

Reviewer #2:

A three-wavelength photo-acoustic soot spectrometer, an aerosol mass spectrometer, and an aerosol chemical speciation monitor were used to measure and show the relationship between the relative abundance of organic aerosol to black carbon and its relationship to the Absorption Angstrom Exponent (AAE) in ambient samples collected in urban and rural areas in the Pearl River Delta region of China. Since the AAE is widely used in attributing the light absorption of brown carbon at shorter wavelengths, the method was improved by statistical analysis and applied to the collected data. The findings from this study are relevant to the readers of ACP as well as the air pollution and aerosol communities. The manuscript includes enough discussions of the limitations and implications of the study. Therefore, I recommend publication of this manuscript upon consideration of the suggested revisions listed below.

### **General Comments**

- Be consistent in the use of brown carbon or BrC, and Absorption Angstrom Exponent or AAE.
- Better tie results with what they mean. It is difficult to follow when several numbers are listed in a long sequence.
- Choose words that are more descriptive of what is being said, or follow up with a brief explanation. Some choices are vague and leave the reader to wonder in what context they are being applied, or are being related to. Example: levels, comparatively, and convenient.
- Remove words that do not add any significance to the text.
- Be consistent with tenses.
- Check for the proper use of “in”, “by”, “on”, etc.
- Cross-referencing different sections within the text makes it for a difficult read.

**REPLY:** The above suggestions were all taken and the relevant sentences have been carefully modified in the revised manuscript.

### **Specific Comments (by line)**

- 55, Unclear when said that, “BrC aerosol could contribute more than 65 and 15% light absorption at 370 nm”

**REPLY:** The sentence has been rephrased in the revised text as below:

“BrC aerosol could contribute more than 65% of light absorption at 370 nm and 15% at a mid-wavelength (Favez et al., 2009).”

- 60, Related to line 45, but too far apart.

**REPLY:** The relevant sentences have been moved closely to each other in the revised text.

- 63, The authors could expand on why East Asia in one of the five regions of atmospheric brown clouds.

**REPLY:** The suggestion was taken and the sentences have been modified in the revised text as below:

“South and East Asia are typical regions of atmospheric brown clouds (ABC) (Alexander et al., 2015). Biomass burning has been recognized as a significant contributor to ABC, including forest burning, crop waste burning, traditional religious activities and residential burning in those countries like India, China, Thailand, etc. (Venkataraman et al., 2006; Yan et al., 2006; Chakrabarty et al., 2013; Chakrabarty et al., 2014; Huang et al., 2012). ”

- 75, What is it meant by universal?

**REPLY:** It has been revised to “widely used”.

- 110, What type of “little local emission”

**REPLY:** It has been revised to “little local fossil fuel combustion emission nearby”

- 117, It is stated that tunnel experiments were performed three times. Does that mean measurements were done three times inside each tunnel? I only see two tunnels, the Tanglangshan and the Jiuweiling tunnel. Is there another one? Please reword this sentence.

**REPLY:** The sentence has been modified in the revised text as below:

“We performed tunnel experiments three times in Shenzhen urban areas: twice in the Tanglangshan tunnel (TL) and once in the Jiuweiling (JW) tunnel.”

- 128, Rearrange order of the sentence to better describe why biomass burning simulation experiments were done in the combustion laboratory. Although it is known that biomass burning is a great source of BrC, as it is, it appears that biomass burning experiments were done without a purpose.

**REPLY:** The relevant sentences have been modified in the revised text as follows:

“Moreover, since biomass burning is recognized as an important source of BrC (Ramanathan, et al., 2007) and is a popular source in rural areas in PRD, especially during the harvest season (He et al., 2011; Zhang et al., 2013), we performed biomass burning simulation experiments in a combustion laboratory to study the spectral dependence of aerosol light absorption in biomass burning smoke.”

- 138,139, Why was the water boiling test protocol developed by the University of California used?

**REPLY:** We just refer to the protocol to make our burning processes normalized and more reliable.

- 152, Please clarify how the data was processed and how AMS data was related to PASS-3 data. Remove “in the later data analysis and discussion.”

**REPLY:** The data processing has been clarified in the revised text as below:

“Then, we processed the 2 min time resolution data of absorption at three wavelengths for half hour averages and made further data analysis based on the half hour time resolution datasets. On the other hand, we also processed the 10 min time resolution data of organic aerosol derived from AMS or ACSM for half hour averages to explore the relationship with the absorption datasets.”

- 163, Expand on how the ACSM is a convenient version of the HR-ToF-AMS.

**REPLY:** The relevant sentences have been expanded and rearranged in the revised text as below:

“An aerosol chemical speciation monitor (ACSM) (Aerodyne Research, MA, US) was used at the HS site and in the tunnel experiments with a time resolution of 10 min. In comparison with HR-ToF-AMS, ACSM was smaller and more convenient to be transported to field sampling sites and setup in a monitoring car with limited space. The detailed description of ACSM was given by Ng et al. (2011).”

