes in the range C_{14}–C_{22}, while n-C_{17} is an indicator for algae and photosynthetic bacteria (Aichner et al., 2010; Ficken et al., 1998; Grimalt and Albaigés, 1987; Han and Calvin, 1969). Due to the large range of possible chain lengths present within sediments, ratios of *n*-alkanes are often used to determine the vegetation composition. The most commonly used ratios are the average chain length (ACL) (Poynter and Eglinton, 1990), the carbon preference index (CPI) (Bray and Evans, 1961), the submerged versus emergent aquatic plants predominance ratio (P_{aq}) (Ficken et al., 2000), and the grass to wood prevalence ratio (Norm31) (Carr et al., 2014). However, it is still unclear to what extent variations in leaf wax composition within paleoenvironmental archives can be explained in terms of changes in the relative proportions of different plant species on the landscape and/or the reaction of a plant community to environmental conditions (Carr et al., 2014).

Revealing human presence in lake catchments often relies on anthropological evidence, but advances in proxy development during the past two decades now allows determining the presence of humans or pastoralism through steroid fecal biomarker concentrations (Bull et al., 2002). FeSts, such as stanols and bile acids, in lake sediments reflect grazing in a lacustrine catchment (D'Anjou et al., 2012). Specific FeSts, such as 5β-stanols, are organic compounds produced by the microbially mediated alteration of cholesterol in the intestinal tracts of most mammals, making them ideal fecal biomarkers (Dubois and Jacob, 2016). Coprostanol and stigmastanol derive from hydrogenation of cholesterol and stigmasterol by bacteria present in the intestines of humans or animals and can indicate human presence and animal husbandry, respectively (Daughton, 2012; Vane et al., 2010). These molecules are also used as chemical indicators of fecal pollution of lakes, rivers, and drinking water (Daughton, 2012; Vane et al., 2010; Wu et al., 2009). In addition, FeSts can originate from vegetation, e.g. β-sitosterol is synthesised by higher vascular plants (Nishimura and Koyama, 1977; Vane et al., 2010) and its derivative β-sitostanol is generated from a reduction reaction in sediments (Martins et al., 2007).

In this study, we ~~reconstructed~~reconstruct fire activity and vegetation changes using a multi-proxy analytical approach applied to lacustrine sediment samples from the south-eastern Tibetan Plateau. This is the first study to combine MAs, PAHs, *n*-alkanes and FeSts analyses into a single analytical method highlighting the ~~interaction~~interactions between fire, climate, and vegetation during the Holocene. This combination of proxies, when synthesized with regional climate records, helps determine the changing role of local and regional fire activity throughout the Holocene.

2 Study Area, Modern Climate, and Holocene Climate History

~~The Qinghai-Tibetan Plateau is a vast plateau in central Asia with an average elevation of approximately 4500 m above sea level (a.s.l.). The TP stretches nearly 1000 km north to south and 2500 km east to west, covering an area of 2 x 10^6 km^2 (Dong et al., 2010). In addition to this wide geographic range, the TP also encompasses altitudes ranging from 1500 to > 8000 m a.s.l., resulting in a broad diversity of landscapes with considerable biodiversity. In general, however, vegetation across much of the TP is dominated by meadow, steppe, and shrubs with increasing species richness with increasing altitude (Shimono et al., 2010). The TP is a pivotal research area due to its sensitivity to century-scale or short-term climatic changes and its influence on global climate (Liu et al., 1998). On the other hand, its remote character limits access to possible paleoclimate archives, resulting in relatively few investigations into species diversity and plant communities (Wang et al., 2006).~~

~~The TP's climate is regulated by the critical and sensitive junction of four climatic systems (image S1 in the Supplementary Information): the Westerlies; the East Asian Monsoon; the Siberian cold polar airflow (or Winter Monsoon); and the Indian Monsoon (Dong et al., 2010). Westerly winds and the Indian Summer Monsoon (ISM) are considered to be the major wind patterns by which atmospheric particulate derived from biomass burning reaches the plateau (Yao et al., 2013). Millennial-scale changes in insolation over the TP affect monsoon variability and the associated moisture reaching the TP. Generally, during periods of increased insolation, the monsoon extended farther north on the TP, resulting in more vegetation. During decreased insolation, colder, drier conditions dominate the TP and regions influenced by the ISM are restricted to more southerly portions of the Plateau, including the study area. During the late Pleistocene (~ 16 cal ky BP), a cold and dry climate resulted in desert-steppe vegetation across much of the TP (Tang et al., 2000). Global paleoclimate studies indicate that this last glacial period concluded with a sudden warming event at ~ 15 cal ky BP (Severinghaus and Brook, 1999). The subsequent transition to the Holocene was characterized by increasing temperature and precipitation that enhanced permafrost and snow melting and facilitated tree growth in the TP after 12 cal ky BP (Saini et al., 2017; Tang et al., 2000). This period was depicted by frequent oscillations between warm and cold phases, in Tibet as well as in other parts of the world (Liping Zhu et al., 2008; Liu et al., 2008, 2009). For example, Tang et al. (2000) suggest that the evolution of ISM has considerably fluctuated throughout the Holocene. Lake Ximencuo (eastern Tibet) sediments record cold events occurring between 10.3 – 10.0, 7.9 – 7.4, 5.9 – 5.5, 4.2 – 2.8, 1.7 – 1.3 and 0.6 – 0.1 cal ky BP, where the cold event at 4.2 cal ky BP had the most substantial impact (Miao et al., 2015; Mischke and Zhang, 2010). Even with these oscillations, the general temperature trends affecting the TP include warm and humid climate in the early to mid-Holocene, as registered in sediments and dust deposits (Liu et al., 2008), and then a cooling trend during the mid-Holocene. The high temperatures during the early Holocene accelerated evaporation and caused many Tibetan lakes to evolve from open freshwater systems to saline lakes (Dong et al., 2010), despite increased~~

~~monsoonal precipitation (Bird et al., 2014). During the mid to late Holocene, warm-wet conditions shifted towards a cooler and drier climate, due to weaker solar insolation, and after 5 cal ky BP temperature and precipitation decreased linearly (Bird et al., 2014; Dong et al., 2010; Liu and Feng, 2012; Tang et al., 2000). More recently, human activities and related climate change have significantly altered the regional hydrology and ecosystem functions of the plateau, with degeneration of vegetation and grassland that led to desertification and frequent dust storms (Wang et al., 2008).~~

~~Paru Co (0.1 km^2) is located in the Nyainqentanglha Mountains (29°47'45.6"N, 92°21'07.2"E; 4845 m a.s.l.; Figs 1a and 1b) and is dammed by moraines from past glaciations in its watershed. The biome surrounding Paru Co is temperate subalpine steppe, where the lake is located near the border of alpine coniferous forest and tropical and seasonal rainforests (Li et al., 2016). The lake's watershed is 2.97 km^2 and consists of a sloping glacial valley measuring 0.5 to 2.0 km with lateral mountain crests higher than 5000 m a.s.l. A central ephemeral stream channel and a second incised channel drain the lake's watershed and feed Paru Co with runoff. Outflow from the lake drains via a small stream channel located approximately 430 m west of the primary outlet (Bird et al., 2014). The Tropical Rainfall Measuring Mission data (TRMM) from 1998 to 2007 show that approximately 92 % of mean annual precipitation (MAP; 1118 mm y^{-1}) at Paru Co occurs between April to September during the ISM season (Fig. 1c, Bird et al., 2014). Previous paleoclimate work at Paru Co (Bird et al., 2014) indicates the occurrence of intense ISM rainfall between 10.1 and 5.2 cal ky BP, when five century-long high lake levels were recorded. The ISM weakened after ~ 5.2 cal ky BP, with the exception of a pluvial event centred at 0.9 cal ky BP. Nir'pa Co, a small lake located near Paru Co, suggests a wet period between 3.3 and 2.4 cal ky and drier conditions from 2.4 to 1.3 cal ky, due to lower silt and lithic content, coincident with elevated sand and clay content and lower lake levels (Bird et al., 2017).~~

~~3 Methods~~

~~3.1 Coring and chronology~~

~~Paru Co core B11 was collected in 2011 and extends from 0 to 435 cm. Seven radiocarbon ages determined by accelerator mass spectrometry (AMS ^{14}C) were measured on seven carbonized grass fragments and one oogonia sample extracted from the surrounding sediments (Bird et al., 2014). The sedimentation rate is approximately 0.35 mm y^{-1} between 10.7 cal ky BP and the present. Between 10.9 to 10.7 cal ky BP sedimentation rates are approximately ten times higher (3.3 mm y^{-1}). The final age-depth model (Fig. 1d) was constructed using a linear regression between 434.9 and 364.1 cm and by fitting a 3rd order polynomial to the AMS ^{14}C, ^{137}Cs (-0.013 cal ky) and sediment-water interface (-0.061 cal ky) ages between 364.1 and 0.0 cm. The associated model error is between 15 and 90 years (see Bird et al. (2014) for further details).~~

The Qinghai-Tibetan Plateau is a vast plateau in central Asia with an average elevation of approximately 4500 m above sea level (asl). The TP stretches nearly 1000 km north to south and 2500 km east to west, covering an area of 2 x 10^6 km^2 (Dong et al., 2010). In addition to this wide geographic range, the TP also encompasses altitudes ranging from 1500 to > 8000 m asl, resulting in highly heterogeneous landscapes with considerable biodiversity. In general, however, vegetation across much of the TP is dominated by meadow, steppe, and shrub communities where species richness increases with increasing altitude (Shimono et al., 2010). The TP is a pivotal research area due to its sensitivity to century-scale or short-term climatic changes and its influence on global climate (Liu et al., 1998). However, its remote nature restricts access to possible paleoclimate studies, resulting in relatively few investigations of past species diversity and plant community changes (Wang et al., 2006).

The TP's climate is regulated by the critical and sensitive junction of four climatic systems (Supplement S1): the Westerlies; the East Asian Monsoon; the Siberian cold polar airflow (or Winter Monsoon); and the Indian Monsoon (Dong et al., 2010). Westerly winds and the Indian Summer Monsoon (ISM) are considered to be the major wind patterns by which atmospheric particulates derived from biomass burning reach the plateau (Yao et al., 2013). Millennial-scale changes in insolation over the TP affect monsoon variability and the associated moisture reaching the TP. Generally, during periods of increased insolation, the monsoon extended farther north on the TP, resulting in more vegetation growth across the plateau. During decreased insolation, colder, drier conditions dominate the TP and regions influenced by the ISM are restricted to more southerly portions of the plateau, including the study area. During the late Pleistocene (~ 16 cal ky BP), a cold and dry climate resulted in desert-steppe vegetation across much of the TP (Tang et al., 2000). Global paleoclimate studies indicate that this last glacial period concluded with a sudden warming event at ~ 15 cal ky BP (Severinghaus and Brook, 1999), in the context of Bølling–Allerød and Younger Dryas events in the region (Liu et al., 2008). The subsequent transition to the Holocene was characterized by increasing temperature and precipitation that enhanced permafrost and snow melting and facilitated tree growth in the TP after 12 cal ky BP (Saini et al., 2017; Tang et al., 2000). This period was depicted by frequent oscillations between warm and cold phases, in Tibet as well as in other parts of the world (Liping Zhu et al., 2008; Liu et al., 2008, 2009). For example, Tang et al. (2000) suggest that the evolution of the ISM has considerably fluctuated throughout the Holocene. Lake Ximencuo (eastern Tibet) sediments record cold events occurring between 10.3–10.0, 7.9–7.4, 5.9–5.5, 4.2–2.8, 1.7–1.3 and 0.6–0.1 cal ky BP, where the cold event at 4.2 cal ky BP had the most substantial impact (Miao et al., 2015; Mischke and Zhang, 2010). Superimposed on these oscillations, the general temperature trends affecting the TP include a warm and humid climate in the early to mid-Holocene, as registered in sediments and dust deposits (Liu et al., 2008), and then a cooling trend during the mid-Holocene. The high temperatures during the early Holocene accelerated evaporation and caused many Tibetan lakes to evolve from open freshwater systems to saline lakes (Dong et al., 2010), despite increased monsoonal precipitation (Bird et al., 2014). TP vegetation also responded to these warmer temperatures with an increase in forests and forest-meadows between 9.2 and 6.3 cal ky BP (Tang et al., 2000). During the mid to late Holocene, warm-wet conditions shifted towards a cooler and drier climate, due to weaker solar insolation, and after 5 cal ky BP temperature and precipitation decreased linearly (Bird et al., 2014; Dong et al., 2010; Liu and Feng, 2012; Tang et al., 2000). More recently, human activities and related climate change have significantly altered the regional hydrology and vegetation distribution of the plateau, with flora degeneration that led to desertification and frequent dust storms (Wang et al., 2008).

Paru Co (0.1 km^2) is located in the Nyainqentanglha Mountains (29º47’45.6"N, 92º21’07.2"E; 4845 m asl; Figs 1a and 1b) and is dammed by moraines from past glaciations in its watershed. The biome surrounding Paru Co is temperate subalpine steppe, where the lake is located near the border of alpine coniferous forest and tropical and seasonal rainforests (Li et al., 2016). The lake’s watershed is 2.97 km^2 and consists of a sloping glacial valley measuring 0.5 to 2.0 km in length with lateral mountain crests higher than 5000 m asl. The maximum water depth of the modern lake is 1.2 m, with gently sloping sides, but may tolerate a total water level of about 3 m. A central ephemeral stream channel and a second incised channel drain the lake’s watershed and feed Paru Co with runoff. Outflow from the lake drains via a small stream channel located approximately 430 m west of the primary outlet (Bird et al., 2014). The Tropical Rainfall Measuring Mission data (TRMM) from 1998 to 2007 show that approximately 92% of mean annual precipitation (MAP; 1118 mm y^{-1}) at Paru Co occurs between April to September during the ISM season (Fig. 1c, Bird et al., 2014). Previous paleoclimate work at Paru Co (Bird et al., 2014) indicates the occurrence of intense ISM rainfall between 10.1 and 5.2 cal ky BP, when five-century-long high lake levels were recorded. The ISM weakened after ~ 5.2 cal ky BP, with the exception of a pluvial event centred at 0.9 cal ky BP. Nir’pa Co, a small lake located near Paru Co, suggests a wet period between 3.3 and 2.4 cal ky and drier conditions from 2.4 to 1.3 cal ky, due to lower silt and lithic content, coincident with elevated sand and clay content and lower lake levels (Bird et al., 2017).

