

We thank the reviewers for their constructive comments. Specific responses to each of the comments are provided below (reviews' comments in black, our responses in blue, and the manuscript text follows in italics with changes in bold).

**Anonymous Referee #1:**

This paper reports laboratory experiments aimed at understanding the formation of brown carbon (BrC) from various VOCs under different conditions. Studies on the evolution (aging) of the BrC formed are also presented. This work is a systematic analysis aimed at helping to interpret a number of ambient studies that showed BrC levels varied between cities with different mixtures of emissions.

The paper is highly relevant and interesting, it highlights the importance of anthropogenic SOA to BrC. I have only minor comments.

Are not many of the VOCs tested and attributed to anthropogenic SOA (eg, re discussions on urban SOA), also produced in biomass burning? If so, I would suggest the results have broader impacts than just what is discussed here.

The reviewer raises a good point. Other than primary BrC, which is directly emitted into the atmosphere from biomass burning, gas-phase emissions from burning are also exposed to sunlight and oxidants and can generate secondary products, including SOA. And as the reviewer suggested, nitrated aromatic compounds, identified in this work as strong light absorber in anthropogenic SOA, were also observed in some SOA samples produced from aging of biomass burning (e.g., Desyaterik et al., 2013; Iinuma et al., 2010). However, studies have also shown that SOA formation in biomass burning plumes is highly variable and dependent on factors such as fuel types, mass combustion efficiency, and aerosol aging, suggesting that the contribution of biomass burning SOA should be examined and parameterized carefully (Hennigan et al., 2011). To avoid "over selling" our results, we have carefully added some discussion regarding to the application of our results to biomass burning in lines 430-435, as shown below:

*Similar light-absorbing compounds have been identified in certain SOA samples originating from biomass burning (Desyaterik et al., 2013; Iinuma et al., 2010); since substantial variations in SOA formation in biomass burning plumes have been observed both chemically and physically due to fuel types and fire aging conditions (Hennigan et al., 2011), we cannot simply assume similar effects of those parameters on SOA produced from biomass burning emissions.*

References:

Desyaterik, Y., Sun, Y., Shen, X. H., Lee, T. Y., Wang, X. F., Wang, T., and Collett, J. L.: Speciation of "brown" carbon in cloud water impacted by agricultural biomass burning in eastern China, *J Geophys Res-Atmos*, 118, 7389-7399, Doi 10.1002/Jgrd.50561, 2013.

Hennigan, C. J., Miracolo, M. A., Engelhart, G. J., May, A. A., Presto, A. A., Lee, T., Sullivan, A. P., McMeeking, G. R., Coe, H., Wold, C. E., Hao, W. M., Gilman, J. B., Kuster, W. C., de Gouw, J., Schichtel, B. A., Collett Jr, J. L., Kreidenweis, S. M., and Robinson, A. L.: Chemical and physical

transformations of organic aerosol from the photo-oxidation of open biomass burning emissions in an environmental chamber, *Atmos. Chem. Phys.*, 11, 7669-7686, 10.5194/acp-11-7669-2011, 2011.

Linuma, Y., Böge, O., Gräfe, R., and Herrmann, H.: Methyl-Nitrocatechols: Atmospheric Tracer Compounds for Biomass Burning Secondary Organic Aerosols, *Environ Sci Technol*, 44, 8453-8459, 10.1021/es102938a, 2010.

Line 179: Suggest changing: which likely explains, to: which could explain,... I really don't know of ambient data supporting Lin et al (2014). For example, Washenfelder et al. (2015, *Geophys. Res. Lett.*, 42, 10.1002/2014GL062444(2015)) saw no evidence that iePOX (isoprene SOA) contributed to ambient BrC at a remote site in Alabama as part of SOAS where the aerosol is acidic (ie, papers show that it was acid catalyzed isoprene SOA, eg, see Xu et al, *P. Natl. Acad. Sci.*, 112(1), 37-42, 2015).

We have changed Line 179 as suggested by reviewer.

We felt that the Lin study deserved citation because it is an example of SOA derived from isoprene producing light-absorbing SOA, which is in contrast to our study. We note that our experiments were conducted under conditions that are expected to inhibit formation of SOA from the IEPOX pathway, in contrast to the Lin study (i.e., no acidic seed particles were used in our experiments). We can say with certainty that the different reaction conditions relative to Lin et al will produce different types of SOA (Liu et al., 2016). Because we inhibited formation from IEPOX, we are unable to make any conclusions about optical properties of IEPOX SOA from our data. Evaluation of the real-world impact of IEPOX SOA formation on aerosol optical properties is beyond the scope of our study.

References:

Liu, J., D'Ambro, E. L., Lee, B. H., Lopez-Hilfiker, F. D., Zaveri, R. A., Rivera-Rios, J. C., Keutsch, F. N., Iyer, S., Kurten, T., Zhang, Z., Gold, A., Surratt, J. D., Shilling, J. E., and Thornton, J. A.: Efficient Isoprene Secondary Organic Aerosol Formation from a Non-IEPOX Pathway, *Environ Sci Technol*, 10.1021/acs.est.6b01872, 2016.

Last line of section 3.1, (lines 228-230) I think ambient data that includes actual light absorption coefficients are need to make this statement. The logic of the line is unclear.

We have edited the sentence as below:

*Since fulvic acid is often cited as a surrogate of **strong light-absorbing** atmospheric BrC associated with biomass burning, this comparison shows that light absorption by BrC produced from anthropogenic VOCs can be significant under certain photochemical condition, consistent with high MAC values measured previously in urban environments when biomass burning impacts were low (e.g., Zhang et al., 2011, 2013; Liu et al., 2013).*

In section 3.4, the authors might also want to consider showing changes in the compete spectra, not just changes in absorption at 365 nm. This may prove useful when comparing to ambient data. This could go in supplementary material.

Complete spectra are now provided in supplemental materials as Figure S4. We have added a description in lines 331-332 that: complete spectra in the wavelength range of 300-700 nm are provided in Figure S4.

Line 372-373 regarding the discussion that alpha pinene and isoprene SOA produces little BrC. Again I would suggest the authors look at Washenfelder et al. As noted above, there is no evidence for isoprene SOA, but maybe pinene SOA from night-time reaction with NO<sub>3</sub> radical.

Discussion on pinene SOA from night-time reaction with NO<sub>3</sub> radical has been added to lines 385-386. We have also edited lines 372-373 to emphasize that our statement refers only to the conditions we investigated in our experiments (see below). We unfortunately did not conduct any experiments investigating BrC formation from NO<sub>3</sub> oxidation of pinene.

Sentence at lines 372-373 (now lines 380-382) is edited as:

*Although  $\alpha$ -pinene and isoprene have large contributions to the global SOA budget, they were shown to produce SOA with very small light absorption coefficients **under the photochemical conditions we investigated**, which agrees with literature data (i.e., Nakayama et al., 2010;Lang-Yona et al., 2010).*

Line 385, typo, ranged ?

The word “ranged” has been changed to “ranging”.