We thank the reviewers for their constructive comments and have made changes, where deemed appropriate. Specific responses to each of the comments are provided below (reviewers' comments in black and our responses in red).

Anonymous Reviewer #3:

General comments:

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, including 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 ~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.

The authors thank the reviewer for insightful comments. We agree that a molecular-level analysis would provide more insights into the chemical nature of BrC constituents, but it is beyond the scope of this paper, which we should definitely consider in future work.

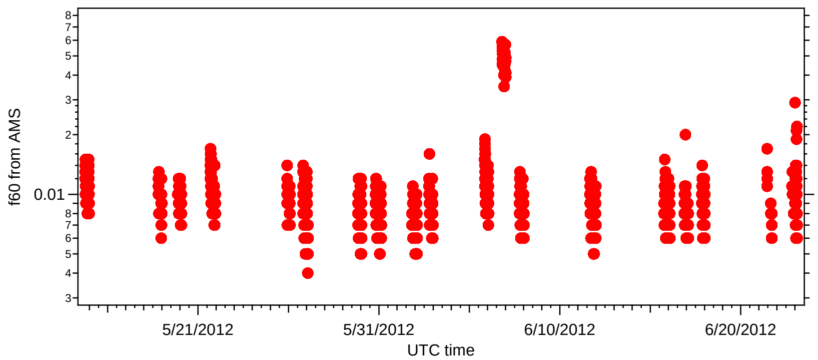
A few minor questions/comments:

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 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?

Good point.

First, observational data suggest that the campaign observations are largely impacted by biomass burning. The plot below shows the time series of f60 from AMS, which is an indicator of biomass burning.



From the plot we see, f60 from AMS is around 1% during the whole campaign, while Cubison et al. (2011) has suggested that a level of f60 ~0.3%±0.06 % is an appropriate background level for this tracer. Therefore we consider the BrC signals observed is largely impacted by biomass burning, although the relative contribution is difficult to estimate.

On the other hand, our observation did suggest that aged BrC tends to be more soluble in methanol, and light-absorbing IEPOX-derived oligomers could be a possible explanation. We believe that some fractions of the campaign observations are certainly impacted by IEPOX chemistry, especially the southeastern US where the isoprene emission is rich. However, a recent study showed that at a remote surface site in the southeast significantly impacted by BSOA, biomass burning dominated the source of BrC (Washenfelder et al., 2015), whereas BSOA had no discernible impact. Therefore, we believe aged biomass burning is the main source of the ubiquitous BrC, but that biogenic SOA cannot be ruled out. We have added this discussion into the main text, section 3.3 (Page 5970-5971).

Refs.

Lin, Y.-H., et al. (2014). "Light-Absorbing Oligomer Formation in Secondary Organic Aerosol from Reactive Uptake of Isoprene Epoxydiols." Environmental Science & Technology 48(20): 12012-12021.

Cubison, M. J., Ortega, A. M., Hayes, P. L., Farmer, D. K., Day, D., Lechner, M. J., Brune, W. H., Apel, E., Diskin, G. S., Fisher, J. A., Fuelberg, H. E., Hecobian, A., Knapp, D. J., Mikoviny, T., Riemer, D., Sachse, G. W., Sessions, W., Weber, R. J., Weinheimer, A. J., Wisthaler, A., and Jimenez, J. L.: Effects of aging on organic aerosol from open biomass burning smoke in aircraft and laboratory studies, Atmos. Chem. Phys., 11, 12049-12064, doi:10.5194/acp-11-12049-2011, 2011.

Washenfelder, R. A., et al. (2015), Biomass burning dominates brown carbon absorption in the rural southeastern United States, Geophys. Res. Lett., 42, 653–664, doi:10.1002/2014GL062444.

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.

The reference has been added into text.