Reviewer #1:

This paper characterizes the Absorption Ångström exponent (AAE) values of aerosols at urban and rural site in the PRD region of China based on the measurements using a three-wavelength photoacoustic spectrometer and reports the AAE values for pure black carbon (BC) and contributions of light absorption by brown carbon (BrC). This manuscript includes sufficient originality, and the topic seems to fit the journal. I recommend publication to ACP after the points below have been addressed.

Major comments:

Question 1 The evaluation of accuracy for the measurement of absorption coefficients is critically important in this study. Therefore, more detailed information on the calibration procedure and uncertainties for the measurements of absorption coefficients at each wavelength should be added. What kind of particles did you use for calibrations? How the theoretical value of 1 was determined? If the authors used similar methods with those used in the previous studies (e.g., Arnott et al. 2010, Nakayama et al. 2015), it would also be better to refer them.

REPLY:

1) More detailed calibration information has been added into section 2.3, 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, the flow rate of sample air was calibrated by a soap film flow meter, with the results shown in Table 1; secondly, the laser power for each wavelength was calibrated by a laser power meter and the error in Table 1 indicated the reading difference between the laser power meter and the laser detector inside the instrument; thirdly, the light absorption calibration was performed by measuring highly absorbing NO_2 (200 ppm) at 532 nm. Then a good linear regression (with R^2>0.99) of the calculated extinction coefficient of NO_2 and the measured light absorption was established. Since the scattering of gas is negligible, the extinction of NO_2 should be very close to the absorption of NO_2, and thus the slope of the fitting curve should be very close to 1, as shown in Table 1. The detection limit of aerosol light absorption with 2 s time resolution was 10, 10, and 3 Mm^{-1} at 405, 532, and 781 nm, respectively."

2）More description of uncertainty calculation has been added into section 2.4, as below:

"The uncertainty of the absorption measurement at a wavelength (U_{Abs_λ}) includes the fit to the absorption calibration slope, the electronic noise within the instrument (Lack et al., 2012a), as well as the drift correction of signals, and can be expressed as below:

$$\Delta X = \sqrt[2]{(\Delta X_{calibration})^2 + (\Delta X_{noise})^2 + (\Delta X_{drift})^2}$$

(5)

$$U_{Abs\_\lambda} = \Delta X / Abs_{\lambda}$$

(6)

Where $\Delta X_{calibration}$ is derived from the uncertainty of the regression slope under a 95% confidence level (p); ΔX_{noise} can be calculated through uncertainty propagation of noise equivalent absorption measured by PASS-3 every 2 minutes; ΔX_{drift} is the standard deviation of the averaged baseline absorption of filtered air. Finally, ΔX is divided by Abs_{λ} to get the corresponding relative uncertainty (U_{Abs_λ}). In result, the relative uncertainties of the absorption measurements at the three wavelengths were ~1.2% for the campaign of $urban_{\_winter}$, 0.8–0.9% for the campaign of $urban_{\_fall}$, and 1.5–1.6% for the campaign of $rural_{\_fall}$."

Question 2 It is also unclear what do the values of error in the laser powers and slopes in Table 1 mean and how did you decide these values. In addition, it is amazing that the all values for $Rural_{\_fall}$ and Tunnel is same in Table 1. How many times did you calibrate the instrument?

REPLY: Clarification has been made in the revised section 2.3 as in the reply to Question 1. The same calibration values of tunnel and $rural_{\_fall}$ were a copy-paste error, which has been corrected now. Calibrations were conducted both at the beginning and at the end of a field campaign in our study.

Question 3 If you used the soot particles for calibration, the systematic uncertainties of the calibration factors for scattering measurements also influence to the determination of calibration factors for the absorption measurements. The estimated systematic uncertainties of the calibration factors for absorption measurements at each wavelength are needed to be taken into account to estimate the uncertainties for AAE and AAE_BC, as well as for the light absorption and contributions of BrC. In addition to the systematic uncertainties, influence of drift of the signals in 30 min should also be added in eq. (5).

REPLY:

1) The author used highly absorbing NO_2 (200 ppm) for absorption calibration and scattering of gas was negligible and thus needs not to be considered.

2) The influence of drift has been added in the total uncertainty calculation as suggested. The details are given in the reply to Question 1.

Question 4 In section 3.2 and 3.3, the authors reported that the linear relation between AAE and r_org/bc was obtained for all cases. However, I think the linear relation between AAE and r_org/(bc+org) may be expected, if a simple mixing rule is assumed.

