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 implement 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.

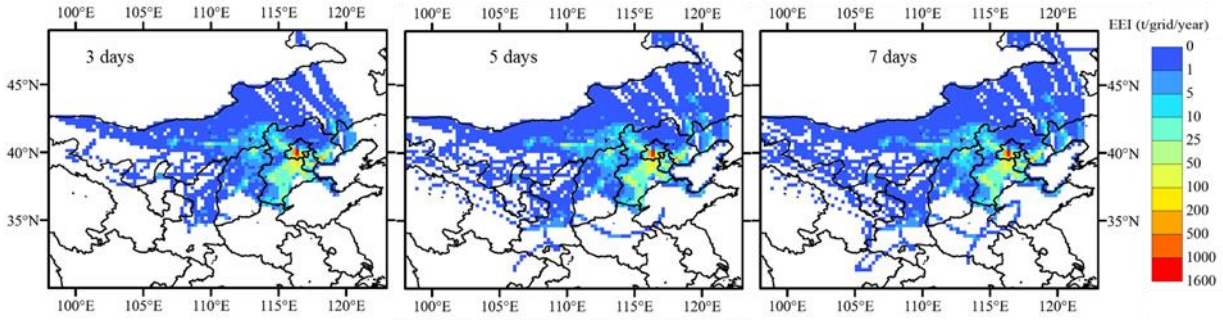

Figure R1 (Fig. S2 in the revised manuscript). Spatial distribution (0.25°×0.25°) of the effective emission intensity (EEI, defined by Lu et al. (2012)) for BC transported to the observation site (40º00'17" N, 116º19'34" E) based on 3 days, 5 days and 7 days back-trajectory. The EEI calculation was stated in our previous study (Zhang et al. 2018).

To make this point clear, we have revised the sentence as "a*nd 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 mNR-CM/mrBC 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 $m_{NR\text{-}CM}/m_{rBC}$. 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.

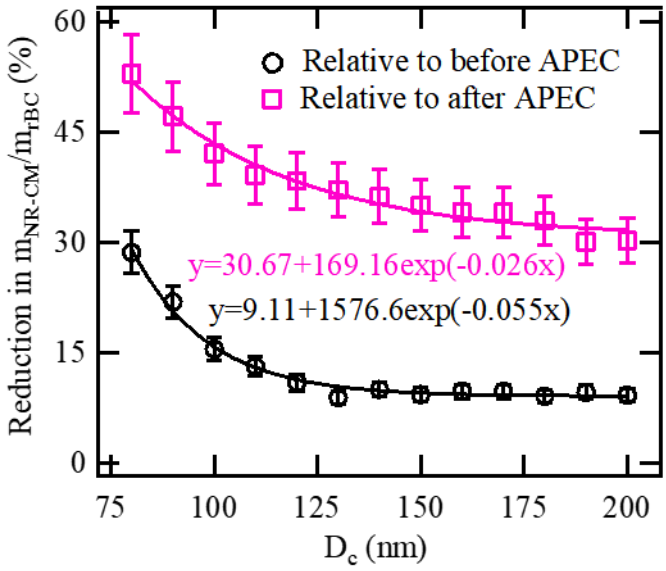

Figure R2 (new Fig. 3b in the revised manuscript). The reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles during APEC relative to that before and after APEC. The error bar shown in (b) represents the uncertainties (~10%) from Mie calculation.

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.*"

