

We would like to thank the reviewer for their valuable suggestions and time. Our responses are given below.

Anonymous Referee #1 Received and published: 20 December 2016

Referee Comment: In this work, three different methodologies were used to calculate the contributions of absorption from BC, BrC and lensing. The authors described the methodologies in the Results and Discussion section (Sec. 3.3). I would suggest the three approaches discussed in the Sec. 2 (Materials and Method).

Author Response: We think this description is very important to our result. We decided to keep descriptions in result and discussion section.

Referee Comment: I would suggest providing a schematic of the instrument setup in the Materials and Method section.

Author Response: Schematic of instrumental setup is provided in the Material and Method section and Figure numbers are updated accordingly.

Referee Comment: The efficiency of the thermal removal mechanism of non-refractory component is a critical point, but in this work is poorly investigated. Incomplete removal of organics (e.g. low-volatile BrC coatings on BC surface) by the thermal denuder would influence the estimate in the relative contributions from BC, BrC and lensing, especially for approach 1. It will be helpful to estimate the removal of low-volatile component after the heating stage. If the author cannot quantify the efficiency of the thermal removal, they can make a sensitive analysis to discuss the uncertainties of relative contributions from BC, rBC and lensing due to incomplete removal of low-volatile organics.

Author Response: We completely agree with the referee that the efficiency of the thermal removal mechanism of non-refractory components by the thermal denuder (TD) is critical and this will influence the estimation from approach 1. In general, it is not straightforward to estimate the removal of low/extremely low volatility components by the TD unless the downstream aerosol is monitored by a combination of SP2, AMS, or SMPS instruments, which was not done in the current study. Saleh et al. 2014 (measuring aerosol from the same burns as this study) estimated about 10 % of the extremely low volatile organic compounds (ELVOCs) would not be evaporated by the TD, but this value will depend on TD specifications (tube diameter, length and flow rate through TD), which are different for our two thermal denuders. Even if the fraction of non-refractory component that is not removed by the TD is accurately estimated, accurate estimation of the influence of ELVOCs in absorption enhancement and fraction of BrC absorption is difficult because they have different absorptivity (imaginary refractive index) than that of low and semi volatile components. As mentioned in page 12 Line 20-26, we strongly believe that our TD did not remove all the organics so our absorption enhancement values estimated by approach 1 are underestimated (assuming no BrC absorption at 660 nm). By approach 1, the relative contribution from BC is over estimated while BrC and lensing are underestimated, but quantifying the actual uncertainties of relative contributions from BC, BrC, and lensing is complex due to the reasons as

discussed above. This is why we present the other approaches, which are much less dependent on the effectiveness of the thermal denuder.

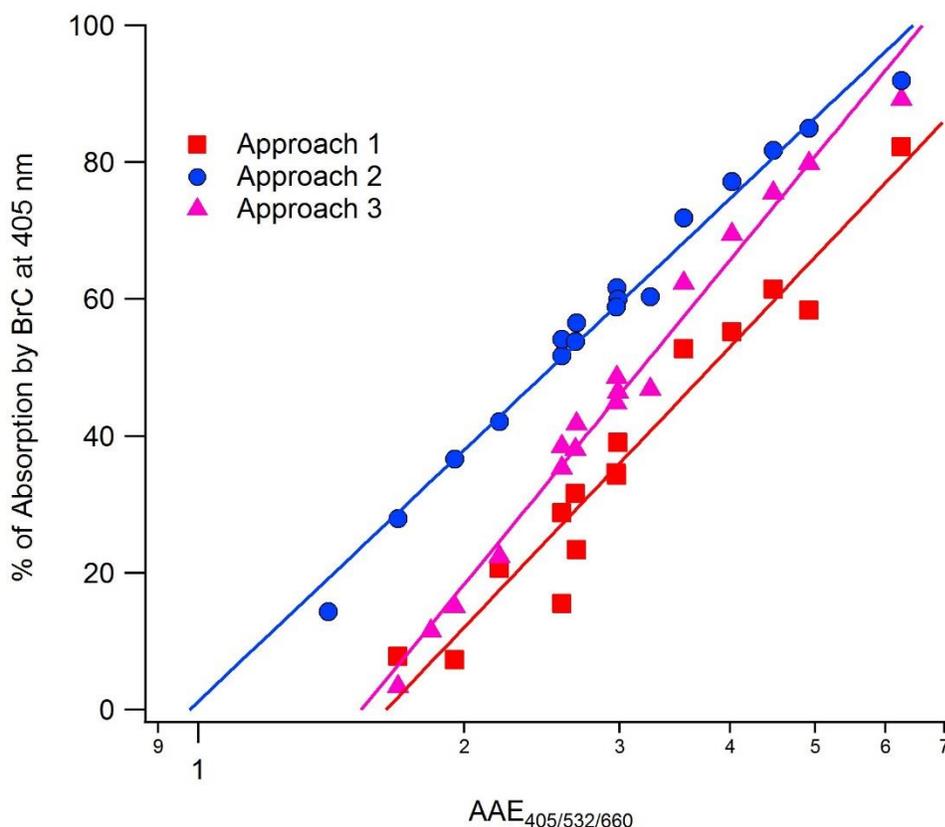
Referee Comment: Due to incomplete removal of low-volatile organics, the E_{abs_660} was underestimated and the $b_{abs_405_den}$ was overestimated in Eq. (3), which would result in an unclear uncertainty (overestimate, underestimate or counteraction) in the fraction of absorption from BrC using approach 1. Meanwhile, the assumption of coated BC with an AAE of 1 in approach 2 led to an overestimate in the fraction of absorption from BrC. I do not know why the author could conclude that the BrC contributions derived from approach 2 was closer to reality than approach 1.

