

Anonymous Referee #1:

The authors measured rBC particles with a single particle soot photometer during APEC, and discussed the effects of multi-pollutant emission reductions on the BC light absorption. The results are interesting, and the science of this work sounds good. However there some potential issues with the data analysis and conclusions. Moreover, the English needs to be further improved. I believe this manuscript can be considered for publication in ACP after minor revision.

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. Page 2 Lines 17–18: Previous studies have demonstrated that a majority of the freshly emitted BC particles are internally mixed from biomass burning emissions (e.g., www.sciencedirect.com/science/article/pii/S135223101830133X). Thus, this expression should be revised.

Response: Thanks. We have revised the sentence as “*When fresh BC particles are emitted from incomplete combustion (e.g., traffic emission) other than biomass burning (Wang et al., 2018; Pan et al., 2017), they are most likely externally mixed with other aerosol components.*”

2. Page 3 Lines 26-27: This sentence is hard to understand. How long is the BC lifetime that the authors considered in this study?

Response: Thanks for the comment. In this study, we defined the transition periods (November 3-5 and 13-16, 2014) considering the lifetime (~3 d) of BC during the campaign period. Figure R1 in the response (Fig. S2 in the revised manuscript) shows a similar distribution of effective emission intensity (EEI,

defined by Lu et al. (2012)) of BC over the site during 3, 5 and 7 days, revealing that the BC transported to the site was mainly from emission within 3 days. The EEI analysis indicated that the BC particles over the site during the campaign period have a lift time of ~ 3 d. For “APEC blue”, the emission control measures were implemented on November 3-12, 2014. Considering the ~ 3 d lifetime, the BC transported to the site during the pollution episode on November 3-5 were the mixtures of particles that were emitted before and during APEC. Similarly, the BC transported to the site during the pollution episode on November 13-16 were the mixtures of particles that were emitted during and after APEC. To clearly distinguish BC characteristics with and without emission control measures, we exclude this two periods (November 3-5 and 13-16, 2014) in this study.

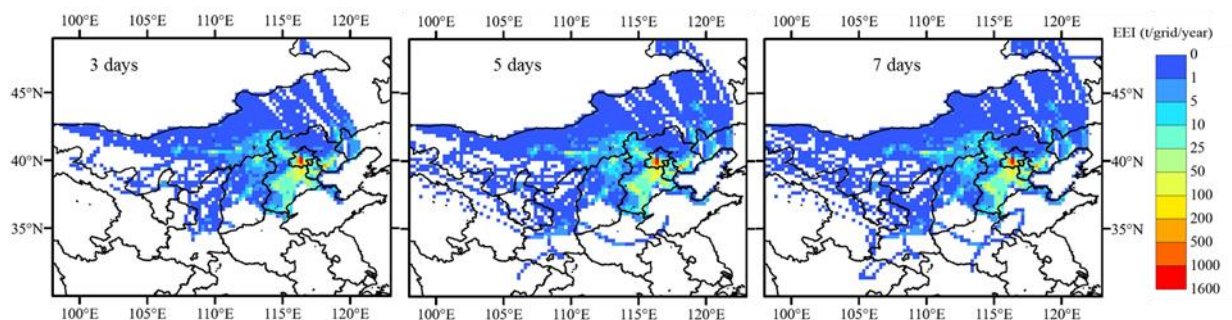


Figure R1 (Fig. S2 in the revised manuscript). Spatial distribution ($0.25^\circ \times 0.25^\circ$) of the effective emission intensity (EEI, defined by Lu et al. (2012)) for BC transported to the observation site ($40^\circ 00' 17''$ N, $116^\circ 19' 34''$ E) based on 3 days, 5 days and 7 days back-trajectory. The EEI calculation was stated in our previous study (Zhang et al. 2018).

To make this point clear, we have revised the sentence as “*and two transition periods (November 3-5 and 13-16, 2014), which were not discussed in this work considering that we could not distinguish the BC particles transported to the site during these days characterized by enforcement of emission control measures or not (Fig. S2 and the associated discussion in the supplementary information).*”

3. Page 4 Lines 25-27: The ratio of m_{NR-CM}/m_{rBC} was calculated based on several assumptions, e.g, RI, density, and core-shell structure, thus, it is better to add some discussion about uncertainties of this method.

