Anonymous Referee #2:

We would like to thank the reviewer for the valuable and constructive comments, which helps us to improve the manuscript. Listed below are our responses to the comments point-by-point, as well as the corresponding changes made to the revised manuscript. The reviewer's comments are marked in black and our answers are marked in blue, and the revision in the manuscript is further formatted as '*Italics*'.

1. General comments

The present manuscript reports variations of the refractory BC (rBC) particles measured by a single particle soot photometer before/during/after APEC summit. The shell diameter of rBC-containing particles were determined according to Mie scattering theory presuming that all rBC particles were in shell-core configuration. The objective of this study is to evaluate the effect of emission control measures on the mixing state of rBC as well as their light absorbing properties, and concluded that coating matters on the rBC core decrease as a result of emission control of pollution precursors such as SO_2 and NO_2. In general, the paper is clear logically and well written; however deficiencies of this study is the absorption enhancement (E_{ab}) is estimated based on calculation, not measurement. Direct measurement of optical properties of rBC particles is essential to better understand the radiative effect of rBC containing particles in Beijing. The paper could be considered for publication after several issues are carefully clarified, as follows.

Response: We thank the reviewer for raising the important issue. Following the reviewer's suggestion, we have added the measured light absorption coefficients to compare with the calculated values (Fig. R1 in the response and new Fig. 7 in the revised manuscript).

