Response to the Reviews' Comments

We thank the reviewer for providing insightful comments and helpful suggestions that have substantially improved the manuscript. Below we have included the reviewer' comments in italic followed by our responses in blue. In the revised manuscript, we have incorporated those changes accordingly.

Comment 1: *Could you state the reason why different measured AAE when choosing different range of wavelength for the calculation. According to Fig. S4, this discrepancy seems large at certain period. Which AAE have you used to calculate the BrC absorption contribution?*

Response:

The BC and BrC optical properties are all wavelength dependent to some extent, i.e., their light absorption coefficients may vary (usually nonlinearly) at different wavelength. Therefore, the calculated AAE will also be different when different wavelength pair are chosen.

In Fig. S4, the time series of particle AAEs are based on the aerosol total absorption ($\sigma_{abs,\lambda}$) measurements by the Aethalometer at various wavelengths. The AAE_{BC} is obtained from the core-shell Mie model calculation based on the BC mass concentrations derived from the Aethalometer measurements. Then, BC absorption at various wavelength ($\sigma_{abs,BC,\lambda}$) can be obtained. Therefore, only the sets of AAE_{BC} are used in the BrC absorption contribution calculation at the corresponding wavelength ranges. The detailed discussion of AAE_{BC} and BrC absorption contribution methodology have been given in the Section 2.2.3 (Line 232).

Comment 2: *Throughout the discussions, one of the main uncertainties is the calculated BC AAE, which causes the uncertainty of BrC calculation. This uncertainty is fairly large, what is the bottom line of this uncertainty and have you put any constraints from your own measurements? Has this uncertainty been included in the value reported in abstract and conclusion regarding the BrC absorption contribution?*

Response:

We certainly agree with the reviewer that the major uncertainty is originated from the calculated AAE_{BC} due to the fact that BC light absorption will vary exponentially with the change of AAE_{BC}. The uncertainty of AAE_{BC} can come from various sources, including the size, mixing states, and morphologies of the BC containing particles (Lack and Langridge, 2013; Scarnato et al., 2013). In this work, the calcualted $AAE_{BC,370–520nm}$ and $AAE_{BC,520–880nm}$ ranged from 0.59 to 0.98 and 0.81 to 1.15, respectively. These values were constrained by *in-situ* BC mass concentration, size-distribution, and mixing state measurements at the same observation site. To evaluate the uncertainty associated with the AAE_{BC} calculation, we have conducted the Monte Carlo simulations to estimate the range of uncertainty in AAE_{BC} results after considering all random errors (with 95% confidence level) possibly associated with the parameters used in the Mie calculations. The results are shown in Fig. S5a (the original Fig. S5). These uncertainties will certainly be propagated into the calculated BrC absorption contributions, too. Hence, we also estimated the corresponding uncertainties in the BrC absorption contribution results, which have been incorporated into Fig. S5 as panel b. Accordingly, the averaged lower limits of BrC absorption contributions were 26.8%±9.1% at 370 nm, 17.5%±8.1% at 470 nm, 10.1%±7.3% at 520 nm, 8.5%±5.8% at 590 nm and 5.3%±4.5% at 660 nm, respectively. The averaged upper limits of BrC absorption contribution ratios were 40.7%±7.2% at 370 nm, 29.5%±6.7% at 470 nm, 21.1%±6.2% at 520 nm, 17.3%±5.2% at 590 nm and 12.0%±4.1% at 660 nm, respectively.

To address these issues, we have added the following statement into the discussion (Line 486): "These uncertainties will certainly be propagated into the calculated BrC absorption contributions, too. Hence, we also estimated the corresponding uncertainties in the BrC absorption contribution results, as shown in Fig. S5b. Accordingly, the averaged lower limits of BrC absorption contribution ratios were 26.8%±9.1% at 370 nm, 17.5%±8.1% at 470 nm, 10.1%±7.3% at 520 nm, 8.5%±5.8% at 590 nm and 5.3%±4.5% at 660 nm, respectively, and the averaged upper limits of BrC absorption contributions were 40.7%±7.2% at 370 nm, 29.5%±6.7% at 470 nm, 21.1%±6.2% at 520 nm, 17.3%±5.2% at 590 nm and 12.0%±4.1% at 660 nm, respectively."

In the conclusion we have inserted the following statement (Line 654): "It should be noted that the calculated BrC light absorption may vary exponentially with the value of AAE_{BC}. According to Monte Carlo simulations under 95% confidence level, we found that BrC light absorption contribution ratios in this work can range roughly from 18% to 48% at 370 nm, 10% to 37% at 470 nm, 3% to 27% at 520 nm, 3% to 22% at 590 nm, and 1% to 16% at 660 nm, respectively. Therefore, proper values of AAE_{BC} have to be carefully obtained for a particular study area, especially needed to be constrained by the BC mass concentration, size-distribution, and mixing state measurements."

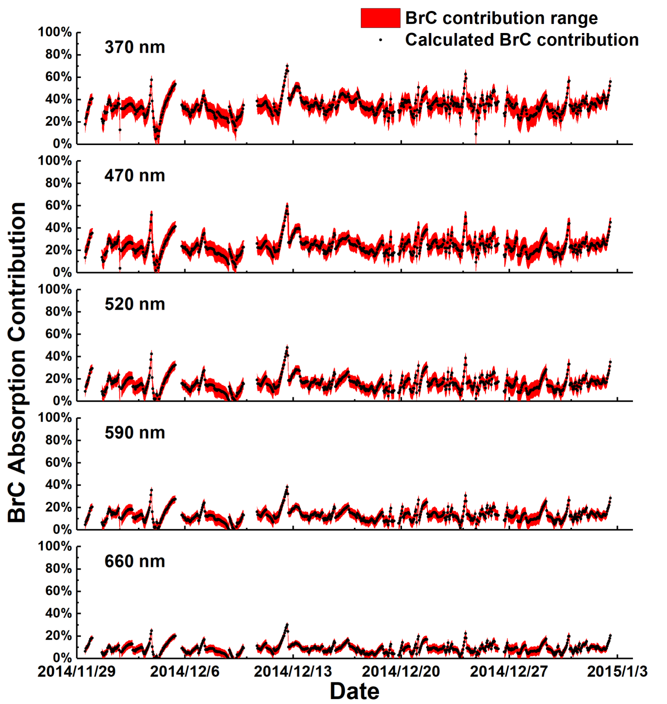

Figure S5b. Time series of the calculated BrC absorption contribution (black dots) with the uncertainty range (the red-colored shadow areas), derived from that lower- and upper-limits of AAE_{BC} obtained from the Monte Carlo simulation. The black dots were the calculated BrC absorption contribution results using the Mie model at five wavelengths, respectively.

References：

Lack, D. A., and Langridge, J. M.: On the attribution of black and brown carbon light absorption using the Ångström exponent, Atmospheric Chemistry and Physics, 13, 10535-10543, 10.5194/acp-13-10535-2013, 2013.

Scarnato, B. V., Nielsen, K., Vahidinia, S., and Richard, D.: Effect of Aggregation and Mixing on optical properties of Black Carbon, 2013,