3 Methods

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Paru Co core B11 was collected in 2011 and extends from 0 to 435 cm. Seven radiocarbon ages determined by accelerator mass spectrometry (AMS ^{14}C) were measured on seven carbonized grass fragments extracted from the surrounding sediments (Bird et al., 2014). The sedimentation rate is approximately 0.35 mm y^{-1} between 10.768 cal ky BP and the present. Between 10.937 to 10.789 cal ky BP sedimentation rates are approximately 10 times higher (3.3 mm y^{-1}). The final age-depth model (Fig. 1d) was constructed using a linear regression between 434.9 and 364.1 cm and by fitting a 3rd order polynomial to the AMS ^{14}C, ^{137}Cs (-0.013 cal ky – determined by direct gamma counting) and sediment-water interface (-0.061 cal ky) ages between 364.1 and 0.0 cm. The associated model error is between 15 and 90 years (see Bird et al. (2014) for further details). As the deepest part of the core shows a much higher sedimentation rate that cannot be clearly explained, with the possibility of data distortion, the subsequent description and discussion of the results exclude the samples aging 10.784-10.937 cal ky BP, limiting the dataset interpretation to the period between 1.347 and 10.768 cal ky BP.

3.2 Sample ~~treatments~~preparation

Sub-samples (n = 72) were selected from the core every 5 cm, spanning from 10.9 to 1.3 cal ky BP with a time resolution of about 130 years on average. Unfortunately, the uppermost samples covering the more recent period (1.3 – 0 cal ky BP) have not been processed for this study due to lack of sufficient sample amounts. The samples were sealed in plastic bags and stored at -20 °C, weighed, freeze-dried~~,~~ and ground and reweighed in order to assure ~ 1 g of dry material, allowing the possibility of determining MAs, PAHs, *n*-alkanes, and FeSts from the same sample. All samples were ground using a Mixer Mill MM 400 (Retsch GmbH, Germany) ball miller.

The 72 Paru Co samples were extracted with a 9:1 v/v mixture of ultra-grade (Romil Ltd., Cambridge, UK) dichloromethane and methanol (DCM:MeOH) with Thermo Scientific Dionex ASE 350 (Accelerated Solvent Extractor system), in order to extract both the polar and non-polar compounds. For each extraction, we used 22 mL steel cells containing a 27 mm ø cellulose filter, diatomaceous earth, the sample, ~ 2 g of Na_2SO_4 (to remove residual moisture) and ~ 2 g of activated copper (to remove sulphur that can interfere with PAHs analysis). We added the following internal standard solutions into each cell: 100 µL of ^{13}C labelled levoglucosan at 1 ng μL^{-1} of concentration, 100 µL of hexatriacontane at 40 ng μL^{-1}, 100 µL of a mixture of ^{13}C labelled PAHs (acenaphthylene, phenanthrene~~ and benzo[a]pyrene) at 1 ng μL^{-1}, 100 µL of cholesterol-3,4-$^{13}C_2$ at 1 ng μL^{-1}. The extractions were performed with three static cycles at 100 °C and 1500 psi. A procedural blank was created and extracted for every batch of 12 samples, where we filled the steel cell with all of the same reagents, but without a sample. Each sample was then purified with three steps to obtain a PAHs/*n*-alkanes fraction, a FeSts fraction and a MAs fraction. We combined and modified published clean-up methodologies in order to obtain the necessary fractions (Battistel et al., 2015; Douglas et al., 2012; Kirchgeorg et al., 2014; Martino, 2016)~~, and benzo[a]pyrene) at 1 ng μL^{-1}, 100 µL of cholesterol-3,4-$^{13}C_2$ at 1 ng μL^{-1}. The extractions were performed with three static cycles at 100 °C and 1500 psi. A procedural blank was created and extracted for every batch of 12 samples, where we filled the steel cell with all of the same reagents, but without a sample~~. Our resulting method uses 12 mL Solid-Phase Extraction cartridges (SPE DSC-Si 10 Tube; 12 mL; 52657 Supelco, Sigma-Aldrich) packed with 2 g of silica gel (particle size 50 µm) and installed on Visiprep™ (SPE Vacuum Manifold standard, Sigma-Aldrich) to accelerate purification. We conditioned each cartridge with 30 mL of DCM and 30 mL of Hexane (Hex). The first non-polar fraction (F1), containing PAHs and *n*-alkanes, was eluted using 40 mL of a Hex:DCM 9:1 v/v mixture. Then, the second polar fraction (F2), containing FeSts, was separated with 70 mL of DCM. This fraction was derivatized, according to Battistel et al. (2015), at 70 °C for 1 h with 100 µL of BSTFA + 1% TMCS (N,O-bis(trimethylsilyl)trifluoroacetamide with 1 % trimethylchlorosilane, Sigma-Aldrich) to increase compound volatility and detectability during gas chromatography – mass spectrometry (GC-MS) analysis. Finally, the third polar fraction (F3) containing MAs was eluted with 20 mL of MeOH. F1 and F2 were evaporated under a stream of pure N_2 using a TurboVap II® system (Caliper Life Science, Hopkinton, MA, USA) in order to reduce the volume to 150 µL. F3 was dried, dissolved in 0.5 mL of ultra-pure water and sonicated to avoid any adsorption of MAs to walls of glass evaporation tubes. Finally, the samples were centrifuged (5 min, 14000 rpm) and transferred using decontaminated Pasteur pipettes to the measurement vials~~.

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3.3 Sample analysis

MAs were detected using methods published in ~~Kirchgeorg et al. (2014) with Ion Chromatography~~Kirchgeorg et al. (2014) with ion chromatography (IC Dionex ICS 5000, Thermo Scientific, Waltham, USA) coupled with a single quadrupole ~~Mass Spectrometer~~mass spectrometer (MSQ Plus™, Thermo Scientific) equipped with CarboPac MA1™ column (Thermo Scientific, 2 x 250 mm) and an AminoTrap column (2 x 50 mm), resulting in a good separation of the isomers levoglucosan, mannosan and galactosan. The injection volume was 50 µL. A solution of MeOH/NH_4OH was added post-column (0.025 mL min^{-1}) to improve ionization of the aqueous eluent before entering the electrospray ionisation (ESI) in negative mode. The analytes were quantified according to specific mass to charge ratios, and with calibration curves and response factors containing unlabelled molecules of L, M, G, as well as an internal standard molecule (^{13}C labelled levoglucosan).

~~The seventeen priority PAHs (according to US ATSDR, 1995) plus retene, *n*-alkanes (from C_{10} to C_{35}) and FeSts (coprostanol, epi-coprostanol, cholesterol, 5α-cholestanol, sitosterol, sitostanol) were analysed with Gas Chromatography (6890-N GC system) coupled to a single quadrupole Mass Spectrometer (MS 5975, Agilent Technologies, Santa Clara, CA, USA) (Argiriadis et al., 2014; Battistel et al., 2015; Gregoris et al., 2014; Martino, 2016; Piazza et al., 2013). Each analysis used the same capillary column (HP5-MS (5%-phenyl)-methylpolysiloxane, Agilent Technologies, Santa Clara, CA, USA). The conditions were an injection volume of 2 µL (split valve open after 1.5 min) and He as a carrier gas (1 mL min^{-1}). The MS was equipped with an electronic impact (EI) source used in positive mode. The analytes were quantified in single ion monitoring mode (Table 1). We created response factors containing all of the target compounds as well as internal standards molecules (^{13}C labelled acenaphthylene, phenanthrene and benzo[a]pyrene; hexatriacontane; cholesterol-3,4-$^{13}C_2$). We ran a response factor after every seven samples in order to monitor possible instrumental drift, as well as running a full calibration curve of external PAHs standards before each set of analyses.~~

~~Target molecules with respective analysed ion and method detection limit (MDL) are listed in Table 1. Further method details and quality assurance can be found in previously published works (Argiriadis et al., 2014; Battistel et al., 2015; Gregoris et al., 2014; Kirchgeorg et al., 2014; Martino, 2016; Piazza et al., 2013). Chromatographic peak identification and calculations were performed using the Chromeleon™6.8 Chromatography Data System Software (Thermo Scientific, Waltham, USA) and Agilent G1701DA GC/MSD ChemStation (Agilent Technologies, Santa Clara, CA, USA).~~

The 17 priority PAHs (according to US ATSDR, 1995) plus retene, *n*-alkanes (from C_{10} to C_{35}) and FeSts (coprostanol, epi-coprostanol, cholesterol, 5α-cholestanol, sitosterol, sitostanol) were analysed with gas chromatography (6890-N GC system) coupled to a single quadrupole mass spectrometer (MS 5975, Agilent Technologies, Santa Clara, CA, USA) (Argiriadis et al., 2014; Battistel et al., 2015; Gregoris et al., 2014; Martino, 2016; Piazza et al., 2013). Each analysis used the same capillary column (HP5-MS (5%-phenyl)-methylpolysiloxane, Agilent Technologies, Santa Clara, CA, USA). The conditions were an injection volume of 2 µL (split valve open after 1.5 min) and He as a carrier gas (1 mL min^{-1}). The MS was equipped with an electronic ionization (EI) source used in positive mode. The analytes were quantified in single ion monitoring mode (Table 1). We created response factors containing all of the target compounds as well as internal standards (^{13}C labelled acenaphthylene, phenanthrene and benzo[a]pyrene at 1 ng µL^{-1} - Cambridge Isotope Laboratories, Inc; hexatriacontane at 40 ng µL^{-1} and cholesterol-3,4-$^{13}C_2$ at 1 ng µL^{-1} - Sigma Aldrich). We ran a response factor after every seven samples in order to monitor possible instrumental drift, as well as running a full calibration curve of external PAH standards before each set of analyses.

Target molecules with their respective analysed ions and method detection limits (MDL) are listed in Table 1. Further method details and quality assurance are available in previously published works (Argiriadis et al., 2014; Battistel et al., 2015; Gregoris et al., 2014; Kirchgeorg et al., 2014; Martino, 2016; Piazza et al., 2013). Chromatographic peak identification and calculations were performed using the Chromeleon™6.8 Chromatography Data System Software (Thermo Scientific, Waltham, USA) and Agilent G1701DA GC/MSD ChemStation (Agilent Technologies, Santa Clara, CA, USA).

3.4 Data ~~elaboration~~Analysis

~~Data elaboration and statistics were performed with Microsoft Excel, R and OriginPro 8. All the concentration values obtained from IC and GC-MS analysis were converted in ng g^{-1}, using the dry weight of each sample, and then transformed into fluxes in order to correct the data for the influence of time and sedimentation. Fluxes (ng cm^{-2} y^{-1}) were calculated by multiplying sedimentation rate (cm y^{-1}), wet density (g cm^{-3}) and concentration (ng g^{-1}) of the respective analyte (Menounos, 1997). 2-tailed Pearson's correlations were calculated in R with a 95% confidence interval (chart S3 in the Supplementary Information) with statistically significative results when p-value < 0.05.~~

~~*N*-alkanes ratios useful for work include: the average chain length (ACL), representing the composite of longer and shorter *n*-alkanes (Poynter and Eglinton, 1990) and encompassing the chain length range of 21 to 33; the P-aqueous ratio (P_{aq}), that can help differentiate between submerged plants that tend to have medium-chain-length *n*-alkanes and terrestrial plants that tend~~

~~to have longer chain lengths (Ficken et al., 2000); and the Norm31 ratio (Carr et al., 2014), that specifically examines changes in the distribution of the longest-chain n-alkanes to identify general changes in vegetation types. These ratios were calculated according to the following equations:~~

~~(1)~~ ~~$ACL_{21-33} = \frac{\sum(n_{21-33})(C_{21-33})}{\sum(C_{21-33})}$~~

All concentration values obtained from IC and GC-MS analyses were converted to ng g^{-1} or µg g^{-1}, and then transformed into fluxes in order to correct the data for the influence of time and sedimentation. Fluxes (ng cm^{-2} y^{-1}) were calculated by multiplying the sedimentation rate (cm y^{-1}), wet density (g cm^{-3}) and concentration (ng g^{-1}) of the respective analyte (Menounos, 1997). As explained in Section 3.1, we investigate the data between 1.347 and 10.768 cal ky BP which has a constant sedimentation rate. The concentrations of each analyte therefore have the same trends as their resulting fluxes (Supplement S2). We present all results as concentrations (ng g^{-1} or µg g^{-1}).

