

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

Anonymous Referee #3 Received and published: 28 December 2016

Referee Comment: For the correlation analyses between Eabs and AAE, EC/OC ratio, or MCE and between the fraction of BrC absorption and AAE, EC/OC ratio, or MCE, the authors sometimes use logarithm but not in some other cases. For example, the author reported that the log(Eabs_Den_405) linearly correlates with AAE (Fig. 4), and the “% of absorption of BrC” linearly correlates with log(AAE) (not AAE itself) (Fig. 7). In contrast, the log-log plot was used for both of Eabs vs. EC/OC and % of absorption of BrC vs. EC/OC. The authors need to add detailed explanation on these choices. For the discussion of these correlations, the term “logarithm of” should be added in the text, if the correlation analysis was conducted for the logarithm.

Author Response: The referee has two good observations. We replaced the term “log” by “logarithm of” in the text. We have also added the following paragraph to the text to explain the criteria by which we chose the variables for regression.

Added Text Location: Section 3.2, Page 9, Line 3

Added Text: It is notable that some regressions are done for a semi-log plot while others are linear or log-log. The type of regression was chosen based on objective criterion for simple regression. Namely that the residuals are equally scattered from the regression line and that the residuals are as close as possible to a normal distribution. The model (either LogY vs LogX, logY vs X, or Y vs LogX) which satisfied these criterion for simple linear regression was chosen.

Referee Comment: This paper reported the small lensing effect for all fuels and burning conditions. Is the magnitude of the lensing effect reasonable, if you assume all OC is used for the coating with the core-shell structure?

Author Response: E_{Abs} at 660 nm, which we attribute to lensing, varied from 0.92 ± 0.09 to 1.43 ± 0.17 (Page 8 Line 24) which is consistent with the range reported in previous literature (McMeeking et al., 2014; Lack et al., 2012). As mention in the text, the average BC core and particle size during FLAME-4 was 100 and 200 nm respectively, which, based on Mie core/shell theory would give E_{Abs} of roughly 1.5 assuming internal mixing and varying slightly depending on the refractive indices utilized in the analysis (McMeeking et al., 2014).

Referee Comment: The main findings of the paper “The fraction of absorption from BrC shows reason-ably good correlation with AAE and EC/OC at both 405 and 532 nm.” are not surprising, because the relative contribution of BrC is expected to increase with increasing OC concentration and that the AAE is expected to increase with increasing BrC (OC). The results imply that the light absorbing properties (such as mass absorption cross section and imaginary part of refractive index) of OC do not largely change with EC/OC ratio, fuel types, and burning conditions. If so, the results obtained in this work may be inconsistent with the results of Saleh et al. (2014). I think that the discussion on this point and the more detailed descriptions on the evidence leading to the conclusion be-low should be added. “This result is distinct but not inconsistent with Saleh et al. (2014) who found that the imaginary index of refraction increases with increasing BC/OA ratio.

These two results can be understood with the idea that brown carbon grows darker as emissions have a higher fraction of black carbon relative to non-refractory organic mass, but the fraction of total absorption caused by brown carbon increases as the amount of organic mass increases and the black carbon to organic carbon mass ratio decreases.”

Author Response: Perhaps the main findings that “The fraction of absorption from BrC shows reasonably good correlation with AAE and EC/OC at both 405 and 532 nm.” are not surprising to the reviewer, but they are certainly notable. There are many variables that contribute to absorption including, most notably, the mixing state of the aerosol and the refractive index of the organic material, but also including the morphology of the black carbon core and other variables. Given all this, it is very interesting that a simple correlation of this type can reasonably represent the fraction of absorption by brown carbon. The reviewer is incorrect in stating that the results show that the imaginary refractive index of the organic carbon does not change with EC/OC ratio. This would be implied if there was a linear relationship between the amount of OC (not EC/OC ratio) and the absorption coefficient. The result shown, a linear correlation between the % of absorption by brown carbon and the EC/OC ratio, is far more complex and is not easily reduced to find the imaginary refractive index of the organic aerosol without knowledge of the black carbon size distribution, the total size distribution, and the mixing state of the aerosol. Because we do not have this data available, we feel that the statement currently made about our results relative to those of Saleh et al. (2014) is all that can be said. We agree with the reviewer that this is an area that needs further study to reconcile these results and we are actively involved in this research now.

Referee Comment: Page 3, Line 2 Material and Methods: If a part of the data (fuel) used in this work is same with that used in Pokhrel (ACP2016), it is better to add the information on that and how the authors chose the data (fuel) used in this work.

