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Comment on acp-2022-70

Anonymous Referee #2

Referee comment on "Characteristics and evolution of brown carbon in western United States wildfires" by Linghan Zeng et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-70-RC2, 2022

This manuscript investigates characteristics and evolution of brown carbon (BrC) using online photoacoustic spectrometer (PAS) that measures dry aerosol absorption of fine particles and offline filter-based approach using liquid spectrophotometric measurements of extracts of particles collected on filters. They compared the measurements at different wavelengths and found that good agreement of BrC absorption at 400 nm. While doing the comparisons, there are several assumptions and limitations, but it still provides useful information and worth publishing. The study claims that investigated samples falls under moderately absorbing class. They also investigated a particular BrC chromophore, 4-nitrocatechol and its evolution with plume ages. Results indicate that 4-nitrocatechol depleted with plume ages, while other BrC was much stable even with increasing temperature in downwind. However, some previous study reported that particulate nitrophenol and nitrocatechol isomers can contribute significantly to BrC absorption at 405 nm in aged wildfire smoke.

This is an interesting study and will be useful for the community. Overall, the manuscript is clearly written, some suggested clarifications are listed below. However, prior to acceptance, the authors should address the following questions/ suggestions and modify the manuscript accordingly.

Specific comments:

The comparison between bap, PASBrc and bap,TSBrC at 405 nm looks good. It might be good to add some discussion why the PAS derived BrC absorption is higher than the TS Brc at higher wavelength. I see that the authors add some discussion about the insoluble chromophores, but it will be good add this discussion in the results section and will be easier for readers to follow.

One of the main concerns of this manuscript is that applied method rely on several assumptions and approximation which can create a large uncertainty in estimation. I appreciate that the authors stated most of the uncertainties for example in extrapolating the wavelength-dependent differences. However, I think the authors should state overall uncertainties in estimating all the absorption values. For example, I think there is a large uncertainty in estimation of the conversion factor itself. How that translate to uncertainties in total absorption?

Page 12: Some more discussion about the absorption Angstrom exponent (AAE) measured by PAS and WS BrC and MS BrC and in context to previous study would be useful, like lower AAE value reported by PAS. I'm bit confused with the AAE values from PAS and from TS reported in Figure 3. And how did the authors calculate the modified combustion efficiency?

Size distribution data and black carbon data from SP2 are missing in the manuscript but it is important to have this information in the SI.

Authors discussed several other factors that may influence the evolution of BrC. I appreciate this discussion. However, some of the supporting data on this are not shown in the manuscript.

Summary section can be improved by proving general applicability of the closure exercise and overall applicability of this study. Another thing I find it difficult to draw some firm conclusions as this study investigated several fires with different scales, while chasing one large fire with sufficient amount time would help to decipher some of the key aspect of BrC evolution. I think this is still an important study but something to discuss in the summary section so future work can be better designed.

Minor comments:

Page 21, line 67: please check the sentence, 405 nm wavelength was mentioned twice

Overall, there are several acronyms and subscripts, it will be difficult for readers to follow, it will be good to have a table with all the acronyms.

Is there a reason to show just show one specific fire events for the wavelength dependencies in the main manuscript? I see the value for each fire events but how about combining all the dataset to get a broader picture?

Figure 4 can be move to SI. Did you calculate the correction factor for each fire events? I suggest adding some sorts of histograms and combining all the events, so reader can get an idea about the spread of the data.