N-alkanes ratios useful for our study include: the average chain length (ACL), representing the composite of longer and shorter *n*-alkanes between the chain length range of 21 to 33 and indicating the weighted predominant length (Poynter and Eglinton, 1990); the carbon preference index (CPI), an expression of odd/even predominance that represents how much of the original biological chain length specificity is preserved in geological lipids (Meyers and Ishiwatari, 1993); and the P-aqueous ratio (P_{aq}), that helps differentiate between submerged plants that tend to have medium-chain-length *n*-alkanes and terrestrial plants that tend to have longer chain lengths (Ficken et al., 2000). These ratios were calculated according to the following equations:

(1) $ACL_{21-35} = \frac{\sum(n_{21-35})(C_{21-35})}{\sum(C_{21-35})}$

(2) $CPI_{21-33} = \frac{1}{2}\frac{\sum C_{odd(21-33)}/\sum C_{even(20-32)}}{\sum C_{odd(21-35)}/\sum C_{even(22-34)}}$

~~(2)~~(3) $P_{aq} = (C_{23} + C_{25})/(C_{23} + C_{25} + C_{29} + C_{31})$

~~(3)~~ ~~$Norm31 = C_{31}/(C_{29} + C_{31})$~~

where n_{21-33} indicates the number of carbons in the *n*-~~alkanes~~alkane chains and C_n represents the concentration of the respective *n*-alkane.

~~Charcoal is the most widely used fire proxy, and we therefore compiled regional charcoal~~ In order to help data ~~from the Global Charcoal Database (GCD) as regional syntheses of past biomass burning events~~interpretation, 2-tailed Pearson's correlations were calculated in ~~the TP. The paleofire~~ R ~~library (Blarquez et al., 2014) entails version 3 of the database (Marlon et al., 2016). The range of metrics used to quantify charcoal (e.g., influx, concentration, charcoal/pollen ratios, gravimetric, image analysis, etc.) results in individual data values that vary over 13 orders of magnitude among and within sites, requiring standardizing data~~ for ~~sites (Power et al., 2010). The standardization protocol used for obtaining the charcoal index is described by Marlon et al. (2008) and Power et al. (2010). After selecting latitude and longitude ranges of regional charcoal data, this compilation~~

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~~provides an independent fire history that can be visually compared with the MAs fluxes in the Paru Co core~~the biomarkers dataset with a 95% confidence interval (Supplement S3) with statistically significant results when p-values are < 0.05.

4 Results

4.1 Paleofire indicators

~~Both groups of fire molecular markers in the Paru Co core – MAs and PAHs – demonstrate similar trends in biomass burning activity (Fig. 2a). The MAs record from Paru Co spans from 10.9 to 1.3 cal ky BP, with a major MAs peak in the early Holocene, with a sharp decrease from 8.5 cal ky BP and then a long decreasing trend to 1.3 cal ky BP. This major peak in MAs values for the samples in the period 10.9-10.8 cal ky BP, coincides with the highest sedimentation rates in the core, suggesting that MAs may have been better preserved due to the high quantity of sediment transported into the lake. In order to better visualize MAs trends, the deepest samples were omitted in Fig. 2b, making it possible to identify the fire peaks at approximately 10, 8.6, 5.6 and 2.5 cal ky BP. Over the entire core, MAs fluxes range from 5 to 2500, from 0.7 to 162 and from 0.9 to 531 ng cm^{-2} y^{-1} for levoglucosan, mannosan and galactosan respectively. The Paru Co MAs results reflect the general observation in the literature that mannosan and galactosan concentrations are almost always less than levoglucosan concentrations, which may be due to the different thermal stability of their precursors, hemicellulose and cellulose, respectively (Kuo et al., 2011; Simoneit, 2002). Although levoglucosan and galactosan may have different precursors, their trends throughout the Paru Co core are generally similar, while peaks in mannosan concentrations slightly differ from the other two isomers.~~

~~The fluxes of the total sum of the PAHs congeners mostly vary between 0 and 20 ng cm^{-2} y^{-1} and contain peaks up to 110 ng cm^{-2} y^{-1} (Fig.~~ MA concentrations values span from 29 to 6497, from 15 to 993 and from 17 to 1722 ng g^{-1} for levoglucosan, mannosan and galactosan, respectively. In the most recent sample from 1347 cal ky BP, none of the three MAs were above the detection limits while in a few other samples mannosan and galactosan were below the MDL. The Paru Co MA results reflect the general observation in the literature that mannosan and galactosan concentrations are almost always less than levoglucosan concentrations, which may be due to the different thermal stability of their respective precursors, hemicellulose and cellulose (Kuo et al., 2011; Simoneit, 2002). Although levoglucosan (Fig. 2e) and galactosan may have different precursors, their trends throughout the Paru Co core are generally similar, while peaks in mannosan (Fig. 2f) concentrations differ slightly from the other two isomers. The MA signal is much higher during the early Holocene (10.8-8 cal ky BP) and then slightly increases again during the periods 7-5 and 3-2 cal ky BP.

The lowest PAH value is 0.2 ng g^{-1} of benzo[b]fluoranthene (BbFl) while the highest PAH concentration is 310.3 ng g^{-1}, of naphthalene (Naph). Phenanthrene (Phe), benzo[e]pyrene (BePyr) and Naph respectively represent 20.9%, 18.9% and 17.5% of the total PAH signal in Paru Co (please see Supplement S4 for single PAHs concentrations). The total sum of PAHs (∑PAHs – Fig 2a) shows higher values in the middle Holocene, with major peaks at 6.3, 5.8, 5.2, 4.8, 3.9 and 3.5-3.3 cal ky BP. The general trend shows increases from 2.2 to 1.3 cal ky BP. The molecular weight and/or number of aromatic rings of PAHs allows investigating the influence of different PAH types through time. The group of 3ring-PAHs (Fig. 2b) includes Phe, anthracene (Ant) and fluoranthene (FluA), demonstrating a similar pattern to the ∑PAHs. The group of 4ring-PAHs (Fig. 2c) encompasses pyrene (Pyr), benzo[a]anthracene (BaAnt), chrysene (Chr), retene (Ret), benzo[b]fluoranthene (BbFl) and benzo[k]fluoranthene (Bkfl), which also has higher values during the middle Holocene and then an increasing trend towards 1.3 cal ky BP. The group of 5-6ring-PAHs (Fig. 2d) is composed of benzo[a]pyrene (BaPyr), BePyr, benzo[ghi]perylene (Bghi), Indeno[1,2,3,-c,d]pyrene (IP) and dibenzo[a,h]anthracene (DBahAnt), with a more noisy trend and dissimilar behaviour from the rest of the groups. 5-6ring-PAHs are high in the early Holocene, peaking at 10.3-9.9 cal ky BP, and then have separate high concentrations at 9.3, 8.6, 7.2, 5.2, 3.9, 3.5, 2, and 1.3 cal ky BP.

The ratios of both MAs and PAHs help reconstruct past vegetation and burning sources. MA ratios can help determine past vegetation types and/or burning temperatures. High combustion temperatures (~ 300 °C) and longer combustion durations result in higher L/M and L/(M+G) ratios, regardless of plant species (Kuo et al., 2011).~~2a). Within the 18 analysed congeners of PAHs, naphthalene, fluorene, fluoranthene, phenanthrene and benzo[e]pyrene reach the highest concentrations, touching values till 70 ng cm^{-2} y^{-1}, primarily between 10.9 and 10.7 cal ky BP. These high peaks of fire activity in the oldest part of the record are similar to the fire history recorded by MAs. The other PAHs had low fluxes near or below MDL (image S2 in the Supplementary Information). Plotting only the samples above the deepest and oldest samples, allows observing trends in PAHs that are otherwise obscured by the high peaks in the deepest part of the core (Fig. 2b). The PAHs record differs from the MAs fire history after the initial major peak in biomass burning; MAs steadily decline throughout the Holocene while PAHs do not mirror this long-term trend.~~

~~MAs and PAHs were then plotted together with biogenic silica (BSi) and total organic matter (TOM) data (Bird et al., 2014) to identify if influences from the monsoon or from the association with organic materials, respectively, could have had a role in their distribution in the sediment core (Fig. 3). Pearson's correlation coefficients (chart S3 in the Supplementary Information) were calculated for the periods 10.7-1.3 cal ky BP between MAs and % of BSi and between MAs and % of TOM, with negative results, i.e. r = -0.54 (p-value 3.46*10^{-6}) and r = -0.38 (p-value 0.00175) respectively. This anti-correlation can also be visually observed in Fig. 3 and indicates the non-dependence between MAs quantities and the organic content of the sediments. The TOM only slightly varied displaying a modestly increasing trend across the Holocene. The BSi signal reflects changes in the ISM, with warmer and wetter climate between 11 to 5 cal ky BP followed by cooler and drier conditions (Bird et al., 2014, 2017).~~

~~Throughout the Holocene, the obtained data for ∑PAHs and ∑MAs do not correlate (r = -0.06, p-value 0.665). However, ∑PAHs and ∑MAs positively correlate during the early Holocene (10.9 – 8.7 cal ky BP), with r = 0.51 (p-value 0.029), while in the following period (8.7 – 1.3 cal ky BP) a slight, but not significant, negative correlation is observed, with r = -0.26 (p-~~

~~value 0.08). Considering that the MAs catchment area varies from local to regional scales, the high early Holocene (10.9 – 10.7 cal ky BP) fluxes of MAs and PAHs may be explained with local fire activity, better preservation and higher original concentrations in the sediments. Then, in the period 10.7 – 8.7 both local and regional fire origin may be hypothesised. After 8.7 cal ky BP PAHs may have had a biogenic origin, due to the fact that TOM and PAHs show increasing trends from 8 to 1.3 cal ky BP.~~

~~High percentages of 4-6 ring PAHs generally suggest the contribution of local high-temperature combustion (Yang et al., 2016), where such combustion may be the source of benzo[e]pyrene, the congener with the second highest concentration in Paru Co. PAHs can also serve as local indicators of their sources, where the organic carbon content can be compared to the presence of PAHs in soils (Abdel-shafy and Mansour, 2016). Both PAHs and TOM trends in Paru Co slightly increase from 8.3 to 1.3 cal ky BP (Figs. 3c and 3d). However, this fact is not statistically confirmed (r = 0.26, p-value 0.08), and may be due to the noisy trend in PAHs. In general, the low amounts of PAHs in Paru Co may derive from their partial solubility in water, especially for the lower molecular weight PAHs (Abdel-shafy and Mansour, 2016). The relative proportions of PAHs originating from land areas are generally independent of the sedimentation rate (Zakir Hossain et al., 2013). Therefore, the relative composition of the PAHs could record details of changes in the terrigenous environment surrounding Paru Co (Matsubara Pereira, 2017), but further investigation is needed to explore these results.~~

~~4.2 Vegetation and human indicators~~

~~The ratios of both MAs and *n*-alkanes help reconstruct past vegetation. MAs ratios can help determine past vegetation types and/or burning temperatures. Higher combustion temperatures (~ 300 °C) and longer combustion duration result in higher L/M and L/(M+G) ratios, regardless of plant species (Kuo et al., 2011). The L/M and L/(M+G) ratios in Paru Core range from 0.6 to 100 and 0.5 to 11.1, respectively. The L/M ratios peak between ~ 6 to 7 cal ky BP; the L/(M+G) values do not peak at the same time, but oscillate throughout the Holocene, with the highest values centred around ~ 2 cal ky BP (Figs. 4a and~~ The L/M and L/(M+G) ratios in Paru Core range from 0.6 to 100 and 0.5 to 11.1, respectively (Supplement S5). The L/M ratios peak between ~ 6 and 7 cal ky BP, with the highest value of 98.8 (Fig. 3a). The L/(M+G) values do not peak at the same time, but oscillate throughout the Holocene, with the highest values centred around ~ 2 cal ky BP. Although MA 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.

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. 2-g,h,i), in order to highlight that the influence of error propagation from the original analysis to the ratio values should be carefully investigated (Hughes and Hase, 2010) 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.
~~4b). Although MAs ratios cannot precisely point to the type of past burnt vegetation, they can classify general vegetation categories. According to their published ranges (Fabbri et al., 2009), our data suggest that grasses dominated the area for the oldest section of the Paru Co core and that softwood began to grow in the region after ~ 10.74 cal ky BP. Grasses, softwood and hardwood may have oscillated until 8.6 cal ky BP. Hardwood generally dominated the vegetation between 8.6 to 7.7 cal ky BP, followed by primarily grasslands until the present. Even though MAs ratios can generally differentiate between grass versus wood burning (Kirchgeorg et al., 2014), specific L/M and/or L/(M+G) ratios do not directly correspond to individual fuel types (Matsubara Pereira, 2017) due to the problem of overlapping values and similar burning conditions that influence the ratios.~~

~~Past vegetation changes can also be derived by variations in *n*-alkane ratios, as *n*-alkanes record the organic input into the lake. Rapid fluctuations in ACL_{21-33} values at 10.9 – 10 cal ky BP may reflect quick transitions between terrestrial and aquatic vegetation, while decreased ACL_{21-33} values between 10-5.5 cal ky BP may result from the prevalence of submerged aquatic plants (Fig. 5b), and then prevalence of C_{27}-associated terrestrial vegetation from 5 to 1.3 cal ky BP. The P_{aq} ratio values closer to 1 indicate a greater percentage of submerged plants, and when the value is closer to 0, these numbers pertain to a greater percentage of terrestrial vegetation. The Paru Co P_{aq} ratio (Fig. 5c) quickly oscillates in the oldest section of the core, suggesting rapid changes between terrestrial and aqueous vegetation as the dominant source of *n*-alkanes to the lake, as also supported by ACL_{21-33} data.~~

~~The Paru Co Norm31 ratios demonstrate alternating vegetation types throughout the Holocene (Fig. 5d). High sitostanol fluxes occur at similar core depths as high Norm31 ratios (Fig. 5e), suggesting that the vegetation may be one of the possible sources of sitostanol in the core. The only FeSts that were above the MDL in the Paru Co samples are sitostanol and sitosterol, where these FeSts highly correlate with each other ($r = 0.94$, p-value $3.79*10^{-8}$) due to the fact that these molecules can be produced by plants and reduction reactions in sediments. All other FeSts were either not present and were below MDL in Paru Co samples, suggesting the virtual absence of local humans and grazing animals in the lake catchment throughout the examined time period.~~