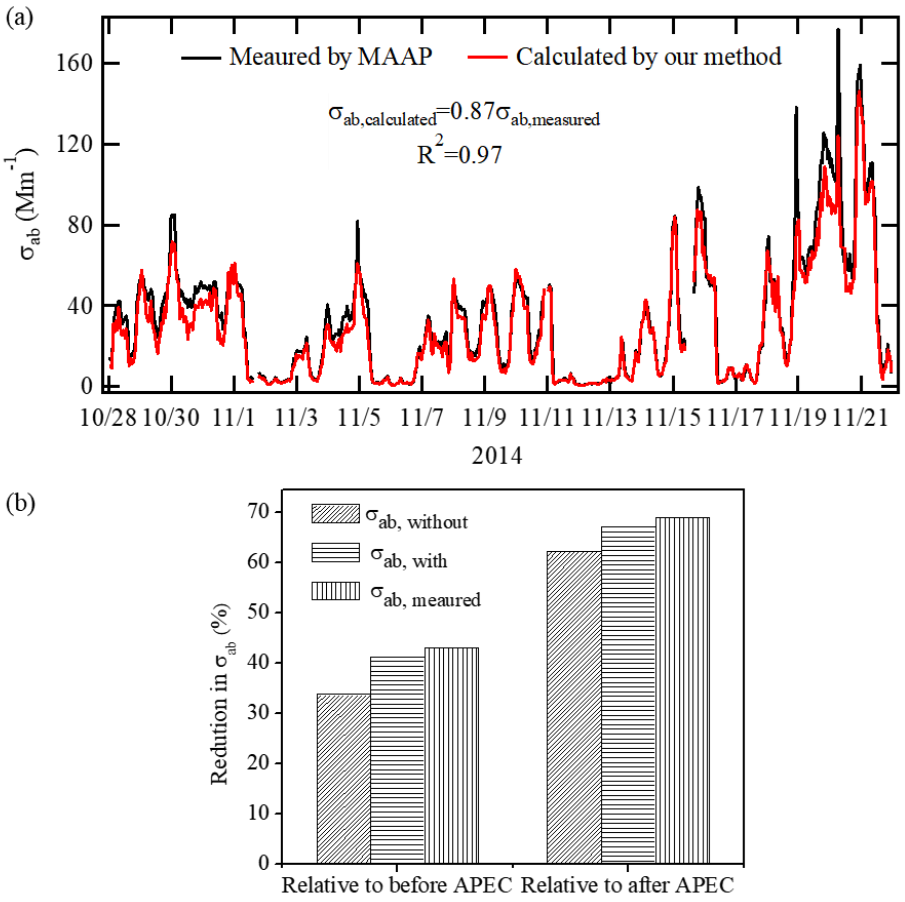

Figure R3 (new Fig. 7 in the revised manuscript). (a) The light absorption coefficient (σ_{ab}) at 670 nm. (b) Reduction in the absorption coefficients (σ_{ab}) of BC-containing particles observed in the pollution episode during APEC relative to that before and after APEC. The correlation between the 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 NO_2 and SO_2 might affect the production of BC coating materials. Whether the reduction of precursor of secondary aerosols (e.g., NO_2 and SO_2) 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, NO_2 and SO_2 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, 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.*"

