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Interactive Comment

Interactive comment on "Brown carbon aerosol in the North American continental troposphere: sources, abundance, and radiative forcing" *by* J. Liu et al.

Anonymous Referee #3

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This well-written manuscript presents results obtained from aircraft measurements on brown carbon (BrC) in aerosol samples collected over central U.S. during a summer period that was impacted by several biomass burning events. Approximately 600 filter samples were collected over a range of altitudes (1 - 12 km) and extracted in water and methanol to measure H2O_Abs(365) and total_Abs(365), where the latter includes absorption data from samples extracted in both water and methanol. This filter extraction procedures are solid and provides a lot of insight into the chemical nature of the BrC constituents. During biomass burning periods, H2O_Abs(365) and total_Abs(365) were highly correlated with other known emissions from biomass burning plumes, in-

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cluding CO, ACN, and BC. Under background conditions, H2O_Abs(365) was somewhat correlated with smoke tracers, but the total_Abs(365) was not well correlated with any specific tracers, but most correlated with WSOC, possibly due to BrC evolving to a more water-insoluble state as it ages. Importantly, these data seemed to be well supported by the online measurements. Further, they estimated the BrC contribution to climate forcing using a radiative transfer model (SBDART). From these model calculations, they find that overall negative TOA aerosol scattering is reduced by $\sim$ 20% due to BrC presence.

This manuscript is really important to appear in Atmospheric Chemistry and Physics, especially since there has been a lot of interest and uncertainty in BrC formation and its potential abundance in the atmosphere. I tend to agree with the other comments made by the other 2 reviewers and also agree this should be published with minor revisions noted. My main questions below relate to the sources of BrC since that is an area that my group and many others have been interested in. One item I hope that the authors will consider in the future is to have their filters analyzed off-line by mass spectrometric techniques to provide more insights into the potential sources of BrC in both the biomass burning and background conditions. That would make this an even better paper, BUT please note I think is is already a GREAT paper. The molecular-level data would have only made this a "dream" paper. I hope they will consider this in future work if it is possible.

A few minor questions/comments:

1.) In discussing the correlation of Total_Abs(365) data with other datasets, I found it interesting that the authors also didn't consider the possibility of SOA constituents aging to produce BrC that is more soluble in the methanol extracts. For example, Lin et al. (2014, ES&T) showed that IEPOX-derived oligomers that absorb in the BrC region are more soluble in methanol. Further, one could consider aldehydes from BVOC oxidations yielding BrC oligomers through cloud droplet formations and evaporation, similar to work of DeHaan, McNeill, Turpin, Noziere, and other groups. I'm especially curious

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to know how BVOCs in this region may play a role in the BrC signals observed in the background air? Could this be important or is what you measure in the background air really just aged biomass burning?

2.) In addition to Limbeck et al. (2003) study cited in the introduction, I think the authors should also highlight work done by Lin et al. (2014, ES&T) that demonstrated isoprene epoxydiols can yield oligomeric species that can absorb light at short wavelengths, and thus, act as a BrC. Some of these oligomers were identified in SE USA fine aerosol collected at the ground sites.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 5959, 2015.