~~4.3 GCD results~~

~~The GCD version 3 allows extracting and compiling individual charcoal records into user-defined syntheses using the paleofire R package. Statistical methods to create these compilations are described in detail in Blarquez et al. (2014). The GCD does~~

~~not include charcoal records located in the TP, but does include records from elsewhere in China and Asia. Here, we selected all available charcoal sites with a radius of ~ 1000 km from Paru Co, and that encompass the time period between 0 and 12 cal ky BP, resulting in a total of 43 sites (Fig. 6a). Due to the wide geographic distribution of these sites, these charcoal records are located in a variety of elevations and ecosystems. The catchment area of an individual charcoal record is only a few km, but this synthesis results in charcoal records across 1000s of km. In addition to only traveling a few km, macroscopic charcoal particles (> 100 µm) usually result from burning at temperatures between 250 and 550 °C , while levoglucosan is produced at temperatures centred around 250 °C and can travel up to thousands of kilometres from its source (Schüpbach et al., 2015; Zennaro et al., 2014). When comparing the charcoal and MAs records the differences in catchments may explain much of the dissimilarities in trends over a millennial scale (Fig. 6). Dating uncertainties among the different records can be carried over into the composite charcoal index and can be a source of misinformation. However, some similarities exist in the short-term variability, as highlighted with coloured bars and arrows in Figs. 6b and 6c. The comparison at this level is merely visual, due to the fact that MAs values are presented in fluxes while charcoal is displayed as a standardised index.~~

4.2 Vegetation and human indicators

The variations in *n*-alkane ratios help reconstruct past vegetation changes, as *n*-alkanes record the organic input into and within the lake. The *n*-alkane concentrations oscillate between 0.6 ng g^{-1} (C10) and 321 µg g^{-1} (C25) with C25 as the most abundant (39.8%) followed by C27 (15.8%) and C29 (9.2%). $ACL_{21–35}$ values fluctuate between 24.9 and 27.9, with a general decreasing trend from 10.8 cal ky BP till 7.2 cal ky BP and then an increasing pattern until 1.3 cal ky BP (Fig. 4b). P_{aq} ratios (Fig. 4c) vary between 0.3 and 1 with a trend that is the opposite of $ACL_{21–35}$. CPI_{21-33} demonstrates a general predominance of odds over evens, with values < 1 only occurring in three cases (1.8, 3.2, 8.6 cal ky BP) and where the maximum value of 41.2 happens at 10.1 cal ky BP (Fig 3b).

FeSts contain very low values for the majority of the analysed compounds. Only three FeSts are above the MDL in Paru Co, but these FeSts are not quantifiable in all samples. These FeSts include sitostanol (5α-Sit) that represents 58% of the quantifiable total, sitosterol (Sit) with 37% and cholestenol (5α-Ch) with only 3% of the total. The maximum FeSt concentration throughout the entire core is from 5α-Sit (282 ng g^{-1}) at 2 cal ky BP (Supplement S7). Due to the generally low concentrations, no diagnostic ratios were calculated for the FeSts.

5 Discussion

5.1 Paleofire activity

~~The TP is ringed by high mountains that create natural barriers that block the transport of smoke aerosols to the TP from the south, west, and northwest (You et al., 2016a). However, the Indian Summer Monsoon~~It is not always easy to distinguish the pyrogenic, biogenic and petrogenic sources of PAHs in a specific place because: i) the same compound can be derived from different sources, ii) PAH profiles depend on the combustion temperature, the duration of the process, the flame conditions (oxygen) and the type of organic material (Daly et al., 2007) and iii) once deposited, PAHs undergo transformation processes that depend on the chemical characteristics of the compounds and environmental variables (Cai et al., 2008; Ma et al., 2005; Maliszewska-Kordybach et al., 2009). Taking these conditions into account, we interpret the PAH profiles in Paru Co as fire-related as no evidence of other sources (e.g. volcanic eruptions, anthropogenic emissions) was found.

During the early Holocene (10.8-8.5 ky BP) levoglucosan, galactosan, mannosan and 5rings-PAHs show similar trends, with a general decreasing pattern and some higher peaks at 10.5-10, 9.2 and 8.5 cal ky BP. During the middle Holocene fires are recorded between 6.5-4 cal ky BP by levoglucosan, and more evidently by PAHs spikes. The late Holocene shows increasing PAHs from 3 to 1.3 ky BP and also some peaks in levoglucosan are visible (Figs 2-a,b,c,d,e,f).

The high concentrations of higher molecular weight PAHs during the early Holocene could be explained with local fires of greater combustion temperatures, due to the fact that higher number of rings requires greater burning energy (Denis et al., 2012). High percentages of 4-6 ring PAHs generally suggest the contribution of local high-temperature combustion origins (Yang et al., 2016), where such combustion may be the source of BePyr, the congener with the second highest concentration in Paru Co, but also of IP and Bghi, which are high temperature markers (Zakir Hossain et al., 2013). When fuel sources are uniform, hotter fires (at and above 500 °C) commonly produce high concentrations of BePyr, IP, Bghi (McGrath et al., 2003; Zakir Hossain et al., 2013). The lower, but not lacking, presence of 3rings and 4rings-PAHs could be due to the fact that lower molecular weight PAHs are more depleted due to weathering processes (Zakir Hossain et al., 2013). Their lower concentrations may also be due to the fact that the 3ring and 4ring-PAHs could have travelled farther since they are more volatile than the 5-6 ring PAHs. In addition, the 3ring and 4ring-PAHs may have photochemically degraded in the gas phase after emission to the atmosphere (Wang et al., 2010).

Higher molecular weight PAHs are more stable compounds compared to 3-4 ring 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).

MAs continue to decrease from 8.5 ky BP to 1.5 ky BP whereas 3, 4, and 5 ring-PAHs start increasing before reaching their greatest values between 6.5 and 4 ky BP (Figs 2-b,c,d,e,f). This difference may due to higher percentages of lignin burning (evidenced by retene peaks – Supplement S6) with respect to cellulose burning (represented by MA concentrations). Pollen profiles (Zhao et al., 2011) indicate an increased presence of trees between 7 and 3 ky BP. The combination of low concentrations of the 5-6 ring PAHs but abundant FluA, Pyr and BePyr suggests geographically small, but more frequent wildfires (Zakir Hossain et al., 2013). We interpret the Paru Co record between 6.5 and 4 cal ky BP as the result of such relatively small, but recurrent fires.

The explanation for the lack of levoglucosan and other MA 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 high temperature flaming 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). Increased presence of PAHs may also be due to the sedimentation itself. PAHs derived from pyrogenic sources generally associate with soot-rich particles that protect them from degradation in the atmosphere, water column and sediments (Yunker et al., 2002b). PAHs from forest fires only travel relatively local distances but are protected from photolytic degradation due to their association with larger particles, helping them survive the transport from the atmosphere into climate archives such as sediments (Yunker et al., 2002b).

5.2 Combustion sources

The diagnostic ratios and associated error propagation (Figs 2-g,h,i) do not allow quantitatively assigning PAH sources. IP/(IP+Bghi) contains values above the 0.5 threshold for combustion of wood, wood soot and/or grasses, creosote, as well as almost all wood and coal combustion aerosols and bush fire (Yunker et al., 2002b). The FluA/(FluA+Pyr) ratio, with values above 0.5 for the majority of the samples, indicates the combustion of grass, wood or coal, although this threshold is not definitive (Yunker et al., 2002b). The Ant/(Ant+Phe) ratio with values > 0.10 is generally related to pyrogenic PAH sources, but overlapping values between petroleum and combustion sources are reported (Yunker et al., 2002b). In Paru Co, when including the error propagation, the majority of samples show values of Ant/(Ant+Phe) > 0.10. Due to the improbability that petroleum sources were burned near Paru Co during the geological time period covered by the analysed core, the obtained values for the ratio Ant/(Ant+Phe) must be related to vegetation combustion. In general, Ant undergoes more rapid photochemical reactions in the atmosphere than Phe. In contrast, FluA/Pyr and IP/Bghi isomer pairs degrade at comparable rates and the original composition information is preserved during atmospheric transport (Yunker et al., 2002b) suggesting that their ratios may be more reliable compared to the Ant/(Ant+Phe) ratio. Given these considerations, we confirm that diagnostic ratios are important tools for the source assignment but cannot be completely trusted due to overlapping values and error propagation that may hinder the correct allocation of the signal origin. However, PAHs in Paru Co can function as pyrogenic markers as we did not find any evidence of other sources (e.g. volcanic eruptions, anthropogenic emissions).

The sum of PAH concentrations demonstrates similarities to the eastern TP tree pollen record from the Zoige basin (Fig. 3; Zhao et al., 2011). The Zoige basin is 450 km to the north-east of Paru Co and, in addition to Hidden Lake (Tang et al., 2000), are among the closest pollen records to Paru Co. Both records demonstrate Holocene vegetation fluctuations in the region and are consistent with other pollen and modelling studies (Dallmeyer et al., 2011; Herzschuh et al., 2006; Lu et al., 2011), which ~~may help transport both mineral and organic aerosols over the mountain ridges and into the TP during the summer monsoon months when winds rush from the south across the Himalayas. The ISM is the main source of precipitation across much of the southern TP, where this rainfall provides moisture for plant growth. The strength of the ISM over millennial timescales is driven by solar radiation, where increased insolation results in the ISM moisture moving northward across the TP.~~ identify decreasing summer monsoon precipitation and changes in warm season temperature as the mechanisms responsible for the vegetation shifts from meadow to conifer forest to alpine steppe. The average forest fraction on the TP shrank by almost one-third from the mid-Holocene (41.4%) to the present (28.3 %). Shrubs quadrupled in their mid-Holocene percentage to present-day (12.3 %), replacing much of this forest. The grass fraction also increased from 38.1% during the mid-Holocene to the current percentage of 42.3% (Dallmeyer et al., 2011). This forest decline and replacement by shrubs from 6 cal ky to present is prevalent across much of the south-eastern TP (Lu et al., 2011).

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 (Bird et al., 2014; Tang et al., 2000) 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 (Fig. 2a).~~Climatic records from areas surrounding the TP demonstrate that the Pleistocene-Holocene transition was characterised by increasing temperatures until approximately 8.2 cal ky BP, when sudden cooling occurred (Mischke et al., 2016). The ISM was more intense than current levels between ~ 10 – 6 cal ky BP due to increased insolation, and reached a maximum in the south-eastern TP at 8 cal ky BP (Tang et al., 2000). The mid-Holocene had higher average summer sea surface temperatures (SST) and a stronger summer monsoon than during the present, resulting in warm and wet climate (Wei et al., 2007; Zhao et al., 2011). This timing is consistent with paleo-monsoon records from southern China and with the idea that the interplays between summer insolation and other large-scale boundary conditions, including SST and sea-level change, control regional climate (Zhao et al., 2011). A drying trend during the past 6 cal ky is documented in many records from the northern subtropics and tropics (Liu and Feng, 2012). The cooling trend after the Holocene Climatic Optimum (6.5-4.7 cal ky BP) correlates with decreasing solar insolation (Zhao et al., 2011) and caused a progressive southward shift of the northern hemisphere summer position of the Inter Tropical Convergence Zone, resulting in a decreasing strength of the Asian monsoon~~

~~systems and in a drier climate across much of the TP. Decreased solar insolation resulted in a dramatic drying at ~ 4.2 cal ky BP, directly or indirectly leading to the observed collapses of many Chinese Neolithic cultures (Liu and Feng, 2012; Wang et al., 2005). During the past 750 years, precipitation changes in the Altai controlled fire-regime and vegetation shifts, and the high sensitivity of ecosystems to occasional decadal-scale drought events may, in the future, trigger unprecedented environmental reorganization under global-warming conditions (Eichler et al., 2011).~~

~~The Paru Co fire record demonstrates similar fire histories in the early Holocene for all three proxy types. During the time interval ~ 10.9 – 9.5 cal ky BP, MAs, PAHs and the regional charcoal composite all have elevated fluxes, suggesting increased regional fire activity (Figs. 2 and 6). These observed high levels of fire recorded in Paru Co are consistent with weaker ISM in the period 10.9-10.7 cal ky BP (Bird et al., 2014) and with the idea that dry conditions in Asia before 10 cal ky BP supported biomass burning, higher in the early Holocene than in the late Holocene (Marlon et al., 2013). However, when the monsoon reaches its peak at ~ 8 cal ky BP, the Paru Co fire records and regional charcoal composite substantially differ from one another (Figs. 6b and 6c). The MAs record demonstrates high fire activity between 8 to 9 cal ky BP, while the PAHs and charcoal records demonstrate decreased fire activity during this time period. These differences may be due to factors such as different catchment sizes (as discussed in Section 4.3) as MAs are a regional record, while charcoal provides specific information for a local point, where an individual record may influence the results of a compilation. In addition, many of these charcoal records are located far from Paru Co. However, the PAHs record is from the same Paru Co core as the MAs. The difference between these two records may be influenced by the burning temperatures that produce the different products, where MAs may reflect low temperature fires around ~ 250 °C (Zennaro et al., 2015 and references therein). Although the ISM reached its maximum at ~ 8 cal ky BP (Tang et al., 2000) resulting in relatively wet conditions with the potential to provide more vegetation growth and hence biomass source for fire (Bird et al., 2014; Marlon et al., 2013), within this generally humid period, the abrupt climate event at ~ 8.2 cal ky BP brought generally cold and dry conditions to much of the northern Hemisphere (Alley et al., 1997; Alley and Ágústsdóttir, 2005; Barber et al., 1999). In the Tibetan area, a dry interval at Shumxi Co, associated with the 8.2 cal ky event, is indicated by pollen and diatom records (Van Campo and Gasse, 1993). A peak in aeolian silt in north-western China (Lop Nur, Xinjiang) is also associated with the 8.2 cal ky event (Liu et al., 2003), and correlates with a cold interval in pollen diagrams from Qinghai Lake (Koko-nur) (Alley and Ágústsdóttir, 2005; Liu et al., 2002). The Paru Co fire peaks between 8 – 8.5 cal ky BP may therefore be associated with this cold, dry climate event following a period of enhanced vegetation growth.~~