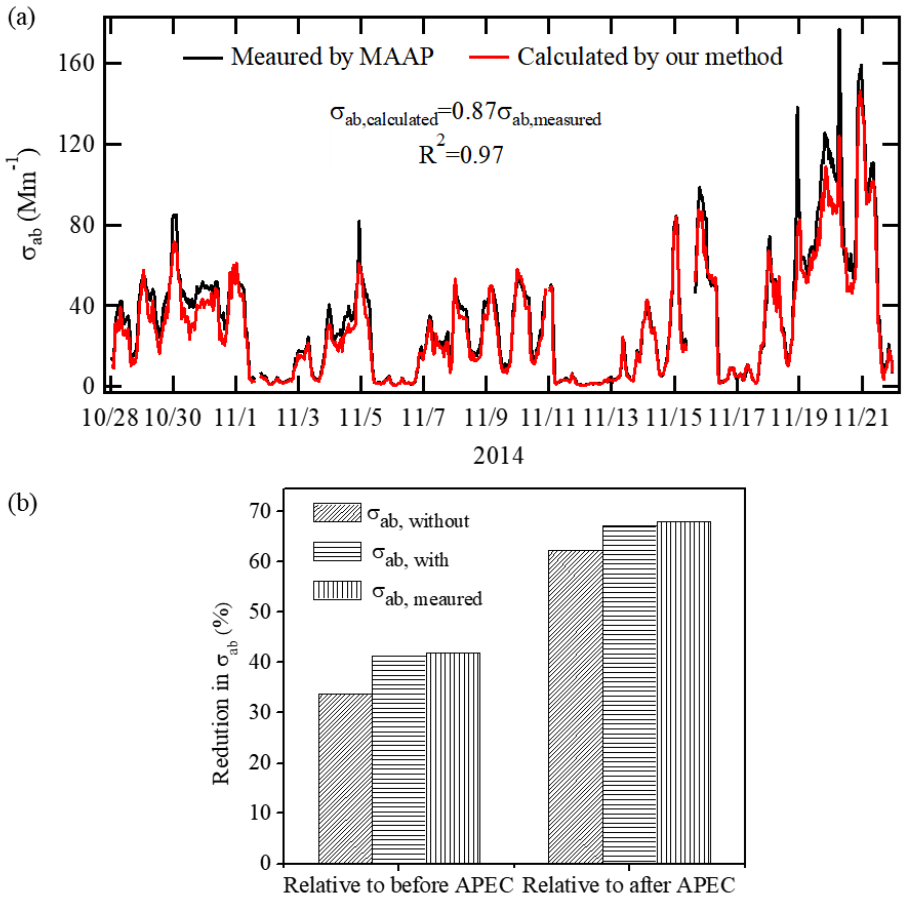

Figure R1 (new Fig. 7 in the revised manuscript). (a) The light absorption coefficient (σ_{ab}) at 670 nm. (b) Reduction in the daytime σ_{ab} of BC-containing particles observed in the pollution episode during APEC relative to that before and after APEC. The correlation between the calculated σ_{ab} ($\sigma_{ab,\ calculated}$) using Mie theory combined with SP2 measurements and the measured σ_{ab} ($\sigma_{ab,\ measured}$) by the MAAP is also shown in (a). The $\sigma_{ab,with}$ and $\sigma_{ab,without}$ values represent $\sigma_{ab,\ calculated}$ with/without, respectively, considering the differences of light-absorption capability of ambient BC-containing particles among the episodes before, during and after APEC.

Correspondingly, the related discussion has been added in the revised manuscript, as "*Figure 7a shows the measured and theoretical light absorption coefficient (σ_{ab}) of BC-containing particles during the campaign period. The measured σ_{ab} revealed that the daytime light absorption of BC-containing particles in the pollution episode during APEC decreased by ~42% and ~68% compared with those in pollution episode*

before and after APEC, respectively. This decrease could attributed to reduction in both the rBC mass concentration and the light-absorption capability of ambient BC-containing particles. In order to separate the contributions of a decrease of rBC mass concentration and a weakening of BC light-absorption capability to the reduction in light absorption during APEC, we calculated the theoretical reduction in σ_{ab} *of BC-containing during APEC with and without considering the weakened light-absorption capability of BC-containing particles due to emission reduction (*$\sigma_{ab,with}$ *and* $\sigma_{ab,without}$ *respectively). When considering the simultaneous reduction in the mass concentration and light-absorption capability of BC, the calculated reduction in daytime* σ_{ab} *of BC-containing during APEC related to non-APEC period showed a good agreement with ones obtained from MAAP measurements (Fig. 7b). This agreement demonstrated that the decrease in the light absorption of BC-containing particles depended not only on the reduction of BC mass concentration, but also on the weakening of their light-absorption capability.*"

2. Specific comments

(1) Page 4 line 16: The authors assumed that all the rBC containing particles were in spherical shape, please make sure they are true or not. As known, on-road vehicle emission is one of the important sources for rBC particles in Beijing mega city, freshly emitted rBC may present in non-spherical shape, and they may turn to be spherical with ageing process. The authors are suggested to check the number size distribution and delay time (delta_t) to clarify this.

Response: Thanks to the reviewer to point this out. Following the reviewer's suggestion, we have checked the number distribution of rBC cores (D_c) and whole BC-containing particles (D_p) (Fig. R2 in the response) to clarify the reasonability of spherical assumption used in Mie calculation in this study. Noted that we focused on investigating the BC-containing particles during pollution episodes. Figure R2 shows that the number distribution of D_p for BC-containing particles

exhibited a peak at 200-400 nm during the pollution episodes, significantly larger than the peak value (D_c of ~95 nm) for number size distribution of bare rBC cores. This revealed fully aged BC-containing particles under polluted conditions. In our previous study (Zhang et al., 2016), we found that the thickly coated BC particles in the north china plain (including Beijing) exhibited near-spherical shape. Therefore, the spherical assumption used in the Mie calculation in this study was reasonable.