Response: Thanks to the reviewer for raising this concern. Following the reviewer’s suggestion, we have add some discussion about uncertainties of Mie calculation. In our previous work (Zhang et al., 2018), we have estimated the uncertainties from the assumptions in Mie calculation. These assumptions were also used in this work to calculate the m_{NR-CM}/m_{rBC} . Following the reviewer’s suggestion, we have added the related discussion in the revised manuscript, as “*The uncertainty of Mie calculation from the assumptions (e.g., RI, density and core-shell structure) was estimated to be ~10% in our previous work (Zhang et al., 2018).*”

Correspondingly, we have added the uncertainty in the new Fig. 3 (Fig. R2 in the response). The related statements was also added in the caption.

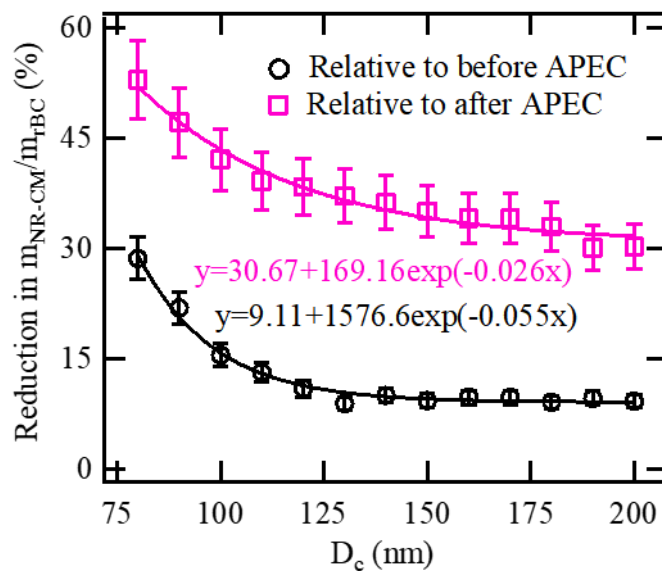


Figure R2 (new Fig. 3b in the revised manuscript). The reduction in m_{NR-CM}/m_{rBC} ratio of BC-containing particles during APEC relative to that before and after APEC. The error bar shown in (b) represents the uncertainties (~10%) from Mie calculation.

4. Page 5 Lines 10–14: According to Zhang et al. (2018), the authors also used AE33 measurements during this campaign. It is better to compare the calculated light absorption coefficients (based on Eq. 3) with the AE33 measured values.

Response: Thanks for the comment. Following the reviewer’s suggestion, we have compared the calculated light absorption coefficients with the measured values (Fig. R3 in the response). However, we used the MAAP measurements instead of AE33 measurements, because the AE33 measurement was not conducted before 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.”*