~~The centennial-scale variability of monsoon precipitations is also characterized by events that correlate with changes in oceanic and atmospheric circulation (Bond et al., 2001; Wang et al., 2005). Recent studies have shown that the climatic change at 5.5 cal ky BP (Bond event 4) was one of the most prominent Holocene climatic events that affected much of the world in the Holocene (Liu and Feng, 2012; Wei et al., 2007). Regional fire history increases during this time period, with elevated MAs~~

~~fluxes in Paru Co, increased regional charcoal, as well as high charcoal concentrations in the south-central TP Nam Co core (Herrmann et al., 2010).~~

~~In general, the Paru Co fire history shows a decreasing trend from 8 cal ky BP to the present, which is consistent with the diminishment of the ISM. This decreasing fire pattern observed in Paru Co may be associated with a regional cooling trend reconstructed from Lake Zigetang with the pollen ratio *Artemisia*/Cyperaceae, a semi-quantitative measure for summer temperature, indicating a general cooling trend throughout the Holocene (Herzschuh et al., 2006). As already mentioned in Section 4.1, the noisy PAHs signal in Paru Co after 8 cal ky BP may not be not fire related but instead may be associated to the biogenic/diagenetic transformation of natural organic matter in the lake (Saber et al., 2006; Stogiannidis and Laane, 2015). Figure 3 shows increasing trends both in PAHs and TOM from 8 to 1.3 cal ky BP, evidencing the possible association of these two variables.~~

~~Regional dust records can provide information regarding past wind speed and direction. However, the transport and source of mineral dust versus the transport and source of organic fire markers may differ, and we would like to highlight that increased dust does not imply increased fire frequency. Dust layers in Genggahai Lake demonstrate weak aeolian activity between 10.3 to 6.3 cal ky BP (Qiang et al., 2014), which may be a response to increased vegetation cover due to the strengthened Asian summer monsoon. In contrast, in central Asia (Lake Zhuyeze, Mischke et al., 2016) the 8.2 cal ky event increased the mobility of aeolian sands which gradually caused the degradation of vegetation because of burial and led to massive and widespread aeolian sand transportation, until ~ 7.5 cal ky BP when the vegetation recovered. Dust layers occurred episodically when the summer monsoon weakened in the periods 6 – 5.5, 4.6 – 4, 1.8 – 1.4 and 0.2 cal ky BP and these abrupt events of sand mobility were associated with enhanced wind strength, probably in response to cooling events at high latitudes (Qiang et al., 2014). This increased aridity, coupled with increased winds, may have influenced the transport of MAs to the analysed lake, as reflected in the fire peak at 5.6 cal ky BP at Paru Co.~~

~~5.2 Past vegetation reconstruction~~

~~Isotopic and pollen information from surrounding lakes support the climatic variation from a cold-dry early Holocene to a warm-humid mid to late Holocene and also ascribe these climate changes to the ISM (Kramer et al., 2010a, 2010b; Ma et al., 2014; Zhu et al., 2010). Pollen assemblages from two transects of lakes (east-west and north-south) across the TP indicate sparse vegetation with low pollen concentrations characterized by *Artemisia*/Cyperaceae alpine steppe (Li et al., 2016). Lake Naleng, also located on the south-eastern TP, records changes that are similar to Paru Co paleoreconstructions (Kramer et al., 2010a). From 10.7 to 4.4 cal ky BP open *Abies*–*Betula* forests reflect intense summer monsoon and an upward treeline shift. Temperature range reconstructions demonstrate climate 2 – 3 °C warmer than present and treeline position 400 – 600 m higher than today. However, within this warm period, the climate had a sudden, intense change between 8.1 and 7.2 cal ky BP with temperatures 1 – 2 °C below early and mid-Holocene levels and forests retreating downslope (Kramer et al., 2010a). Multiple~~

~~pollen studies confirm the severe early Holocene cold events at 8.7 – 8.3 and 7.4 cal ky BP (Miao et al., 2015; Mischke and Zhang, 2010). During the mid-Holocene (7.3 – 4.4 cal ky BP), dense temperate steppe vegetation dominated the TP (Li et al., 2016; Zhao et al., 2011). Tree pollen (primarily *Picea*) peaks during the mid-Holocene at 6.5 cal ky BP, and then decreases until 2 cal ky BP (Zhao et al., 2011). During the same time period, Cyperaceae becomes the dominant regional steppe vegetation, and altitudinal vegetation belts shifted downslope in response to reduced temperatures (Li et al., 2016). These alpine steppes contain desert vegetation elements (a composite of Cyperaceae, Poaceae, Chenopodiaceae, and characteristic high-alpine herb families) between 4.4 – 0 cal ky BP (Herzschuh et al., 2006; Tang et al., 2000). Lake records from Nam Co and Taro Co, located in the same vegetation zone as Paru Co, suggest a weakening in the ISM and the increased influence of the westerlies from 5.6 to 0.9 cal ky BP (Bird et al., 2014; Li et al., 2011; Ma et al., 2014). This synthesis on changes in Holocene vegetation suggests that variations of monsoonal precipitation and insolation-driven temperature are the predominant driving forces for changes in alpine vegetation in the central TP (Li et al., 2016).~~

~~A regional synthesis of pollen records along a south-north transect indicates that climate and vegetation reliance on the monsoon through time is prevalent across the eastern TP (Zhao et al., 2011) suggesting that Paru Co paleoreconstructions may also be considered within this framework. In general, vegetation density and productivity increased during the early to mid-Holocene, as suggested by relatively high pollen concentrations in the Dunde ice cap (northern TP) during ca. 10 – 4.8 cal ky BP, with a limited abrupt to the previously-mentioned dry conditions during 8.0 – 7.7 cal ky BP (Liu et al., 1998). Pollen data are consistent with the ice core oxygen isotope record showing a gradual ^{18}O enrichment throughout the Holocene with a warm period centred at 8 – 6 cal ky BP. Finally, the intervals at 2.7 – 2.2, 1.5 – 0.8 and 0.6 – 0 cal ky BP were comparatively humid periods with higher vegetation density and productivity, with characteristic pollen taxa of alpine meadow (Cyperaceae, *Polygonum*) (Liu et al., 1998). These studies suggest that increased biomass availability provided fuel for fires and was probably the pivotal driver for the kindling of intense fire activity periods in Paru Co.~~

~~A pollen record from the nearby Hidden Lake (Figs. 1 and 4), demonstrating Holocene vegetation fluctuations in the area (Tang et al., 2000), contains similarities with the Paru Co L/M trend in vegetation changes (Figs. 4b and 4c). These records are consistent with the literature and demonstrate that ~ 8 cal ky BP meadows began to be replaced with softwood and then between 5.3 to 3 cal ky BP, these conifers began to be substituted by steppe vegetation (Tang et al., 2000). Data-model comparisons also help depict TP Holocene climatic trends and insert Paru Co vegetation reconstruction in the Tibetan context. Model reconstructions primarily identify decreasing summer monsoon precipitation and changes in warm season temperature as the mechanisms responsible for the vegetation shift (Dallmeyer et al., 2011).~~

In addition to the PAH ratios, L/M ratios can also help determine combustion sources (Fig. 3a). 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). Therefore, the Paru Co data suggest that the fire signal from MAs after 10.74 cal ky BP is likely due to conifer burning in the region. Successively, grasses, softwood and hardwood burning oscillated until 8.6 cal ky BP, where hardwood combustion prevailed until 7.7 cal ky BP, followed by the predominance of grassland burning. Although MA ratios can generally differentiate between grass versus wood burning (Kirchgeorg et al., 2014), specific L/M and/or L/(M+G) ratios do not directly correspond to individual fuel types (Matsubara Pereira, 2017) due to the problem of overlapping ratios and similar burning conditions that influence the ratios.

In order to obtain more information from the burning conditions, we compared CPI values to L/M and PAHs (Fig. 3). 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 C_{23} to C_{35}, 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 the 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, potentially due to the thermal generation of *n*-alkanes of lower CPI from macromolecular material (Schefuss et al., 2003; Standley and Simoneit, 1987). 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). Using these considerations, we assume that when CPI and L/M are parallel to each other, they record both fire from the surrounding areas as well as from near the lake catchment.

5.3 Vegetation in the lake catchment

Past vegetation changes can also be derived from variations in *n*-alkane ratios, as *n*-alkanes can record the organic input into and within the lake. The average chain length (ACL) represents the composite of longer and shorter *n*-alkanes (Poynter and Eglinton, 1990), encompassing the chain length range of 21 to 35. The peak in ACL_{21-35} values at 10.9 – 10 cal ky BP may reflect fewer submerged aquatic plants, while decreased ACL_{21-35} values between 10-5.5 cal ky BP may result from the prevalence of submerged aquatic plants (Fig. 4b). The P-aqueous (P_{aq}) ratio can help differentiate between submerged plants that tend to have medium-chain-length *n*-alkanes and terrestrial plants that tend to have longer chain lengths (Ficken et al., 2000). This ratio is calculated as $(C_{23} + C_{25})/(C_{23} + C_{25}+C_{29}+C_{31})$. When the P_{aq} ratio is closer to 1, these values indicate a greater percentage of submerged plants, and when the value is closer to 0, these numbers pertain to a greater percentage of terrestrial vegetation. The Paru Co P_{aq} ratio (Fig. 4c) rapidly fluctuates in the oldest section of the core, suggesting quick transitions between terrestrial and aqueous vegetation as the dominant source of *n*-alkanes to the lake. The ACL_{21-35} and P_{aq} are highly negatively correlated (r = -0.89; p-value $4.8*10^{-22}$) throughout the Paru Co core demonstrating that both ratios record similar vegetation changes during the same time periods (Fig. 4). Higher ACL ratios and lower P_{aq} values demonstrate higher percentages of terrestrial plants, and vice versa.

Fluctuations in lake levels (Fig. 4d) can be associated with fluctuations in P_{aq}, suggesting a general relationship between higher lake levels and the prevalence of submerged plants between 10 and 5 cal ky BP. The opposite situation occurs between 5 and 1.3 cal ky BP, when a decreasing trend in lake level corresponds to diminishing P_{aq} values. ACL confirms this trend where the majority of values near 25 occur during higher lake levels (10-5 cal ky BP) and the majority of values around 27 occur from 5 to 1.3 cal ky BP. These high lake levels (Fig. 4d) are consistent with wet conditions from a more intense ISM prevailing until ~ 6 cal ky BP, as evidenced by dD wax and pollen records (Figs. 3, 4 and 5).

After 5.2 cal ky BP, lake levels decreased, suggesting diminished ISM rainfall, reduced clastic deposition, leading to an invasion of the littoral zone on the core site and an increase in sand deposition~~The average forest fraction on the TP shrank by almost one-third from the mid-Holocene (41.4%) to the present (28.3 %). Shrubs quadrupled in their mid-Holocene percentage to present-day (12.3 %), replacing much of this forest. The grass fraction also increased from 38.1% during the mid-Holocene to the current percentage of 42.3% (Dallmeyer et al., 2011). This forest decline and replacement by shrubs from 6 cal ky to present is prevalent across much of the south-eastern TP (Lu et al., 2011).~~

~~Paru Co data demonstrate the correlation between ACL_{21-33} and P_{aq}, where prevalence of submerged aquatic plants is associated to lower ACL and higher P_{aq} values. Locally, after 5.2 cal ky BP lake levels decreased probably causing opposite fluctuations in both ACL and P_{aq}, suggesting diminished ISM rainfall, reduced clastic deposition and lowered lake levels, which led to an invasion of the littoral zone on the core site and an increase in sand deposition (Bird et al., 2014). Generally, according to reconstructed changes in summer radiation (Berger and Loutre, 1991), the insolation is suspected to be the major driver for millennial-scale changes in vegetation around Paru Co (Figs. 5a, 5b, 5c). Residues from leaf waxes also help provide additional information on the links between climate, vegetation and fires in the Paru Co region. Modern leaf *n*-alkanes from plants in Qingjiang (Hubei province, China) demonstrate remarkable seasonal variation in their CPI and ACL values (Cui et al., 2008). During warmer months, the CPI values of all plant species decrease gradually due to the fading process of the leaves. The ACL values are greater in mid-summer than in May and November, suggesting that temperature influences these values. Compared to the fresh leaves, the defoliated leaves have an elevated abundance of *n*-alkanes, possibly due to degradation by microorganisms and associated biotransformation (Cui et al., 2008). We can therefore infer that biotransformation of fresh leaves occurs inside the sediments, increasing the quantities of these biomarkers, which were then detected in high abundances in the Paru Co samples. Moreover, because of the fact that the climate of the Nyainqentanglha Mountains is dominated by the ISM and deposition of leaves only occurs at Paru Co during the boreal summer when the lake is ice-free, the interpretation of sedimentological, and perhaps also *n*-alkanes, variability reflects summer climatic changes~~ (Bird et al., 2014). ~~As prolonged charring reduces the average chain length of *n*-alkanes by up to four carbons and creates a balanced odd/even distribution of~~

~~carbons in the leaf waxes (Knicker et al., 2013), seems that *n*-alkanes could help determine fire history. However, significant correlation between *n*-alkanes ratios and MAs ratios is only observed for Norm31 and L/(M+G), with r = 0.36 (p-value 0.009) insinuating a starting point for a more detailed study of the potentiality of MAs ratio.~~The fluctuations in both ACL and P_{aq} are consistent with these lake level changes (Figs. 4 and 5). The mid to late Holocene changes in the lake levels and vegetation respond to decreased summer radiation and associated ISM precipitation (Berger and Loutre, 1991).

