

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

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Anonymous Referee #3 General Comments: In this manuscript, Callegaro 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 bet-

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ter interpretation of data, as well as better figures and presentation of data. The data and the author's interpretation seemed to conflict with each other, 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).

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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 $\text{Ant}/(\text{Ant}+\text{Phe})$, $\text{IP}/(\text{IP}+\text{Bghi})$ and $\text{FluA}/(\text{FluA}+\text{Pyr})$. 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 $\text{Ant}/(\text{Ant}+\text{Phe})$, $\text{IP}/(\text{IP}+\text{Bghi})$ and $\text{FluA}/(\text{FluA}+\text{Pyr})$. $\text{Ant}/(\text{Ant}+\text{Phe})$ values generally discriminate between petroleum (< 0.10) and combustion (> 0.10) sources; $\text{IP}/(\text{IP}+\text{Bghi})$ 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; $\text{FluA}/(\text{FluA}+\text{Pyr})$ 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 $\text{Ant}/(\text{Ant}+\text{Phe})$, > 0.5 for $\text{IP}/(\text{IP}+\text{Bghi})$ and > 0.5 for $\text{FluA}/(\text{FluA}+\text{Pyr})$ 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

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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.,

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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.

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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, it's 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 5-ring-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 5-6-ring PAHs but abundant FluA, Pyr and BePyr suggests relatively geographically

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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

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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 each other.

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