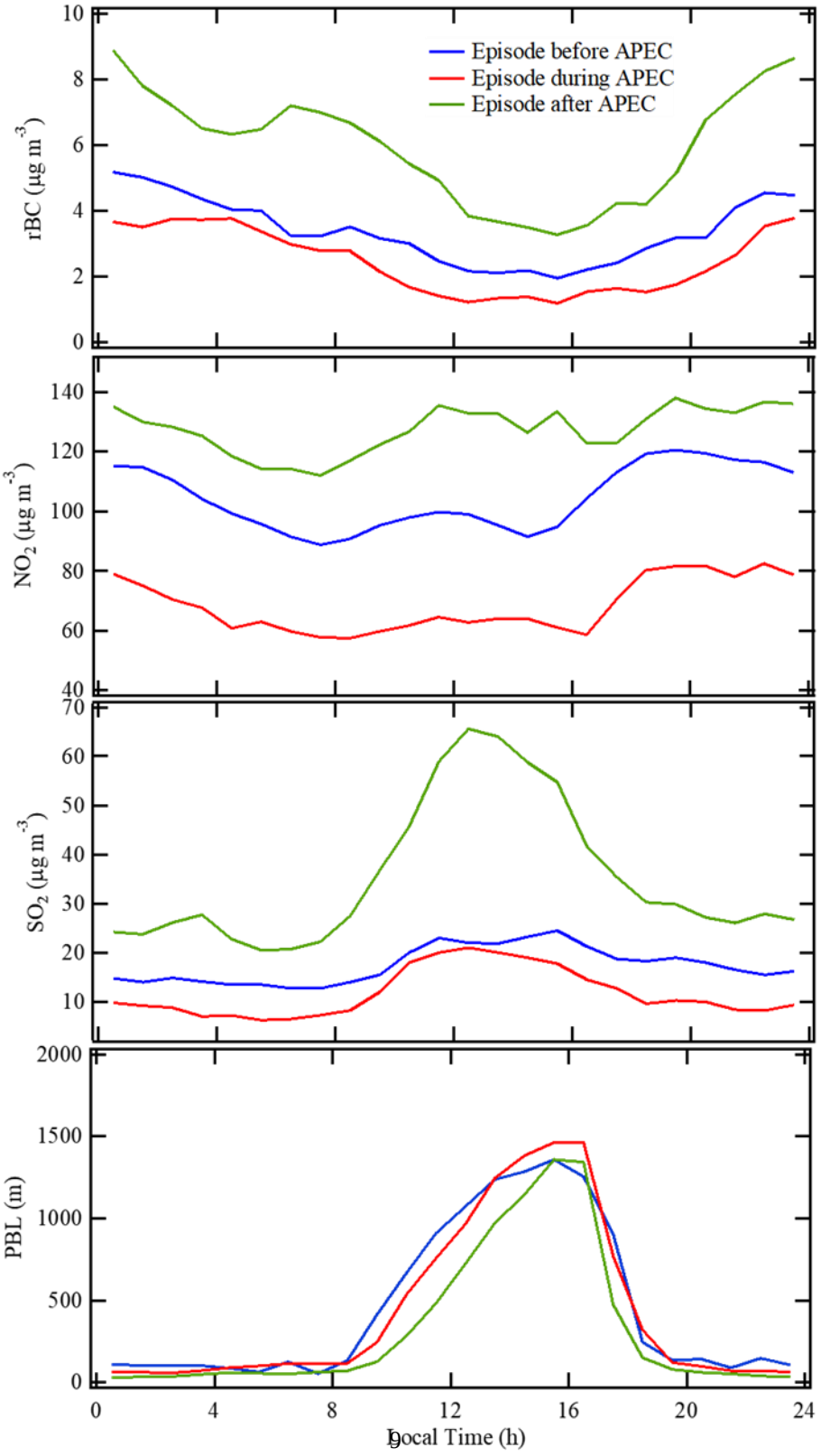
Episode before APEC
Episode during APEC
Episode after APEC
rBC (μg m-3)
NO2 (μg m-3)
SO2 (μg m-3)
PBL (m)
Local Time (h)

Figure R4 (Fig. S4 in the revised manuscript). Diurnal variations of the rBC, NO_2 and SO_2 concentrations and the PBL for the pollution episodes before, during and after APEC.

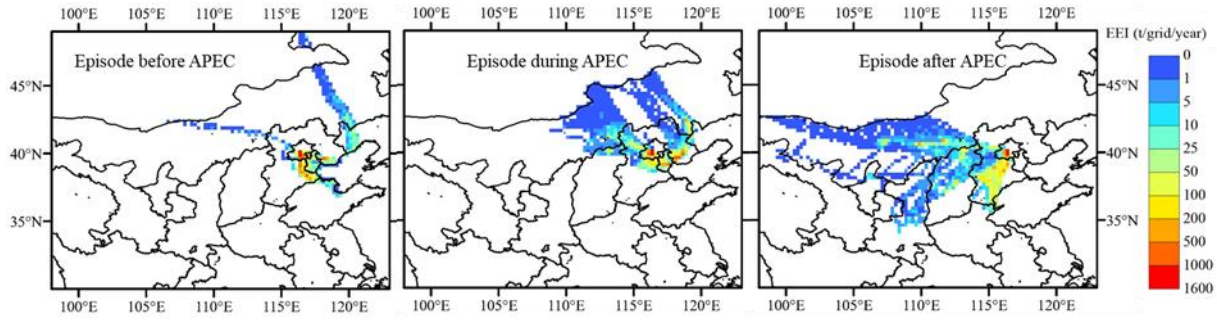

Figure R5 (Fig. S3 in the revised manuscript). Spatial distribution (0.25°×0.25°) of the effective emission intensity (EEI, defined by Lu et al. (2012)) for BC transported to the observation site (40º00'17" N, 116º19'34" E) for the pollution episodes before, during and after APEC. The EEI calculation was stated in our previous study (Zhang et al. 2018).

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).