REPLY: Firstly, the authors think that the meaning of the linear relation between AAE and r_org/bc is very similar to that between AAE and r_org/(bc+org). When org=0, they all get an intercept representing the AAE for pure BC. Secondly,

converting the absorption at 781 nm to the mass concentration of BC will introduce additional uncertainties because mass absorption efficient (MAE) needs be assumed. Thirdly, the ratio of OC and EC was also used to explore the correlations with ambient AAE values in a previous study (Utry et al., 2014), and proved to be a good index of the relative amount of OC and EC. Therefore, we believe that using r_org/bc is a better choice.

Question 5 The plot between AAE and OC/EC was used in Utry et al. (2014). I recommend to adding some information and discussion in the introduction and discussion sections.

REPLY: The following discussion has been added into the text as recommended.

1) In the introduction: "Some previous studies showed that ambient AAE was significantly affected by aerosol OC/EC (organic carbon/elemental carbon) ratio, suggesting a potentially important role of organic matter in aerosol light absorption (Utry et al., 2014)."

2) In section 3.2: "Utry et al. (2014) also revealed a strong correlation between AAE and aerosol OC/EC at an urban site in Hungary, where OC was mainly emitted from wood burning and contained a large amount of BrC"

Question 6 In section 3.2, the authors reported the difference in AAE_BC values at SZ site and those at HS site and pointed out the difference of sources (fuel combustion and biomass burning) as a source of the difference of AAE_BC. Although it is interesting findings, more detailed discussion on the relationship between source (size, shape, and mixing state of BC) and AAE_BC value should be added.

REPLY: More discussion about the difference of $AAE_{\_BC}$ between the urban and rural sites has been added as below:

"In PRD, Lan (2013) ever found that the BC diameters of both vehicular exhaust and biomass burning were generally above 100 nm, using a single particle soot photometer to measure, and the BC diameters of vehicular emissions were even larger. On the other hand, Gyawali et al. (2009) found that the AAE value would decrease as the BC diameter increases in the range of 0.1–1 μm by theoretical modeling. Therefore, the larger $AAE_{\_BC}$ obtained at the rural site could be a result of the smaller BC diameters of biomass burning in PRD."

Question 7 In section 3.4, the authors reported that "BrC could play a more important role under polluted condition". I recommend to adding some discussions on the source of BrC in SZ site in winter. It seems to be nice to calculate the AAE values for BrC for the discussion of the source of BrC.

REPLY: The following relevant information and discussion has been added into section 3.4 as recommended.

1) "The higher BrC contribution in the $urban_{\_winter}$ campaign than that in the $urban_{\_fall}$ campaign suggested that BrC could play a more important role in polluted continental air mass, since Shenzhen had a higher frequency of continental air mass from the

north than that of marine air mass from the south in winter."

2) "On the other hand, the highest BrC contribution at 405 nm in the rural$_{-fall}$ campaign could be attributed to the influence of biomass burning in the farmland nearby, which was supported by the biggest difference of BrC absorption between 405 and 532 nm: the AAE_{405_532} of BrC was calculated to be 1.7, 2.5, and 4.3 for the campaigns of urban$_{-winter}$, urban$_{-fall}$, and rural$_{-fall}$, respectively. High AAE_{405_532} was found to be a feature in the biomass burning simulation experiments, as in Table 4. Especially strong absorption at 404 nm of biomass burning-emitted BrC was also found by Lack et al. (2012b). The lowest AAE_{405_532} of the urban$_{-winter}$ campaign indicated that fossil fuel combustion, rather than biomass burning, seemed to be the major source of BrC in Shenzhen in winter."

Minor comments:

Question 1 Page 28454, lines 12-13 and page 28466 lines 17-18: I think it better to revised the sentence from ".. AAE values … at 405 nm, and … at 532 nm" to ".. AAE values … between 405-781 nm, and … between 532-781 nm".

REPLY: The suggestion was taken and the sentences have been modified.

Question 2 Page 28459, lines 12-13: Are these detection limit values for 2min data?

REPLY: These detection limit values are for 2s data, which has been stated in the revised sentence.

Question 3 Page 28461, lines 20-22: Please add the uncertainties and their definition.

REPLY: The sentences have been revised as below:

"The campaign-average ambient AAE_{405_781} values (± relative uncertainties) were calculated to be 1.05 (±0.01%), 0.92 (±0.10%), and 1.22 (±0.002%), respectively, for the urban$_{-winter}$, urban$_{-fall}$, and rural$_{-fall}$ campaigns, while those of AAE_{532_781} were 0.98 (±0.01%), 0.82 (±0.05%), and 1.00 (±0.001%), respectively. The corresponding uncertainties in the brackets were calculated through the uncertainty propagation of the absorption measurement uncertainties based on Equation 1. The relatively higher values of AAE_{405_781} and AAE_{532_781} in the rural$_{-fall}$ campaign might be related to the biomass burning in the farmland surrounding the HS site."

Question 4 Page 28466 line 16: "the absorption of pure BC" => "the AAE of pure BC"?

REPLY: Corrected.