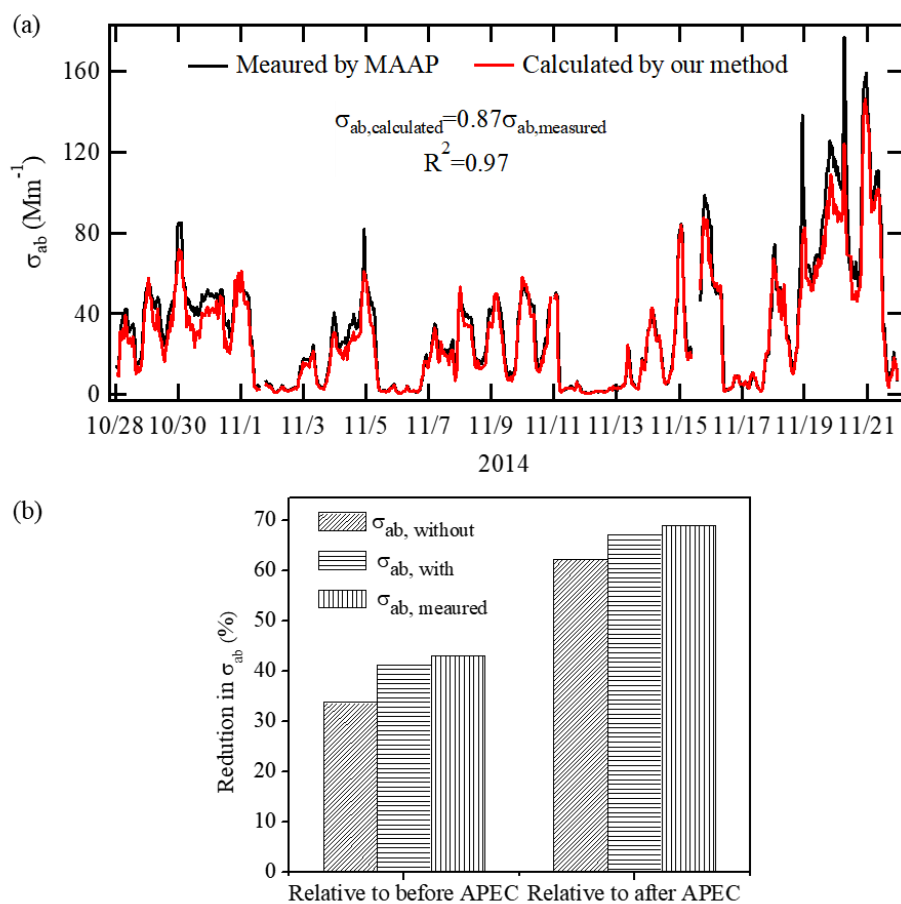


Figure R3 (new Fig. 7 in the revised manuscript). (a) The light absorption coefficient (σ_{ab}) at 670 nm. (b) Reduction in the absorption coefficients (σ_{ab}) of BC-containing particles observed in the pollution episode during APEC relative to that before and after APEC. The correlation between the 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.

5. Page 5 Lines 17–18: What’s the standard used to define the pollution episodes?

Response: We thank the reviewer for raising this question. Following Sun et al. (2016), the pollution episodes were briefly separated by clean days through the study period (Fig. 1 in the manuscript). Air flows were predominately southerly and meteorological conditions were generally stagnant (low wind speed and high relative humidity) during these episodes. Because emission controls were mainly implemented in cities to the south and east of Beijing, we use these episodes to study the impact of regional emission controls on aerosol chemistry in megacity Beijing. This approach isolates the influences of clean periods with air masses from the north and northwest where far fewer emission controls were implemented.

To make it clear, the related statement has been added in the revised manuscript, as *“Three pollution episodes (briefly separated by clean days) on October 28–November 1, November 6–11 and November 17–21 were observed before, during and after APEC, respectively. Following APEC study in Sun et al. (2016), we focused on comparing the BC characteristics among the pollution episodes to investigate the effect of emission reduction. During the three pollution episodes, the air masses over the site were mainly from the south and east of Beijing (Fig. S3) where emission control measures were implemented during APEC. On the other hand, the pollution episodes in Beijing were characterized by low wind speed and planetary boundary layer, as well as high relative humidity (Sun et al. 2016; Zheng et al. 2015).”*

6. Page 6 Lines 5–6: The emission control can reduce the concentrations of NO₂ and SO₂. That’s right, and a lot of APEC publications have demonstrated. However, the authors can not obtain the statement of BC coating materials being affected by emission controls from this, though the authors prove this may be right in the following discussion (Fig. 5).

Response: Thanks for the comments. Here, we stated that reduction in the concentrations of NO₂ and SO₂ might affect the production of BC coating

materials. Whether the reduction of precursor of secondary aerosols (e.g., NO₂ and SO₂) will affect the coating materials on the BC is complex, which not only depends on the decrease in BC amount versus secondary aerosols but also controlled by secondary components condensed on BC-containing versus non-BC containing particles. As expected, in the following section (3.2), we found the reduction in BC coating materials during APEC compared with that before/after APEC based on SP2 measurement.