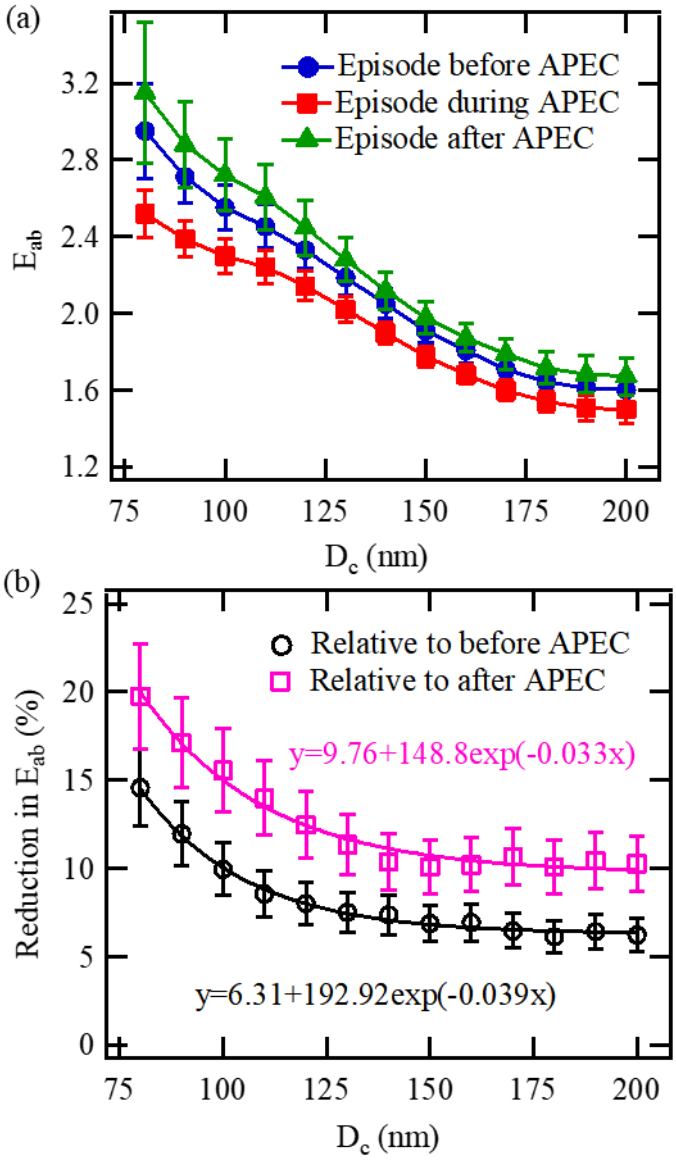

Figure R6 (new Fig. 6 in the revised manuscript). Comparison of the light-absorption capability of BC-containing particles during the day for the pollution episodes before, during and after APEC: (a) light absorption enhancement (E_{ab}) of BC-containing particles and (b) the reduction in E_{ab} of BC-containing 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.

References:

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Wang, Q., Huang R., Zhao, Z., Cao, J., Ni, H., Tie, X., Zhu, C., Shen, Z., Wang, M., Dai, W., Han, Y., Zhang, N., and Prévôt, A.: Effects of photochemical oxidation on the mixing state and light absorption of black carbon in the urban atmosphere of China, *Environ. Res. Lett.*, 12, 044012, 2017.

Zhang, G., Bi, X., He, J., Chen, D., Chan, L. Y., Xie, G., Wang, X., Sheng, G., Fu, J., and Zhou, Z.: Variation of secondary coatings associated with elemental carbon by single particle analysis, *Atmos. Environ.*, 92, 162-170, 2014.

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Zhang, Y., Zhang, Q., Cheng, Y., Su, H., Li, H., Li, M., Zhang, X., Ding, A., and He, K.: Amplification of light absorption of black carbon associated with air pollution, *Atmos. Chem. Phys. Discuss.*, https://doi.org/10.5194/acp-2017-983, in review, 2018.

Zheng, G. J., Duan, F. K., Su, H., Ma, Y. L., Cheng, Y., Zheng, B., Zhang, Q., Huang, T., Kimoto, T., Chang, D., Pöschl, U., Cheng, Y. F., and He, K. B.: Exploring the severe winter haze in Beijing: the impact of synoptic weather, regional transport and heterogeneous reactions, *Atmos. Chem. Phys.*, 15, 2969-2983, 2015.