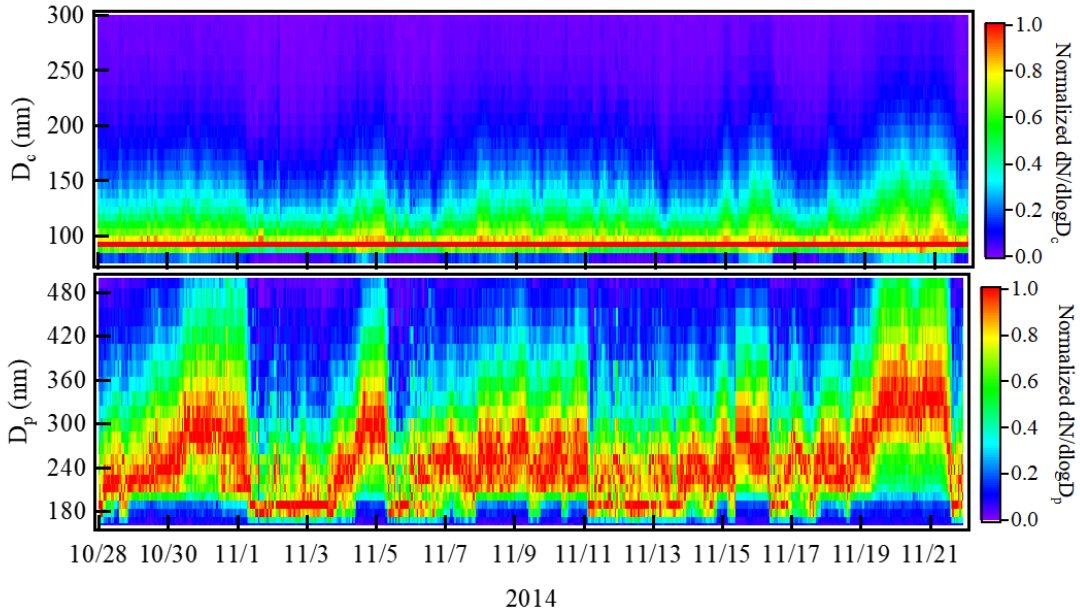

Figure R2 (Fig. 1 in the revised manuscript). Time series of the number size distribution of rBC cores (D_c) and whole BC-containing particles (D_p).

To make this point clear, we have added the Fig. S1 in the supplement and the related statement was "*In this study, we focused on investigating the BC-containing particles during pollution episodes. Under polluted conditions, we have found fully aged BC-containing particles in Beijing, China (Zhang et al., 2018). In our previous study (Zhang et al., 2016), we found that the thickly coated BC particles in the north china plain (including Beijing) exhibited near-spherical shape and a core-shell structure used in the Mie calculation was reasonable.*"

(2) Page 5 section 2.3.2: As mentioned, Eab is determined on Mie theory, not measurements. Please point out the uncertainty of such calculation related to RI values. Adding an uncertainty in Figure 6 is encouraged.

Response: Thanks for the comments. Following the reviewer's suggestion, we have point out the uncertainty (~10%) of Mie calculation related to refractive index (RI). In this study, we used the RI values of coating materials and rBC cores were 1.5-0i and 2.26-1.26i, respectively, which were same with the ones used in our previous study (Zhang et al., 2018). In that work, we have estimated the uncertainty of ~10% from the assumptions of *RI* in Mie calculation. To make this point clear, we have added the related discussion in the revised manuscript, as "*The uncertainty of Mie calculation from the assumptions was estimated to be ~10% in our previous work (Zhang et al., 2018).*"

Correspondingly, we have added the uncertainty in the new Fig. 3 and Fig. 6 (Fig. R3 and Fig. R4 in the response). The related statements was also added in the captions.