To make it clear, we have revised the sentence as “*Previous studies have identified a reduction in the concentrations of secondary components (e.g., sulfate and nitrate) in aerosols during APEC compared to that before and after APEC (Zhang et al., 2016a; Han et al., 2015). However, the change of coating materials on the BC due to the reduction of secondary components was complex, which not only determined by the decrease in BC versus secondary components, but also depend on secondary components condensed on BC-containing versus non-BC particles.*”

7. Page 6 Lines 7-9: It's better to add previous studies in Beijing (e.g, <http://iopscience.iop.org/article/10.1088/1748-9326/aa64ea/meta>) to explain the importance of photochemical reactions in BC aging process.

Response: Thanks for the suggestion. Following the reviewer's suggestion, we have added previous studies in Beijing (Wang et al., 2017; Metcalf et al., 2013; Zhang et al., 2014; Peng et al., 2016) to explain the importance of photochemical reactions in BC aging process. The related discussion was as “*Previous studies have pointed out the importance of photochemical reactions in BC aging process (Wang et al., 2017; Metcalf et al., 2013; Zhang et al., 2014; Peng et al., 2016), indicating that changing the daytime concentrations of rBC and coating precursors might play a more important role in affecting BC aging than altering the nighttime concentrations.*”

8. Page 6 Lines 7-19: The large reductions in the daytime levels of rBC and coating

precursors may be due to the higher boundary layer, which favors diffusion of pollutants. How to evaluate the impacts of meteorological conditions and emission reductions on the rBC coatings?

Response: We thank the reviewer for raising this question. In order to segregate possible meteorological effects and to quantify approximately the influence of the emission reductions on BC coatings, we picked up observational data during pollution episodes before, during and after APEC, in which the local meteorological conditions were similar. On the other hand, we have given diurnal variations of the rBC, NO₂ and SO₂ concentration and the PBL for the pollution episodes before, during and after APEC (Fig. R4 in the response and Fig. S4 in the supplement), as well as back trajectories of air mass during APEC and non-APEC period (Fig. R5 in the response and Fig. S3 in the supplement) to support the decrease in ambient rBC, NO₂ due to the emission controls.

The related discussion was added “*Figure S4 shows that the diurnal variations of the rBC, NO₂ and SO₂ 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., NO₂ and SO₂) 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, NO₂ and SO₂) 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, NO₂ and SO₂ concentration during APEC.”