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).

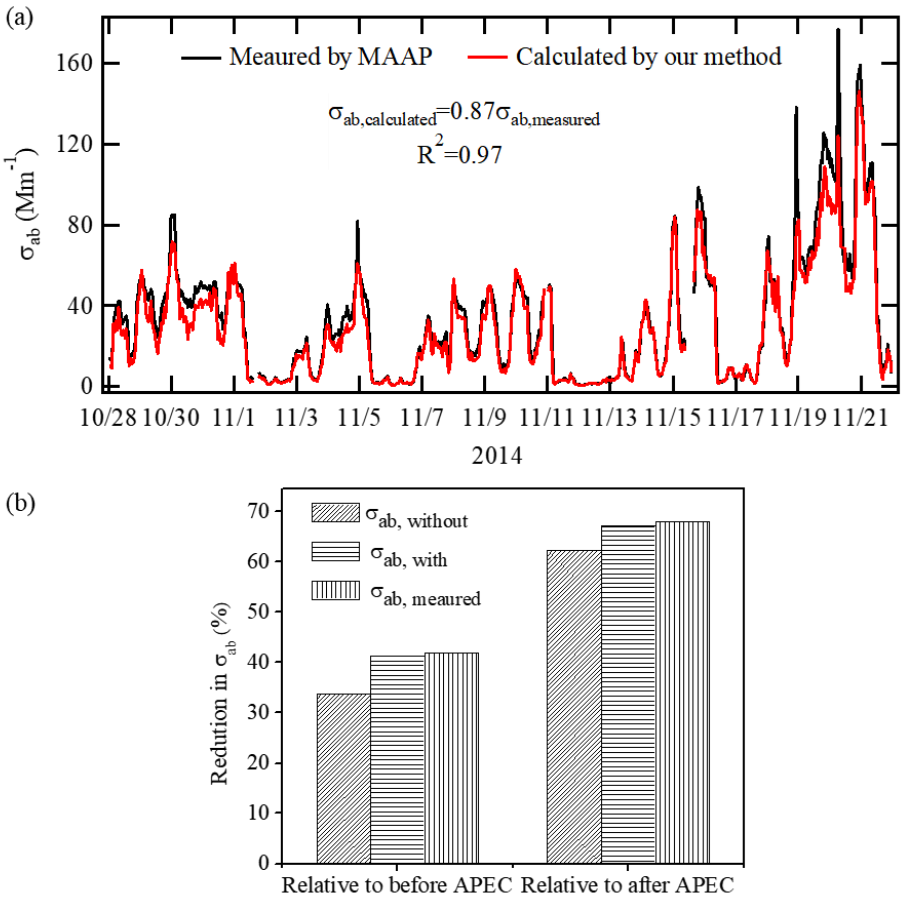

Figure R1 (new Fig. 7 in the revised manuscript). (a) The light absorption coefficient (σ_{ab}) at 670 nm. (b) Reduction in the daytime σ_{ab} of BC-containing particles observed in the pollution episode during APEC relative to that before and after APEC. The correlation between the calculated σ_{ab} ($\sigma_{ab,\ calculated}$) 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.