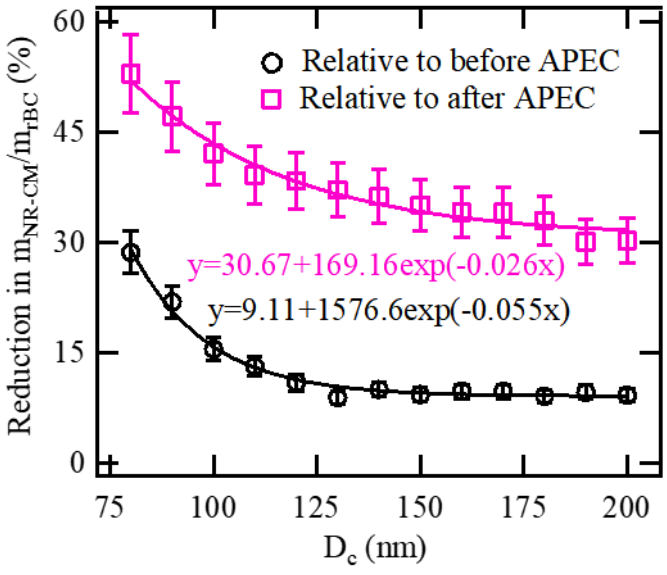

Figure R3 (new Fig. 3b in the revised manuscript). The reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles during APEC relative to that before and after APEC. The error bar shown in (b) represents the uncertainties (~10%) of $m_{NR\text{-}CM}/m_{rBC}$ ratio from

Mie calculation.

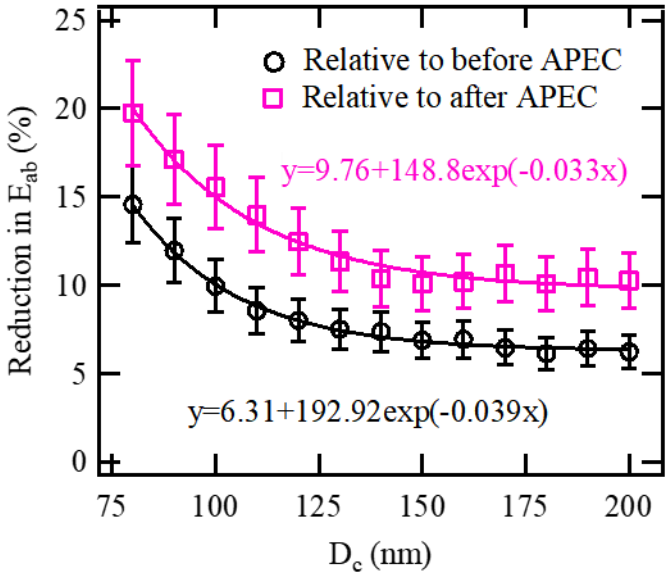

Figure R4 (new Fig. 6b in the revised manuscript). The reduction in E_{ab} of BC-containing particles during APEC relative to that before and after APEC. The error bar shown in (b) represents the uncertainties (~15%) of E_{ab} from Mie calculation.

(3) Page 6 line 11: The authors attribute the decrease in ambient rBC and NO2 on the emission controls. It is encouraged to give more discussion show diurnal variations of their concentration (Sun, Y. et al. "APEC Blue": Secondary Aerosol Reductions from Emission Controls in Beijing. Sci. Rep. 6, 20668; doi: 10.1038/srep20668 (2016), as well as back trajectories of air mass during APEC and non-APEC period to support it.

Response: Thanks for the comments. Following the reviewer's suggestion, we have given more discussion showing diurnal variations of rBC and NO_2 concentration (Fig. R5 in the response and Fig. S4 in the supplement), as well as back trajectories of air mass during APEC and non-APEC period (Fig. R6 in the response and Fig. S3 in the supplement) to support the decrease in ambient rBC,

NO_2 due to the emission controls. The related discussion was added "*Figure S4 shows that the diurnal variations of the rBC, NO_2 and SO_2 concentration and the PBL during the pollution episodes before, during and after APEC. Comparing the diurnal variations between the rBC concentration and the PBL revealed that the rBC concentrations during the pollution episodes were dominated by the PBL. However, the precursor concentration of secondary aerosol (i.e., NO_2 and SO_2) during the pollution episodes exhibited different diurnal variations with a peak at noontime and early afternoon, which was most likely attributed to regional transport. The back-trajectory analysis (Fig. S3) revealed that the air mass during the pollution episodes was mainly from polluted regions (i.e., Hebei and Tianjin). This indicated that regional emission controls would reduce the pollutant (i.e., rBC, NO_2 and SO_2) concentration in Beijing under polluted conditions. The Sun et al., (2016) has demonstrated significant reductions in the precursors of secondary aerosol during APEC compared to those in non-APEC period due to emission controls during over a regional scale (i.e., Beijing and adjacent areas). The similar PBL (Fig S4) during the pollution episodes before, during and after APEC further identified the important contribution of emission reduction to the decrease of rBC, NO_2 and SO_2 concentration during APEC.*"