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The decline in forest vegetation and the rise in steppe vegetation from 5-4 cal ky BP seems to coincide with an increased human presence on the TP. Grazing indicators (increases in *Rumex*, *Sanguisorba*, and Apiaceae ~~pollens~~pollen), imply a human influence on the environment since approximately 3.4 cal ky BP near the south-eastern TP Lake Naleng (Kramer et al., 2010a), in the ~~south-eastern TP, the general~~ region of Paru Co. Humans slashed and burned the forests near Lhasa to open lands through fire over the past 4600 years (Miehe et al., 2006). ~~Other studies also suggest links between fire activity and forest clearance in the southern and south-eastern TP during the late Holocene (Kaiser et al., 2009a, 2009b).~~Other studies also suggest links between fire activity and forest clearance in the southern and south-eastern TP during the late Holocene (Kaiser et al., 2009a; Kaiser et al.,2009b). Although evidence exists that humans altered TP vegetation through burning in the late Holocene, the extent of human~~ activity on~~-related vegetation change across the TP is still unknown. The absence of anthropogenic FeSts in Paru Co sediments indicates that human and associated pastoralism were not present in the local area. In Paru Co, the only FeSts above the MDL were sitosterol and sitostanol~~.~~ (Supplement S7). Sitosterol can derive from ~~higher~~terrestrial vegetation, ~~but~~ and the presence of its ~~lower amounts respect to the~~derivative molecule sitostanol can indicate the microbial reduction of sitosterol~~ into sitostanol~~ in the stomach of ruminant animals (Vane et al., 2010), as well as ~~its~~sitosterol hydrogenation in sediments (Martins et al., 2007)~~.~~. Sitostanol and sitosterol highly correlate with each other throughout the Paru Co core (r = 0.94, p-value 3.79*10^{-8}). The fact that sitosterol and sitostanol were the only FeSts detected in Paru Co suggests the absence of ruminant animals that would also deposit other FeSts, and we consider vegetation and ~~reduction reactions~~hydrogenation in sediments as the main ~~source~~sources of Paru Co FeSts. Due to the absence of other human/animal indicators, we are inclined to describe the variations~~ found~~ in fire regimes and vegetation as primarily climate-driven signals.

Isotopic and pollen information from surrounding lakes, support the climatic variation from a warm-humid early Holocene to a cold-dry mid to late Holocene and also ascribe these climate changes to the ISM (Kramer et al., 2010a, 2010b; Ma et al., 2014; Zhu et al., 2010). Pollen assemblages from two transects of lakes (east-west and north-south) across the TP indicate sparse vegetation with low pollen concentrations characterized by *Artemisia*/Cyperaceae alpine steppe (Li et al., 2016). Lake Naleng, also located on the south-eastern TP, records changes that are similar to Paru Co paleoreconstructions (Kramer et al., 2010a). From 10.7 to 4.4 cal ky BP, open *Abies–Betula* forests reflect intense summer monsoon and an upward treeline shift. Temperature range reconstructions demonstrate climate 2-3 °C warmer than present and treeline position 400–600 m higher than today. However, within this warm period, the climate had a sudden, intense change between 8.1 and 7.2 cal ky BP with temperatures 1-2 °C below early and mid-Holocene levels and forests retreating downslope (Kramer et al., 2010a). Multiple pollen studies using compilations of Chenopodiaceae, Asteraceae, Cistaceae, Tamaricaceae, and Pottiaceae confirm the severe early Holocene cold events at 8.7–8.3 and 7.4 cal ky BP (Miao et al., 2015; Mischke and Zhang, 2010). During the mid-Holocene (7.3–4.4 cal ky BP), dense temperate steppe vegetation dominated the TP (Li et al., 2016; Zhao et al., 2011). Tree pollen (primarily *Picea*) peaks during the mid-Holocene at 6.5 cal ky BP, and then decreases until 2 cal ky BP (Zhao et al., 2011). During this same time period, Cyperaceae becomes the dominant regional steppe vegetation, and altitudinal vegetation belts shifted downslope in response to reduced temperatures (Li et al., 2016). These alpine steppes contain arid vegetation elements (including Cyperaceae, Poaceae (grass family), Amaranthaceae (pigweed and amaranths), and characteristic high-alpine herb families) between 4.4-0 cal ky BP (Herzschuh et al., 2006; Tang et al., 2000). Lake records from Nam Co and Taro Co, located in the same vegetation zone as Paru Co, suggest a weakening in the ISM and the increased influence of the westerlies from 5.6 to 0.9 cal ky BP (Bird et al., 2014; Li et al., 2011; Ma et al., 2014). This synthesis on changes in Holocene vegetation suggests that variations of monsoonal precipitation and insolation-driven temperature are the predominant driving forces for changes in alpine vegetation in the central TP (Li et al., 2016).

5.4 Atmospheric transport

The TP is ringed by high mountains that create natural barriers that block the transport of smoke aerosols to the TP from the south, west, and northwest (You et al., 2016a). However, the ISM system may help transport both mineral and organic aerosols over the mountain ridges and into the TP during the summer monsoon months when winds rush from the south across the Himalayas. The ISM is the main source of precipitation across much of the southern TP, where this rainfall provides moisture for plant growth. The strength of the ISM over millennial timescales is driven by solar radiation, where increased insolation results in the ISM moisture moving northward across the TP. ~~The differences in trends between the *n*-alkanes vegetation data, pollen records and MAs ratios may be due to their differing provenance. MAs, in general, record regional fire, except when associated to PAHs, where local biomass burning can be detected. Conversely, *n*-alkanes are local indicators and can also originate from living plants. The *n*-alkanes may differ from pollen as angiosperms produce more *n*-alkanes than do gymnosperms. This discrepancy is the probable reason why *n*-alkanes demonstrate a different grass/wood prevalence than that recorded by MAs ratios and pollens (Figs. 4 and 5). MAs and pollens are able to be transported hundreds to thousands of kilometres, and so primarily reflect regional environmental modifications. Conversely, *n*-alkanes register that vegetation changes in Paru Co may be related to local-scale lake levels fluctuations and to global-scale solar radiation (Fig. 5). Moreover, sitostanol is associated to local degradation of sitosterol derived from higher plants present near the lake. FeSts are markers of local human and animal presence as well, and their absence in Paru Co exclude the anthropogenic influence on the lake environment. Comparing data resulted from this multi-proxy study is helpful to understand past environmental processes happened within the lake and the south-eastern Tibetan Plateau, highlighting how diverse fire and vegetation markers are needed to obtain both the local and the regional paleoreconstruction information, using the same sediment core.~~

Climatic records from areas surrounding the TP demonstrate that the Pleistocene-Holocene transition was characterised by increasing temperatures until approximately 8.2 cal ky BP, when a sudden cooling occurred (Mischke et al., 2016). The ISM was more intense than current levels between ca 10-6 cal ky BP due to increased insolation, and reached a maximum in the south-eastern TP at 8 cal ky BP (Tang et al., 2000).

The mid-Holocene (~6 cal ky BP) had higher average summer sea surface temperatures (SST) and a stronger summer monsoon than during the present, resulting in a warm and wet climate (Wei et al., 2007; Zhao et al., 2011). This timing is consistent with paleo-monsoon records from southern China and with the idea that the interactions between summer insolation and other large-scale boundary conditions, including SST and sea-level change, control regional climate (Zhao et al., 2011). A drying trend during the past 6 cal ky is documented in many records from the northern subtropics and tropics (Liu and Feng, 2012). 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. Decreased solar insolation resulted in a dramatic drying at ~ 4.2 cal ky BP, directly or indirectly leading to the observed collapses of many Chinese Neolithic cultures (Liu and Feng, 2012; Wang et al., 2005). During the past 750 years, precipitation changes have influenced fire-regimes and vegetation shifts in the Altai, where ecosystems are highly sensitive to occasional decadal-scale drought events which, in the future, may trigger unprecedented environmental reorganization under global-warming conditions (Eichler et al., 2011).

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 (Section 5.1), 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 (Fig. 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.

Even though modern transport is not indicative of atmospheric circulation in the early Holocene, recent studies can depict the distribution of air masses affecting the southern Tibetan Plateau under current monsoon conditions, and demonstrate the geographic reach of possible source areas. Modern transport patterns demonstrate that air masses over Ranwu, a sampling site in the Tibetan Plateau, 450 km east of Paru Co (Wang et al., 2016), mostly arrive from Bangladesh and the Indo-Gangetic Plain during both the winter (64.3%) and spring (70.2%) seasons (Wang et al., 2016). The remaining fraction of air masses in winter (35.7%) come from the Middle East, Afghanistan, Pakistan, and northwest India, while the spring air masses largely derive from northwest India (26.3%). Winter air masses over Beiluhe, a sampling site 600 km north of Paru Co, in the central Tibetan Plateau, come mostly from the southern slope of the Himalayas (79.6%) with the remaining air masses originating in the Middle East and Central Asia (20.4%). However, spring air masses largely originate from north-western China (45.9%), followed by Central Asia (32.4%) as well as the southern slope of the Himalayas (21.6%). The Paru Co levoglucosan record therefore encompasses possible source regions that may extend beyond the TP.

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, and 5.2-3.2 cal ky B (Fig. 4), as more intense rainfall results in greater lithic deposition (Bird et al., 2014). 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 cal ky BP 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).

The late-Holocene PAHs fire signal in Paru Co is consistent with charcoal records demonstrating an increasing fire trend in the eastern monsoonal region of China during this same time period (Xue et al., 2018). The increasing fires in Paru Co and surrounding areas are synchronous with the general change in the regional fire pattern during the late Holocene, coincident with increases in population and crop areas (Marlon et al., 2013). Although human FeSts were absent in Paru Co, FeSts are a very local indicator of the presence of humans, and anthropogenic activity could still influence fire records across regional scales. For example, Miao et al. (2017) link NE Tibet Bronze Age sites and associated human activity to increasing charcoal concentrations from 3.6 cal ky BP to the present.

6 Conclusions

This study is the first multi-proxy work ~~on~~of paleofire activity in lacustrine sediments from the ~~TP ,~~Tibetan Plateau (TP) and provides a starting point for future investigations~~ in this field of research.~~. The combination of MAs, PAHs, FeSts, and *n*-alkanes as fire and vegetation ~~markers, helps~~biomarkers help reconstruct the biomass burning history of the south-eastern ~~Tibetan Plateau using innovative biomarkers. The results reveal intense climate-induced fire activity in the period 10.9-8 cal ky BP and then a long-term decreasing trend in fire.~~TP. Vegetation reconstructed from MA and *n*-~~alkanes~~alkane ratios was characterised by short-term oscillations ~~and~~ alternating ~~softwood~~between conifer and ~~grasses~~grass/steppe ~~vegetation composition, and~~ communities influenced by a long-term pattern ~~due to~~of orbital-induced insolation changes. The apparent absence of human impact indicators as determined by the lack of human FeSts above the MDL~~,~~ excludes local anthropogenic influence on fire and vegetation changes. ~~Fire~~

The fire and vegetation records in Paru Co are instead primarily driven by climatic factors as follows:

~~1)~~ **Early Holocene: 10.~~9 –~~7-7.5 cal ky BP.** The period ~~10.9 – 7.5 cal ky BP~~ was characterised by an increasingly ~~warming and drying~~warm climate followed by an intensification phase of the ISM until the mid-Holocene. These conditions may have favoured vegetation growth ~~that led to fire activity recorded in Paru Co by both MAs and PAHs, with major peaks of fire activity around 10.9 – 9.5 cal ky BP. Regional charcoal compilations also depict increased biomass burning from 11 to 9.5 cal ky BP. However, the charcoal records cannot be related to the increased fire activity recorded in Paru Co between 9 to 8 cal ky BP. This difference suggests a relatively local fire source for the Paru Co biomass burning, as the closest available charcoal records are still hundreds of kilometres away. The decrease in fire activity may have been affected by the dramatic dry event of ~ 8 cal ky BP.~~. Early Holocene ~~MAs ratios and~~ pollen records indicate meadows as the prevalent regional vegetation in the surroundings of Paru Co, where *n*-~~alkanes~~alkane ratios depict ~~vegetation~~ oscillations between grass and conifer communities during much of this time period~~, as well as submerged/emergent vegetation alternation.~~

- ~~2) Mid-Holocene: 7.5 – 3.8 cal ky BP. Increased forest vegetation cover due to the strengthened ISM is observed in Paru Co, yet the 5.6 cal ky BP fire event is the unique peak recorded in this core in the mid-Holocene. This ~ 5.6 cal ky BP fire peak~~. The predominance of high molecular weight PAHs suggests high-temperature local fires during this time period. Levoglucosan also peaks during the early Holocene, suggesting that smouldering fires also ~~occurs in the charcoal composite record too, possibly indicating a~~ occurred, although these fires may be regional ~~fire source, as it can be likewise associated with enhanced aeolian activity most likely related to drying and cooling events.~~rather than local. The presence of both high- and low-temperature fires is consistent with the grassland and conifer forests suggested by the *n*-alkane ratios.
- **Mid-Holocene: 7.5-3.8 cal ky BP**. The strong ISM during the early mid-Holocene increased forest vegetation near Paru Co. The peak in L/M suggests sustained fires at temperatures centred around ~300° C, which is consistent with conifer fires. The forest cover decreases beginning ~5 cal ky BP, never regaining its dominance. As the ISM decreased in strength, PAHs record intense fires, which may be due to the combination of the initial vegetation growth followed by an extended dry period. Cold and dry conditions ~~reach the~~reached their maximum at ~~circa~~ ~4.2 cal ky BP, coinciding with the expansion of ~~Bronze Age~~local civilizations, whose eventual impact on the landscape is still unknown. However, the lack of quantifiable FeSts in the Paru Co core suggests that local populations did not live within the lake's watershed during this time period.
- ~~3)~~ **Late Holocene: 3.8~~ –~~ -1.3 cal ky BP.** ~~The past few thousand years were characterized by low fire activity in Paru Co, with some weak exceptions around 3 – 2 cal ky. Emergent terrestrial plants, steppe and grasses~~Grassy steppes dominated the vegetation ~~at this site, in correspondence with~~near Paru Co, due to the weaker summer insolation, cooler conditions, and ~~the~~ reduced precipitation. Although grassland dominated the region, the MAs demonstrate that relatively few smouldering fires occurred during the late Holocene. However, all combustion PAHs, including the high-temperature

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PAHs, demonstrate a steady increase throughout the late Holocene even though the climate and vegetation types remained stable. This PAHs trend suggests a net increase in local to regional combustion that is separate from vegetation and climate change.