- 169-171 What are satisfied results. Please also reword this sentence.

**REPLY:** The words "satisfied results" were removed and the sentences have been reworded as below:

“The calibrations of PASS-3 for flow rate, laser power, and absorption were conducted following the standard procedures provided by the operational manual, which were also applied in relevant previous studies (Arnott et al., 2000; Lan et al., 2013; Nakayama et al., 2015). Firstly...”

- 178, Table 1. I see how the calibration adds to the validity of the data, but I would consider adding such information to a supplement and not to the main text.

**REPLY:** As reviewer #1 cared about the calibration information very much, the authors think it is better to keep the calibration part in the main text.

- 224, Please explain unfavorable meteorological conditions in PRD.

**REPLY:** The reasons have been added into the revised text as below:

“The higher aerosol pollution observed in the winter could be attributed to the unfavorable meteorological conditions in PRD in the winter, when the air mass came from the polluted northern continent with a overwhelming frequency and the atmospheric boundary layer became shallower due to lower ambient temperatures (Huang et al., 2014).”

- 226, Please consider a better location to compare the data with. I do not see how Denver, CO relates to the PRD region.

**REPLY:** The comparison with Denver has been deleted in the revised text. No comparable study has been found in the literature currently and thus better comparison cannot be made.

- 231, Reword this sentence – “suffering from the severe polluted outflow air from its

northeastern the Guangzhou...”

**REPLY:** The sentence has been modified in the revised text as “suffering from the polluted air outflow from the northeast, where the megacity of Guangzhou was located, during the fall and winter seasons (Gong et al., 2012).”

- 233, Refer to HS again, if this is the place you are still talking about.

**REPLY:** The suggestion was taken and the sentence has been modified in the revised text as “The relatively higher values of  $AAE_{405\_781}$  and  $AAE_{532\_781}$  in the rural<sub>-fall</sub> campaign might be related to the biomass burning in the farmland surrounding the HS site.”

- 246, Clarify if dust events happened. It is mentioned that there were no dust events, but the authors follow to say that they scarcely happened.

**REPLY:** The sentence has been modified in the revised text as “there was no dust event during the three campaigns”

- Fig. 1, Explain why there are gaps in the data in Urban-winter (January) and Urban-fall (September).

**REPLY:** The data missing was due to the failure of power supply or pump repairmen.

- 260 - 271, Seems to be redundant information. The information was already stated previously.

**REPLY:** The first sentence has been removed in the revised text, while other sentences are remained because in the previous part, the authors just gave a brief introduction of our method.

- 299, This is the third time it has been stated that the assumption of  $AAE = 1$  for BC is not reasonable. I believe this point has been made.

**REPLY:** The sentence has been deleted in the revised text.

- 328-344, Please reword.

**REPLY:** We have modified this paragraph to make it clearer.

- 350-359, Data is too sporadic. It is difficult to follow what set of data goes with what site or wavelength.

**REPLY:** The relevant sentences have been modified in the revised text as below: “In result, the average light absorption of BrC at 405 nm was 3.0, 1.4, and 3.9  $Mm^{-1}$  in the urban<sub>-winter</sub>, urban<sub>-fall</sub>, and rural<sub>-fall</sub> campaigns, respectively, contributing 11.7%(±5%), 6.3%(±4%), and 12.1%(±7%) of the total aerosol light absorption, respectively. Here, the values in the brackets were the relative uncertainties calculated through Equation 4. The average light absorption of BrC at 532 nm was 1.9, 0.7, and 1.2  $Mm^{-1}$  in the urban<sub>-winter</sub>, urban<sub>-fall</sub>, and rural<sub>-fall</sub> campaigns, respectively, contributing 10.0%(±2%), 4.1%(±3%), and 5.5%(±5%) of the total aerosol light absorption, respectively.”

- 363, What is it meant by level?

**REPLY:** The word has been modified as “fraction”

#### Technical Comments

- Figure 2, Any reason why when r\_org/bc reach 2 the AAE's drop for all cases?

**REPLY:** The authors do not see the drop for all cases, although there are a few dropping points, which are believed to be influenced by random errors in this study. More measurements are needed to explore whether the dropping points represent some special mechanisms.

- How many points were used for the figures?

**REPLY:** For each field campaign, we processed the original data to half hour averages and thus have at least 700 data points of light absorption at each wavelength. Based on these data, further data analysis was made, including plotting the figures. This information has been added into section 2.2 in the revised text.

- Table 4, Please specify the burning modes for all biomass types. My understanding from looking at the table is that only the modes for Peanut stalk and Short straw are specified. Perhaps arranging the AAE's in ascending or descending order would make it easier for the reader to compare results.

**REPLY:** We have specified burning mode for each biomass type and the data in the table was arranged in descending order according to the AAE<sub>405-532</sub> values.