Author Response: Due to incomplete removal of low-volatile organics, estimated fraction of absorption due to BrC using approach 1 is most likely underestimated. The logic is that Eq. (3) can be simplified as

$$b_{abs_405_BrC} = b_{abs_405_dry} - E_{abs_660} \times b_{abs_405_den}$$

$$b_{abs_405_BrC} = b_{abs_405_dry} - \frac{b_{abs_660_dry}}{b_{abs_660_den}} \times b_{abs_405_den}$$

The denuded absorption at both 660 and 405 nm will be overestimated due to incomplete removal of organics, but the problem is expected to be worse at 405 nm because both brown carbon and lensing increase the 405 nm denuded absorption while lensing is the dominant effect for the 660 denuded absorption. Given this, the ratio $\frac{b_{abs_405_den}}{b_{abs_660_den}}$ is expected to be larger than one and hence BrC absorption will be underestimated because both the dry absorptions (405, 660 nm) will not be affected.



The figure above is a combination of panels d-f from Figure 7 shows the fraction of absorption due to BrC at 405 nm vs AAE for all approaches. As depicted from the figure, approach 1 is close to approach 3 when AAE is small but approach 2 is close to approach 3 when AAE is high. This clearly shows that if aerosol is dominated by BrC, the fraction of absorption by BrC estimated by approach 1 is much smaller than that estimated by approach 2 or 3. Approach 3 is thought to be the maximum possible impact from lensing (AAE = 1.6) and therefore the fact that approach 1 is below approach 3 strongly suggests that approach 1 is low. This result may be different from other studies if TD removes non-refractory materials more efficiently. This is why we concluded approach 2 would be close to reality.

Added Text Location: Section 3.3, Page 10, Line 17

Added Text: Due to incomplete removal of low-volatile organics, estimated fraction of absorption due to BrC using approach 1 is most likely underestimated. The logic is that Eq. (3) can be simplified as

$$b_{abs_405_BrC} = b_{abs_405_dry} - E_{abs_660} \times b_{abs_405_den}$$

$$b_{abs_405_BrC} = b_{abs_405_dry} - \frac{b_{abs_660_dry}}{b_{abs_660_den}} \times b_{abs_405_den}$$

The denuded absorption at both 660 and 405 nm will be overestimated due to incomplete removal of organics, but the problem is expected to be worse at 405 nm because both brown carbon and lensing increase the 405 nm denuded absorption while lensing is the dominant effect for the 660 denuded absorption. Given this, the ratio $\frac{b_{abs_405_den}}{b_{abs_660_den}}$ is expected to be larger than one and hence BrC absorption will be underestimated because both the dry absorptions (405, 660 nm) will not be affected.

Referee Comment: Page 1 Line 17: The temperature unit should be “C”.

Author Response: The temperature unit has been changed to C.

Referee Comment: Page 1 Line 18-20: Three approaches were used to calculate the contributions of absorption from BC, BrC and lensing. However, the author only described two methodologies (i.e. with one: : : : .and the other: : : .).

Author Response:

We describe the two extreme approaches in the abstract and leave the approach that lies between these two extremes for the body of the paper.

Referee Comment: Page 7 Line 12-14: Please define the absorption Angstrom exponent (AAE), such as using an equation.

Author Response: The AAE is now defined as an Equation.

References:

Saleh, R., Robinson, E. S., Tkacik, D. S., Ahern, A. T., Liu, S., Aiken, A. C., Sullivan, R. C., Presto, A. a, Dubey, M. K., Yokelson, R. J., Donahue, N. M. and Robinson, A. L.: Brownness of organics in aerosols from biomass burning linked to their black carbon content, Nat. Geosci., 7, 647–650, doi:10.1038/ngeo2220, 2014.