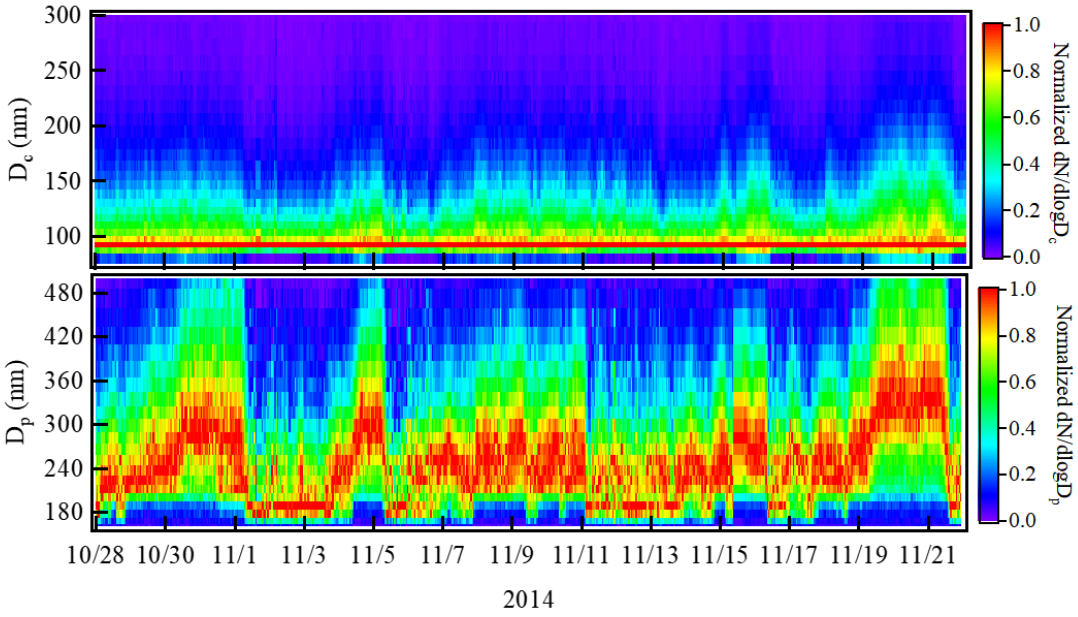

Figure R2 (Fig. 1 in the revised manuscript). Time series of the number size distribution of rBC cores (D_c) and whole BC-containing particles (D_p).

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.

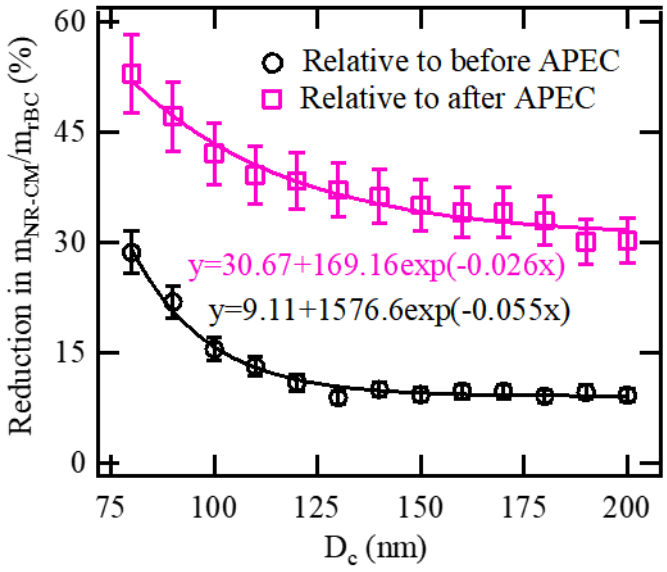

Figure R3 (new Fig. 3b in the revised manuscript). The reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles during APEC relative to that before and after APEC. The error bar shown in (b) represents the uncertainties (~10%) of $m_{NR\text{-}CM}/m_{rBC}$ ratio from Mie calculation.

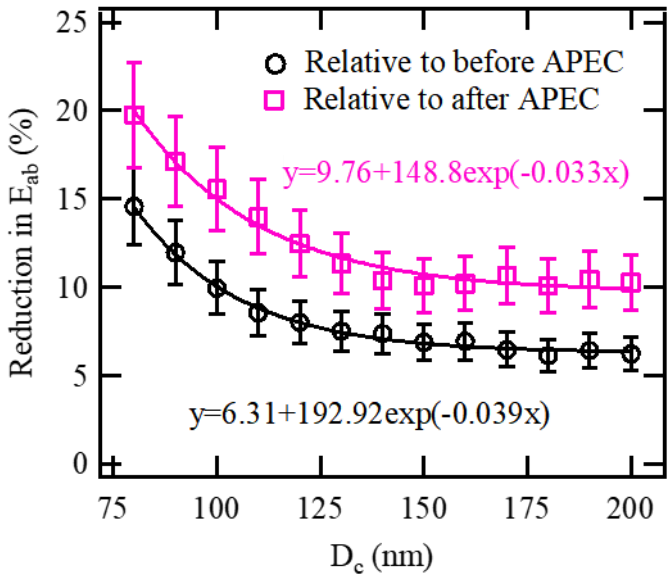

Figure R4 (new Fig. 6b in the revised manuscript). The reduction in E_{ab} of BC-containing particles during APEC relative to that before and after APEC. The error bar shown in (b) represents the uncertainties (~15%) of E_{ab} from Mie calculation.