Data availability. The full dataset is available in the Supplementary Information and can be accessed at the NOAA/World Data Service for Paleoclimatology archive (https://www.ncdc.noaa.gov/paleo-search/study/24410). Additional data used in this study were retrieved from Berger and Loutre (1991), Bird et al. (2014) - https://www.ncdc.noaa.gov/paleo/study/16399, Huffman et al. (2009) and Zhao et al. (2011).

Competing interests. The authors declare that they have no conflict of interest.

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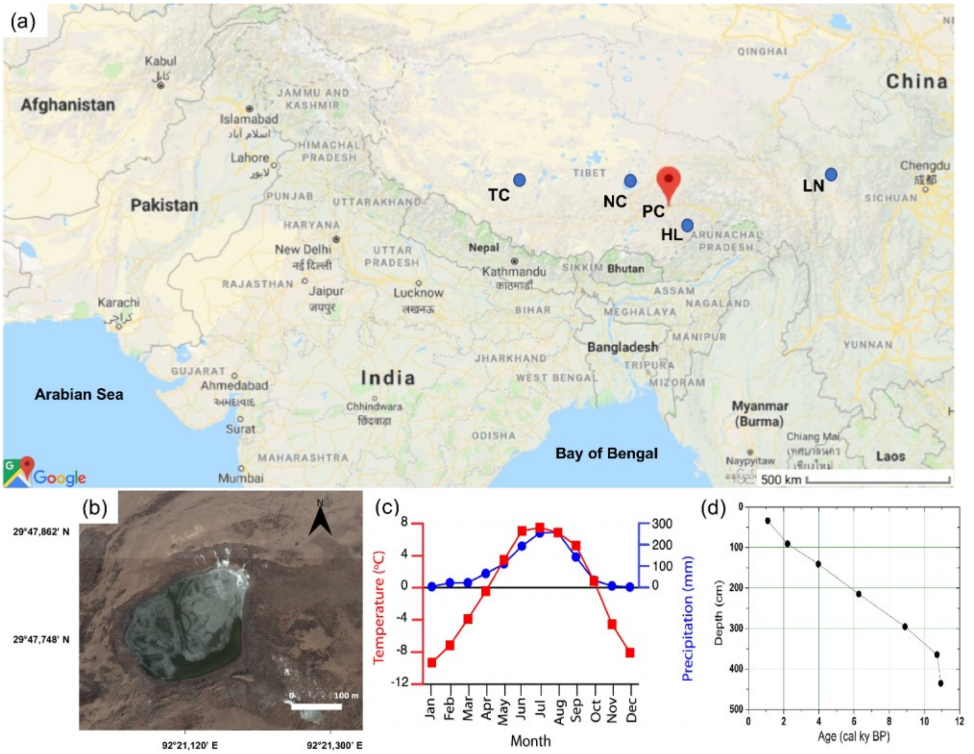(a)
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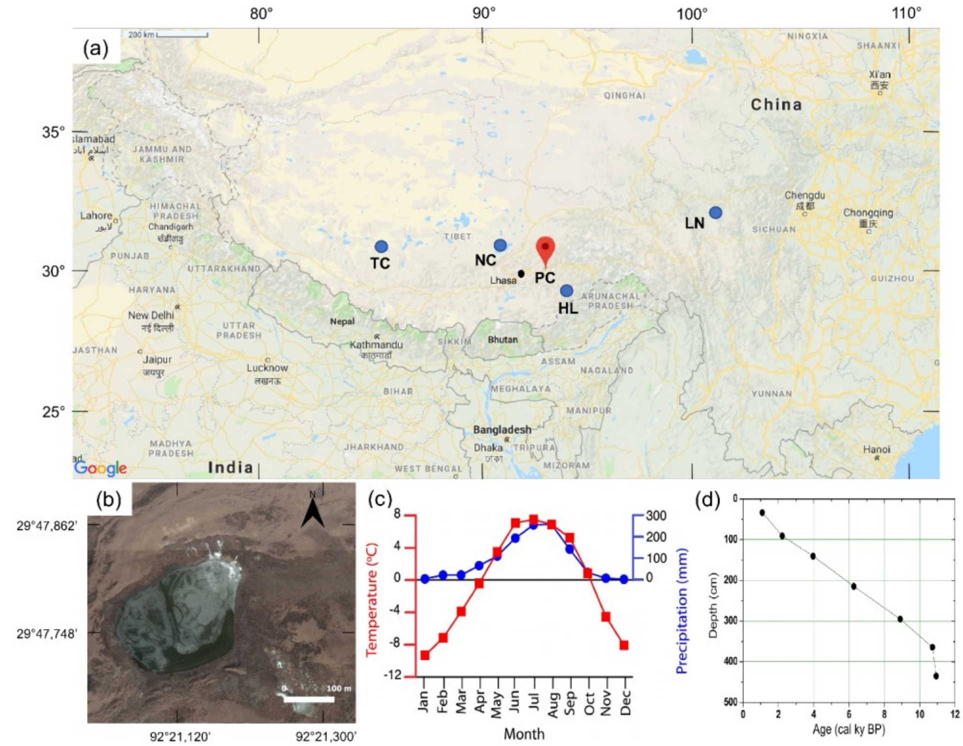
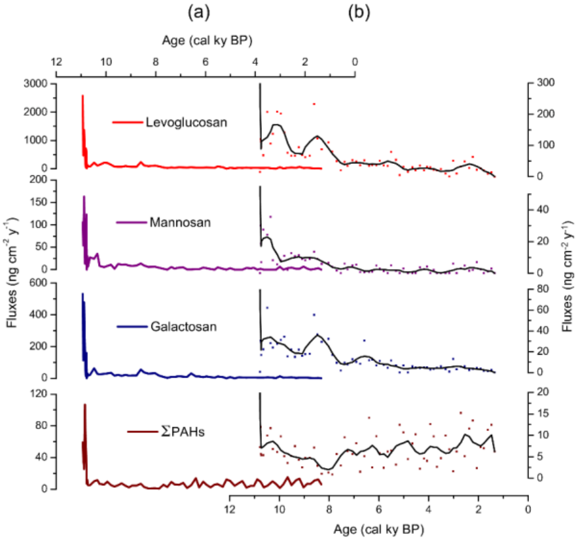
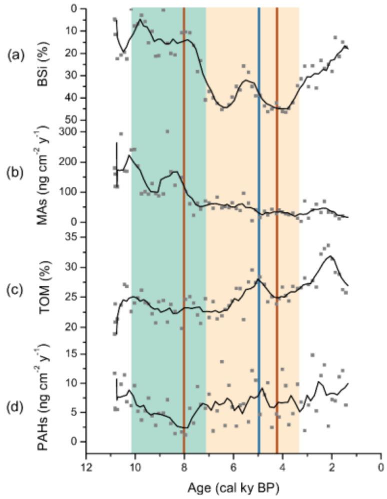


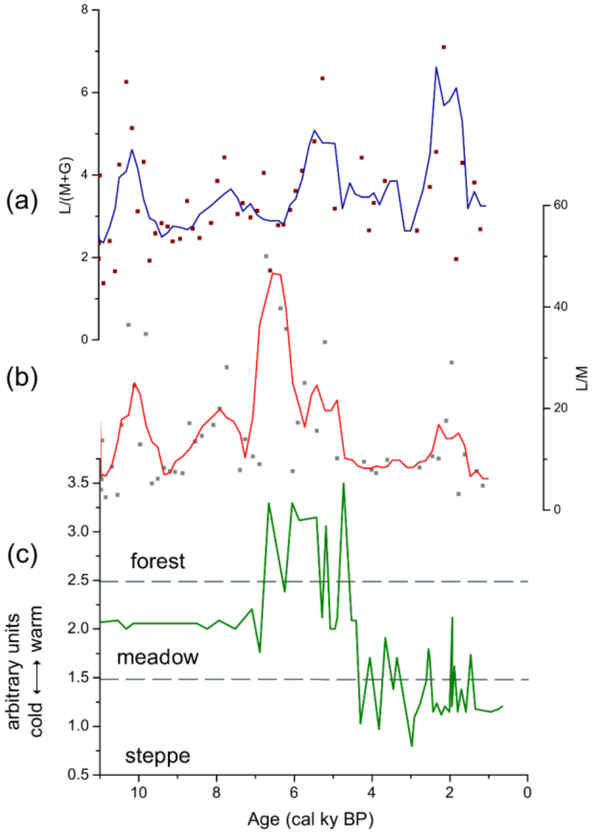
Figure 1~~: (a) Map of the Tibetan Plateau and surrounding territories showing the location of Paru Co (red pin) and of the other lakes mentioned in the text (blue circles): Taro Co (TC), Nam Co (NC), Hidden Lake (HL) and Lake Naleng (LN). (b) Satellite image of Paru Co. (c) Average monthly precipitation at Paru Co based on TRMM data from 1998 to 2007 and average monthly temperatures at Paru Co (4845 m asl) from Lhasa (3650 m asl) weather station data using a lapse rate of – 6.4 °C km^{-1}. (d) Plot of the age/depth model for Paru Co according to Bird et al. (2014).~~



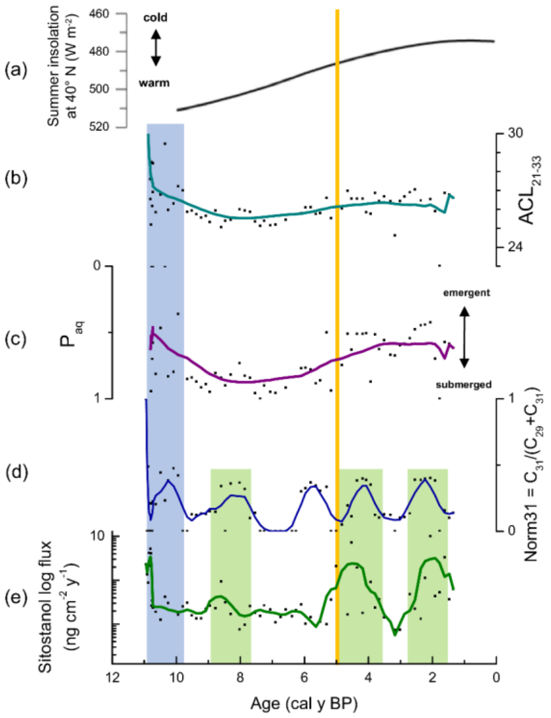
~~**Figure 2:** Fluxes of levoglucosan, mannosan, galactosan and PAHs in Paru Co (a) along the whole core and (b) zooming in after removing the deepest samples older than 10.7 cal ky BP (scatters and trends, obtained with 5-points weighted-moving averages).~~



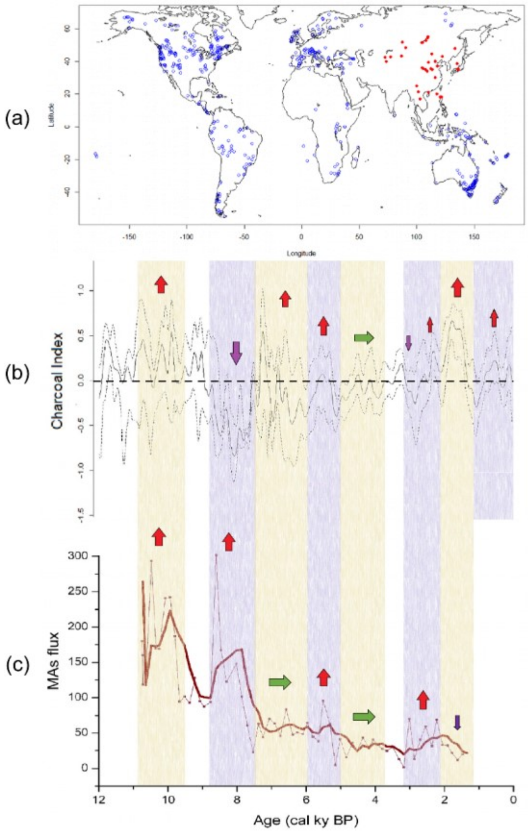
~~**Figure 3:** Results from Paru Co showing data and trends (5-points moving averages) (a) % BSi; (b) ΣMAs; (c) % TOM; (d) ΣPAHs. Blue box: ISM rainfall increased from 10.1 to 7.1 cal ky BP. Peach box: decrease of ISM rainfall to a minimum between 7.1 and 3.4 cal ky BP. Red lines: Bond events 5 (~ 8 cal ky) and 3 (4.2 cal ky). Blue line: division between warmer/wetter ISM and successive cooler/drier conditions.~~