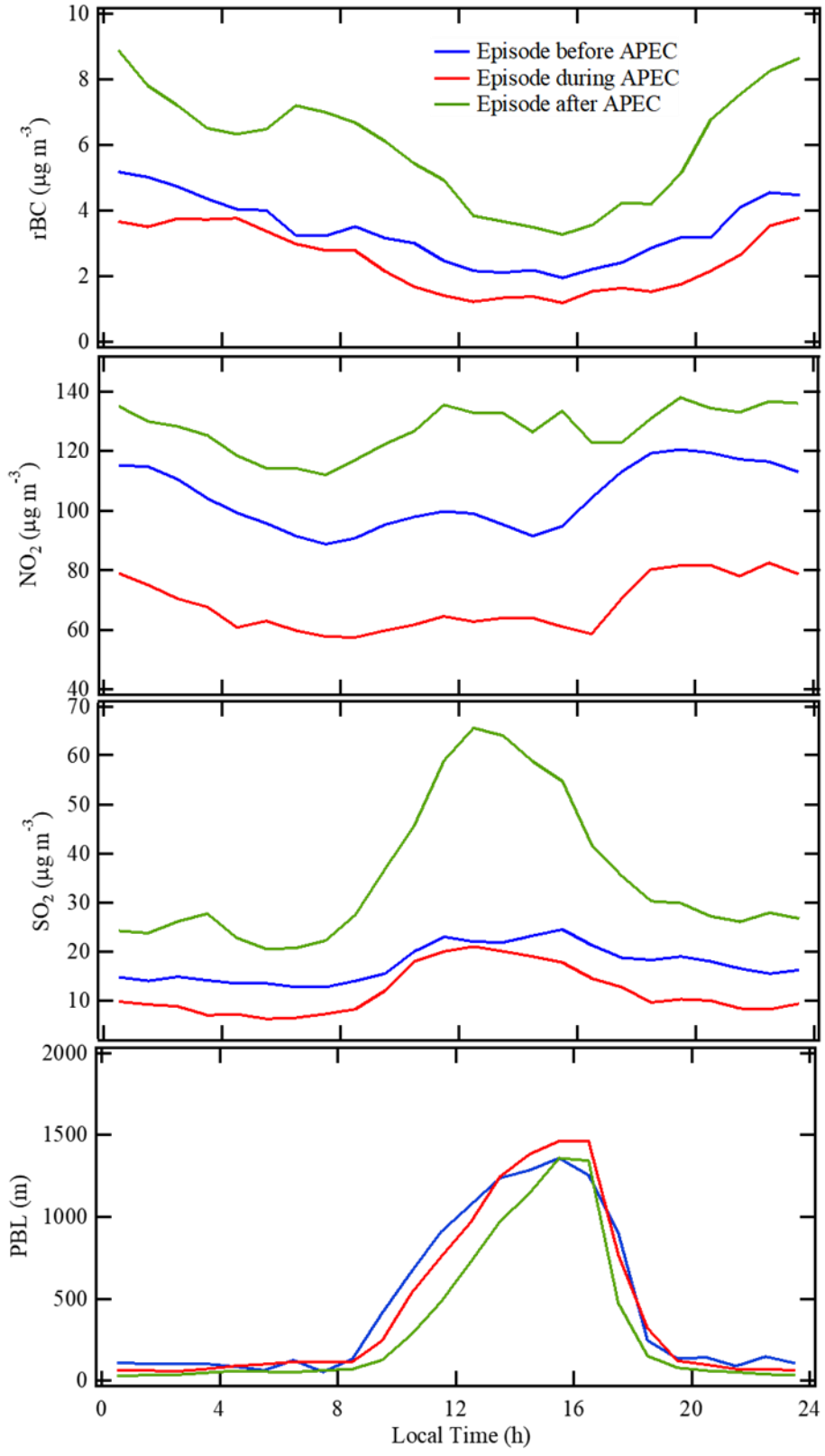

Figure R5 (Fig. S4 in the revised manuscript). Diurnal variations of the rBC, NO_2 and SO_2 concentration and the PBL for the pollution episodes before, during and after APEC.