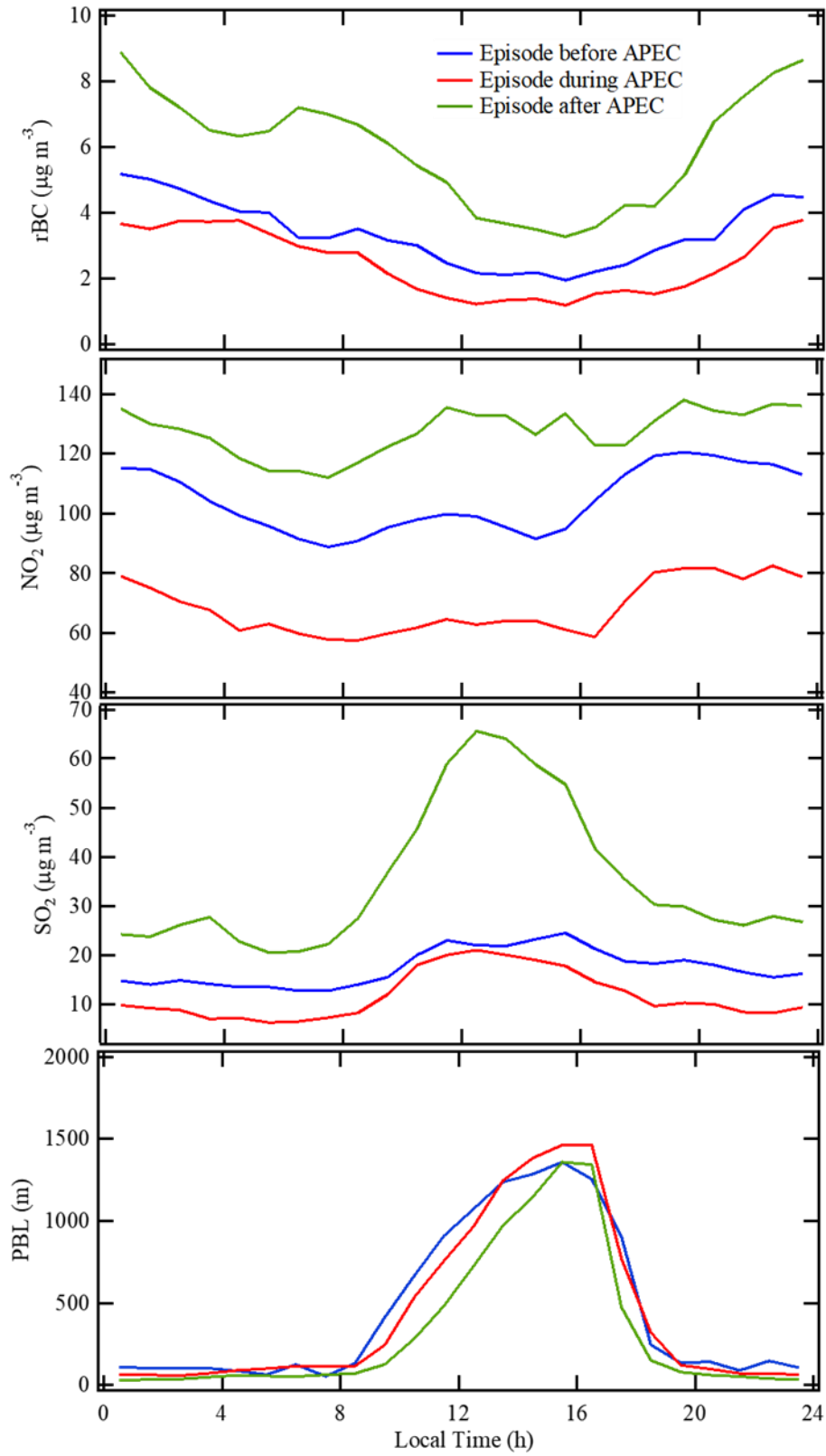


Figure R4 (Fig. S4 in the revised manuscript). Diurnal variations of the rBC, NO₂ and SO₂ concentrations and the PBL for the pollution episodes before, during and

after APEC.

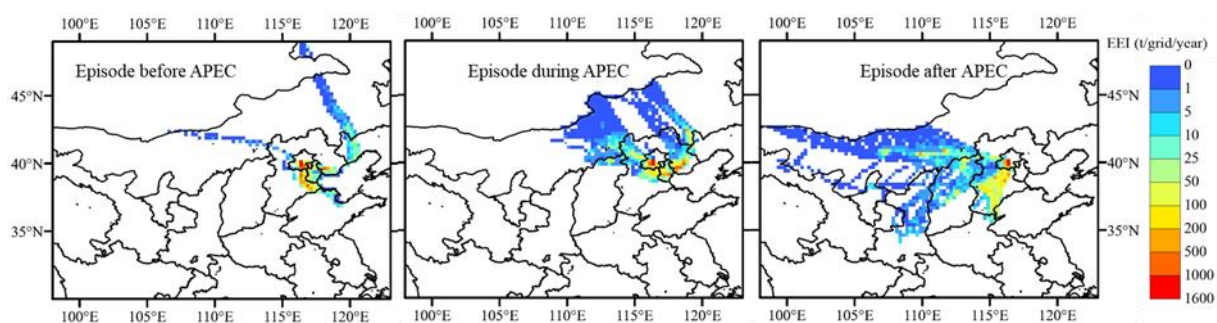


Figure R5 (Fig. S3 in the revised manuscript). Spatial distribution ($0.25^{\circ} \times 0.25^{\circ}$) of the effective emission intensity (EEI, defined by Lu et al. (2012)) for BC transported to the observation site ($40^{\circ}00'17''$ N, $116^{\circ}19'34''$ E) for the pollution episodes before, during and after APEC. The EEI calculation was stated in our previous study (Zhang et al. 2018).