~~**Figure 4:** (a) Paru Co L/(M+G); (b) Paru Co L/M (trends obtained with a 5-points moving average); (c) Hidden Lake pollen inferred vegetation, data graphically obtained with GetData Graph Digitizer from Tang et al., (2000).~~



~~**Figure 5:** Comparison between vegetation indicators in Paru Co and association with the solar summer radiation. (a) summer insolation at 40° N (Berger and Loutre, 1991); (b) average chain length *n*-alkanes ratio; (c) P_{aq} *n*-alkanes ratio; (d) Norm31 *n*-alkanes ratio; (e) Sitostanol log fluxes detected in Paru Co. Trends obtained with 5-points (Norm31 and sitostanol) and 10-points (ACL_{21-33} and P_{aq}) weighted moving averages. Gold line: division between warmer/wetter ISM and successive cooler/drier conditions. Blue bar: early Holocene variability of the proxies. Green bars: association between sitostanol and Norm31 oscillations.~~



~~**Figure 6:** (a) Map indicating the 43 sites (red dots) used for the charcoal index elaboration; (b) charcoal index calculated with the GCD; (c) MAs (ng cm^{-2} y^{-1}) data points and trend (5-points moving average) resulted from Paru Co analysis.~~

: (a) Map of the Tibetan Plateau and surrounding territories showing the location of Paru Co (red pin) and of the other lakes mentioned in the text (blue circles): Taro Co (TC), Nam Co (NC), Hidden Lake (HL) and Lake Naleng (LN). (b) Satellite image of Paru Co. (c) Average monthly precipitation at Paru Co based on TRMM data from 1998 to 2007 and average monthly temperatures at Paru Co (4845 m asl) from Lhasa (3650 m asl) weather station data using a lapse rate of −6.4 °C km^{-1} (Huffman et al., 2009). (d) Plot of the age/depth model for Paru Co according to Bird et al. (2014).

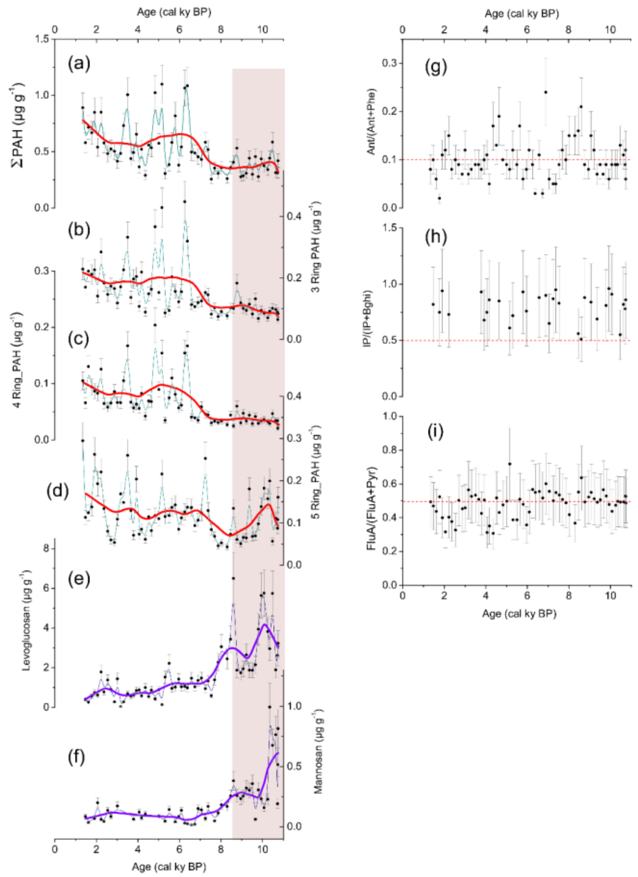
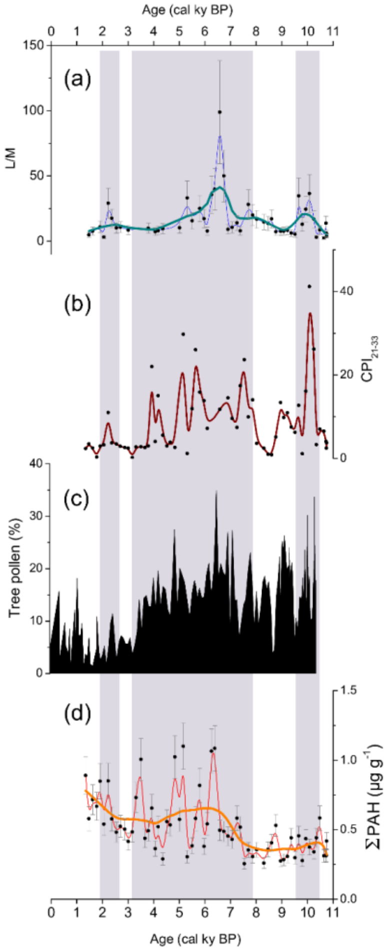


Figure 2: (a) Sum of PAHs concentrations; (b) Sum of 3 ring PAH concentrations (Phe, Ant, FluA); (c) Sum of 4 ring PAHs concentrations (Pyr, BaAnt, Chr, Ret, BbFl, Bkfl); (d) Sum of 5-6 ring PAHs concentrations (BaPyr, BePyr, Bghi, IP, DBahAnt). Data points (black) with absolute error range (grey), LOWESS smoothing with SPAN parameter 0.2 (red), b-spline interpolation (cyano). (e) Levoglucosan concentration; (f) Mannosan concentration. Data points (black) with absolute error range (grey), LOWESS smoothing with SPAN parameter 0.2 (purple), b-spline interpolation (dark blue). Pink bar indicates the early Holocene period where levoglucosan and 5rings PAHs show high concentrations. (g) Ant/(Ant+Phe); (h) IP/(IP+Bghi); (i) FluA/(FluA+Pyr). Ratios values (black points) with absolute error bars (grey) and diagnostic thresholds (red dashed lines).



5 **Figure 3: (a) L/M ratio values (black points) with absolute error bars (grey); LOWESS smoothing with SPAN parameter 0.2 (teal), b-spline interpolation (blue). (b) CPI ratio values (black points); b-spline interpolation (dark red). (c) Tree pollen (%) from Zhao et al., 2011 - http://apps.neotomadb.org/Explorer/?datasetid=14619. (d) Sum of PAH concentrations, data points (black) with absolute error range (grey), LOWESS smoothing with SPAN parameter 0.2 (orange), b-spline interpolation (red).**

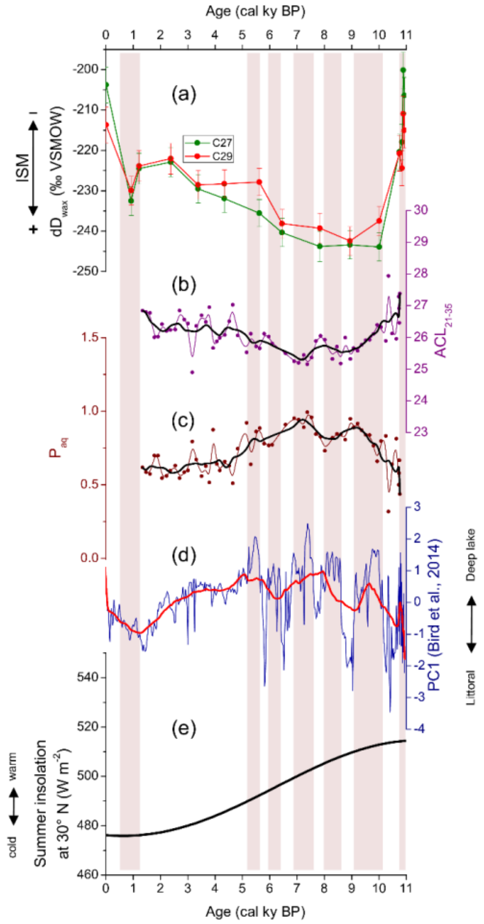


Figure 4: The response of Paru Co aqueous vegetation to changing summer insolation and associated monsoon intensity. (a) δD wax for C27 and C29 n-alkanes referenced to Vienna Standard Mean Ocean Water scale, data from Bird et al., 2014 - https://www.ncdc.noaa.gov/paleo/study/16399. (b) ACL ratio values (purple points), adjacent-average smoothing with 5 points (black), b-spline interpolation (purple line). (c) Paq ratio values (brown points), adjacent-average smoothing with 5 points (black), b-spline interpolation (brown line). (d) Principal component 1 values (blue) as indicative of lake level changes, adjacent-average

smoothing with 40 points (red), data from Bird et al., 2014 - https://www.ncdc.noaa.gov/paleo/study/16399 (e) Summer insolation, data from Berger and Loutre (1991).

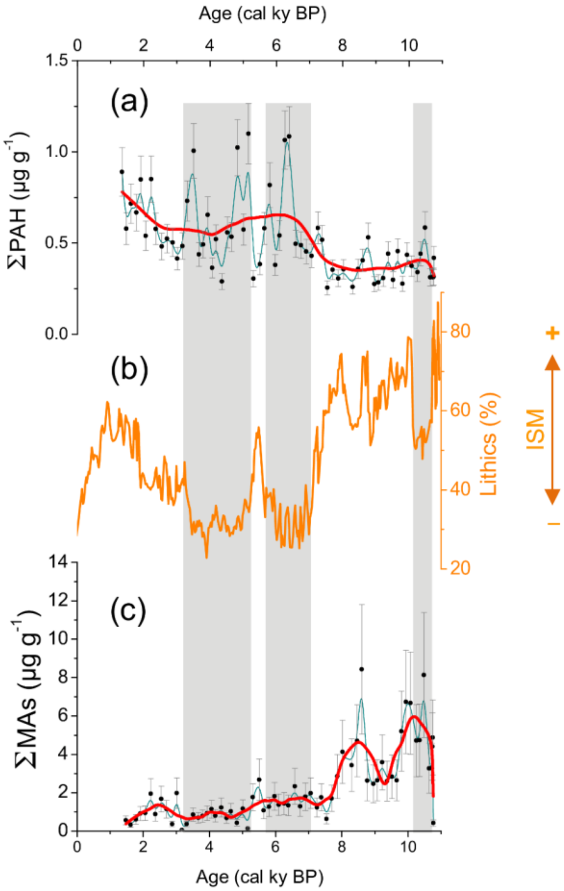


Figure 5: The response of combustion proxies to changes in ISM intensity in Paru Co. (a) Sum of PAHs concentrations, data points (black) with absolute error range (grey), LOWESS smoothing with SPAN parameter 0.2 (red), b-spline interpolation (teal). (b) lithics (%), data from Bird et al. (2014) - https://www.ncdc.noaa.gov/paleo/study/16399. (c) MAs concentrations, data points (black) with absolute error range (grey), LOWESS smoothing with SPAN parameter 0.2 (red), b-spline interpolation (teal).

Table 1: Target molecules with their abbreviations, detected mass to charge ratio (m/z) and method detection limit (ng) calculated as blank values plus three standard deviations or with the signal to noise ratio when no analytes were detectable in the blanks.

Molecular classes	Compounds	Abbreviation	Targeted ion (m/z)	MDL (ng)
MAs	Levoglucosan	L	161	~~226~~26.1
	Mannosan	M	161	12.2
	Galactosan	G	161	9.8
PAHs	Naphthalene	Naph	128	~~16.0~~5
	Acenaphthylene	Acy	152	~~8.4~~800 pg
	Acenaphthene	Ace	154	~~17.8~~1.2
	Fluorene	Flu	166	~~8.7~~3.3
	Phenanthrene	Phe	178	~~21.4~~8
	Anthracene	Ant	178	~~19.3~~750 pg
	Fluoranthene	~~Fluo~~FluA	202	~~11.1~~2.4
	Pyrene	Pyr	202	~~19.4~~2.8
	Benzo(a)anthracene	BaAnt	228	~~0~~50 pg
	Chrysene	Chr	228	~~0~~250 pg
	Retene	Ret	~~234~~219	~~0~~250 pg
	Benzo(b)fluoranthene	BbFl	252	~~0~~80 pg
	Benzo(k)fluoranthene	BkFl	252	~~0~~80 pg
	Benzo(a)pyrene	BaPyr	252	~~15.8~~700 pg
	Benzo(e)pyrene	BePyr	252	~~0~~860 pg
	Benzo(ghi)perylene	~~BghiPer~~Bghi	276	~~0~~300 pg
	Indeno(1,2,3-c,d)pyrene	~~IPyr~~IP	276	~~0~~350 pg
	Dibenzo(a,h)anthracene	DBahAnt	278	~~0~~800 pg
***n*-alkanes**	C_{10}-C_{35}	C_{10}-C_{35}	71	~~4735.0~~500
FeSts	Coprostanol	Cop	215	~~0~~50 pg
	Epicoprostanol	e-Cop	215	~~0~~50 pg
	Cholesterol	Chl	370	100.4
	Cholestanol	5α-Ch	355	~~0~~50 pg
	Sitostanol	5α-Sit	215	0.5

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Sitosterol	Sit	215/396	2.3