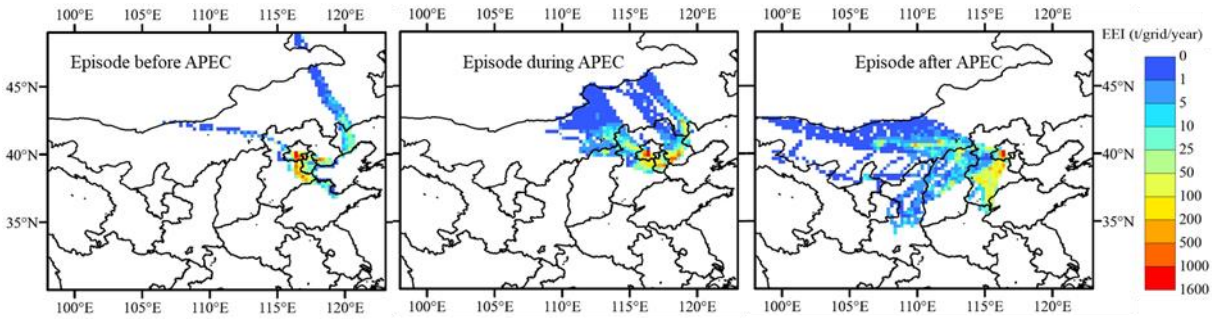

Figure R6 (Fig. S3 in the revised manuscript). Spatial distribution (0.25°×0.25°) of the effective emission intensity (EEI, defined by Lu et al. (2012)) for BC transported to the observation site (40º00'17" N, 116º19'34" E) for the pollution episodes before, during and after APEC. The EEI calculation was stated in our previous study (Zhang et al. 2018).

(4) Page 6 line 24: It is hard to understand the meaning of the sentence "The similar mode of … and similar atmospheric processes (coagulation and wet removal) for rBC particles … APEC." The predominance of rBC in ~100 nm range is mostly due to incomplete combustion processes (vehicle engine etc.). It is better to remove it.

Response: Thanks. Following the reviewer's suggestion, we have removed it.

(5) Page 7 line 4: "condensational growth was more effect". Please provide more information to support such statement.

Response: Thanks to the reviewer for raising this concern. Sorry for inappropriate statement. We have revised the sentence as "*The size-dependent $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles revealed that particle growth was more effective for smaller particles, which followed the diffusion-controlled growth law (Seinfeld and Pandis 2006).*"

(6) In the third paragraph, Where is "Fig. 4b"? What is the reason for the difference in diurnal variability of Dc before and after APEC period?

Response: Thanks to the reviewer for raising these questions. Fig. 4b is described in the fourth paragraph, as "*However, the reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of*

BC-containing particles for the pollution episode during APEC compared to that after APEC showed a different diurnal cycle, with maxima at ~10:00-12:00 LT and with minima at ~15:00-17:00 LT (Fig. 4b)…."

To the second question, sorry for the misleading and Figure 4(a) and (b) do not show the difference in diurnal variability of Dc before and after APEC period. Figure 4 shows the difference in diurnal variability of the reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles with size-resolved D_c during APEC compared with those before (Fig. 4(a)) and after (Fig. 4(b)) APEC period. The difference was mostly likely to be attributed to different formation mechanism of BC coating materials before and after APEC. In the pollution episode before APEC, the production of BC coating materials was dominated by the photochemistry. However, the effect of other processes (e.g., heterogeneous chemistry) on the production of BC coating materials were enhanced during the pollution episode after APEC. We have discussed this point in the third and fourth paragraphs of page 7 in the manuscript.

References:

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