Author Response: Both this paper and Pokhrel et al., 2016 are from data collected during FLAME-4 and they are based on the same data set. This is already described in the paper and Pokhrel et al., 2016 is repeatedly referenced. The papers are very different. Pokhrel et al (2016) discusses the single scattering albedo and absorption angstrom exponent dependence on MCE and EC/OC. These optical properties are very different from E_{abs} and the fraction of absorption from brown carbon discussed in this work. The only real overlap between the results presented in the two papers is the use of AAE values.

Referee Comment: Page 4, 2.1 Inlet system

- a) Have you estimate the particle losses in the Nafion dryer and the activated carbon monolith?

Author Response: We do not discuss the particle losses in the Nafion dryer and the activated carbon monolith because both of these were on upstream of all PAS channels any losses will have similar effects on all channels. Since our results are all intensive properties particle losses will not affect the results unless the dramatically alter the size distribution, which is not expected because large aerosol was removed with the cyclone impactor and the vast majority of the mass was typically much smaller than a micron.

- b) Did you check the possible contribution of removal of semi-volatile organic compounds to the amount and optical properties of BrC through the change in the gas/particle partitioning?

Author Response: There is always concern that semi-volatiles could be lost in an inlet system, but there is no way to quantify if that occurred in the current dataset because all data was collected through the inlet. The PAS and the EC/OC instruments were on different inlets.

- c) Line 17: How often did you insert the filter for the baseline measurements?

Author Response: A filter was performed every 5 to 10 minutes. This statement has been added to the text.

Referee Comment: Page 5, 2.4 Particle loss in Thermal denuder

- a) How did you measure the particle size dependence of particle loss? Did you use two SMPS system and place them upstream and downstream of thermal denuder?

Author Response: A poly-disperse aerosol was measured going through the denuder and bypassing the denuder and the two size distributions were divided by one another to generate the size dependent particle losses shown.

- b) In Fig. 1, the particle transmittance only above 100 nm were given. How was the transmittance for smaller particles? If the particles with a diameter less than 100 nm are negligible, I recommend to the authors to give some information on this point.

Author Response: Particle transmission for particles less than 100 nm size was noisy. In terms of mass (on which absorption coefficient depends), these particles have a small contribution.

Referee Comment: Page 6, lines 22-24 “The calibration of the dry 405 nm channel determined without the high ozone points (what was done for most of the project) was consistently closer to the slope determined using all ozone concentrations (including the high ozone points) than the calibration of the denuded 405 nm channel without the high-ozone points.” => I recommend to adding more qualitative information on this point.

Author Response: Except for the last three days of experiments, ozone calibration of the PAS was performed at concentration levels which gives maximum absorption of 20 Mm^{-1} at 405 nm. During last three days, the ozone calibration was performed at much higher concentrations of ozone and absorption at 405 nm peaked at 40 Mm^{-1} . For the dry 405 nm channel, the calibration constant (Slope of PAS IA vs CRDS extinction) remained fairly constant for both conditions whereas calibration constant of the denuded 405 nm channel showed significant drift. Hence we decided to adjust denuded channel to match dry channel. We feel the paragraph stated is sufficient for the paper but have added detail here.

Referee Comment: Page 7, line 31 “For fires without backup filters or those that were below the detection limit, the average OC correction for that fuel type was applied: rice straw ($2.0 \pm 0.4 \%$), ponderosa pine (1.2%), black spruce ($2.9 \pm 1.6 \%$), and peat ($3.1 \pm 0.8 \%$). For fuel types without backup filters collected, the study average OC artifact ($2.4 \pm 1.2 \%$) was subtracted.” => It seems

that the authors assume the amount of carbonaceous gas adsorption is proportional to the mass concentration of OC. The assumption may not be reasonable if the filter is saturated.

Author Response: We agree with referee about the assumption that is made in applying this artifact correction. We have added a statement to this effect, as well as evidence that the filter was not saturated.

Added Text Location: Section 2.7, Page 8, Line 3

Added Text: This approach to artifact correction assumes that the amount of carbonaceous gas adsorbed is proportional to the mass concentration of OC; this assumption is considered to be reasonable because the back-up filters contained less than $5.6 \mu\text{g OC cm}^{-2}$ and similar quartz fiber filters become saturated above $6 \mu\text{g OC cm}^{-2}$ (Turpin et al., 1994).

Referee Comment: Page 9, line 24 Are the particle sizes (100 and 200 nm) given here volume-based average diameters or number based average diameters?

Author Response: These are the number based average diameters

Referee Comment: Page 14, line 2 “Figure 5” may be “Figure 7”

Author Response: We changed this to Figure 7.

References:

Turpin, B. J., Huntzicker, J. J., and Hering, S. V.: Investigation of organic aerosol sampling artifacts in the Los-Angeles basin, Atmos. Envir., 28, 3061–3071, 1994.