9. Page 6 Line 23-24: The similar rBC core size during different periods may be due to the similar emission sources, but the statement of “similar atmospheric processes” is not right. The atmospheric processes not only affect rBC core but also the BC-containing particles. Thus, this statement should be reworked.

Response: Thanks to the reviewer to point this out. Following the reviewer’s suggestion, we have deleted the statement of “similar atmospheric processes” and the sentence was revised as “*The similar modes of the rBC cores could have resulted from similar emission sources for BC-containing particles observed before, during and after APEC.*”

10. Page 6 Lines 28-32: Based on the AMS measurements in Beijing, primary organic aerosol (POA) is also an important species. Therefore, POA may be also an important contributor to BC coatings.

Response: Thanks for the comment. We have revised the sentence in the revised manuscript, as “*In the pollution episodes before and after APEC, the particle size of the whole BC-containing particles exhibited sustained growth from ~ 180 nm to ~ 320 and ~ 400 nm, respectively, which could be attributed to the gradual*”

coagulation and condensation of other species (i.e., primary aerosol and secondary components) on the BC surface.”

11. Page 8 Lines 20–21: Here it would be good to know if the reductions are statistically significant.

Response: We thank the reviewer for raising this question. Following the reviewer’s suggestion, we have shown the statistical results in the new Fig. 6 (Fig. R6 in the response).

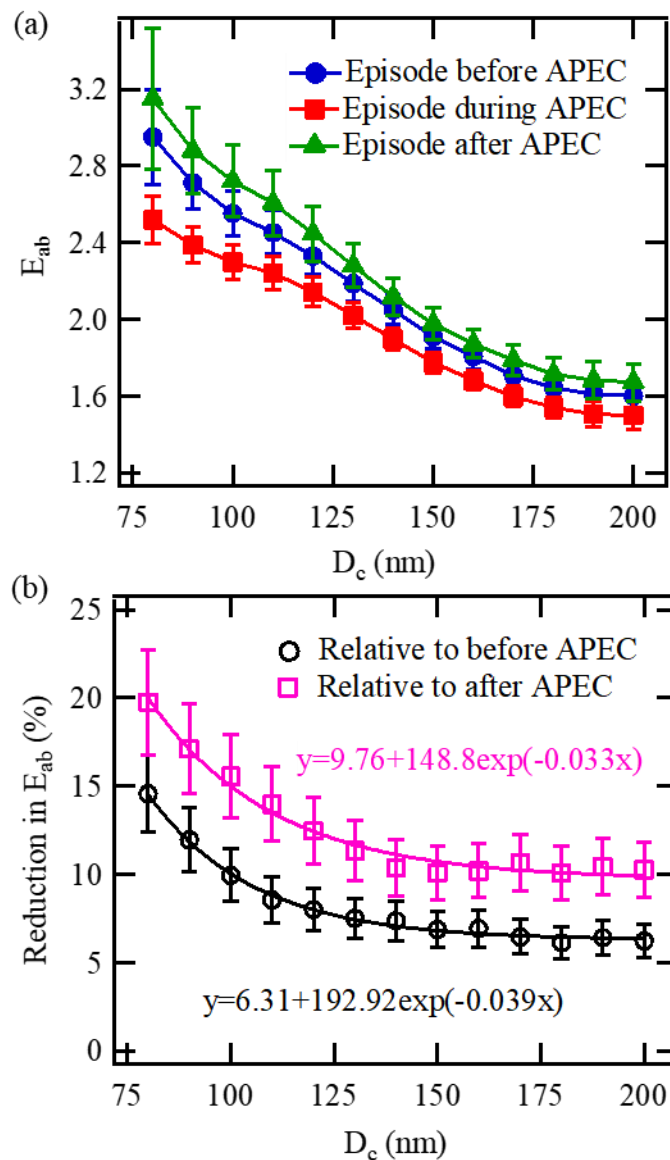


Figure R6 (new Fig. 6 in the revised manuscript). Comparison of the light-absorption capability of BC-containing particles during the day for the

pollution episodes before, during and after APEC: (a) light absorption enhancement (E_{ab}) of BC-containing particles and (b) 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.

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