(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."

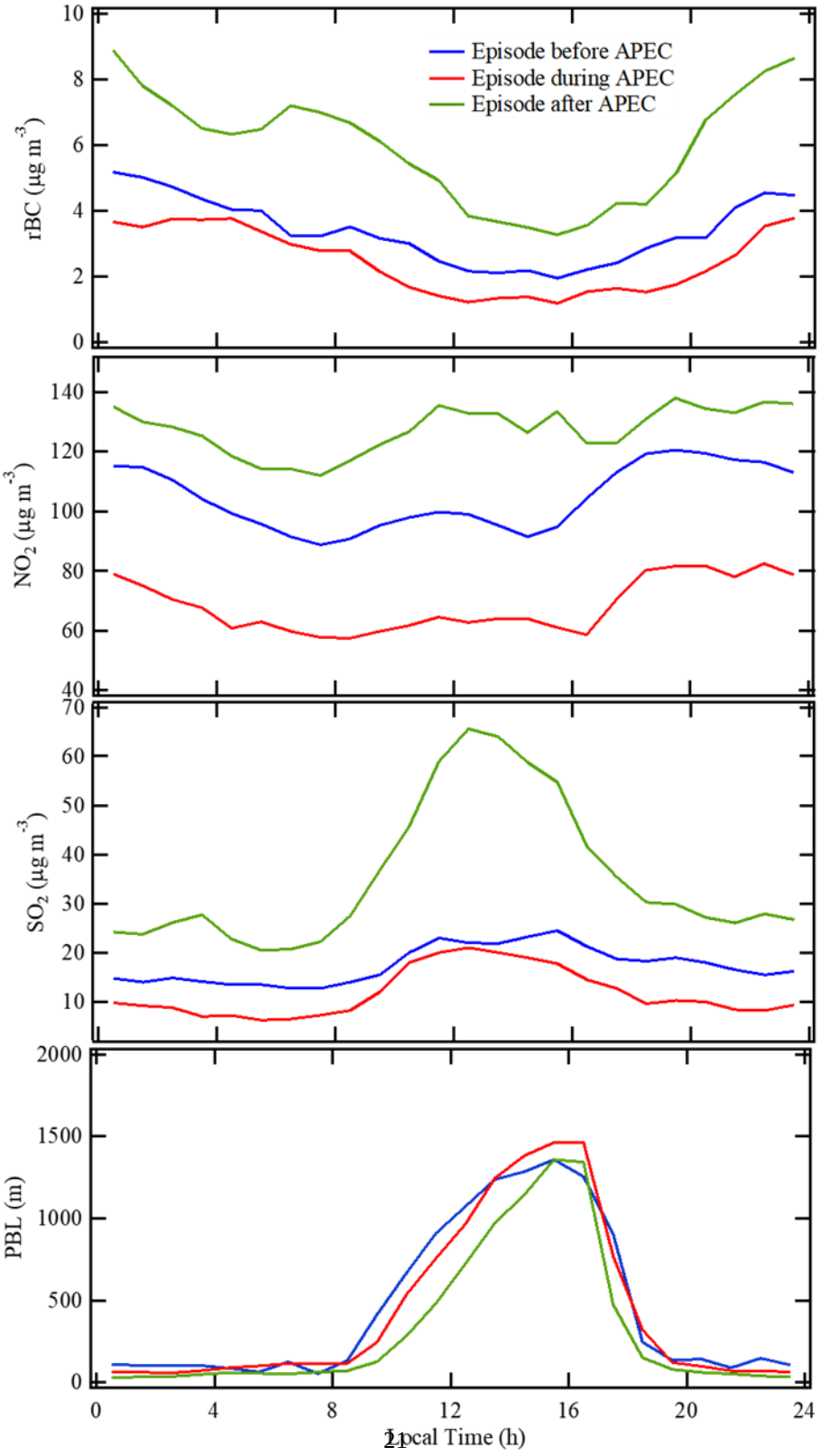
Episode before APEC
Episode during APEC
Episode after APEC
rBC (μg m-3)
NO2 (μg m-3)
SO2 (μg m-3)
PBL (m)
Local Time (h)

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.

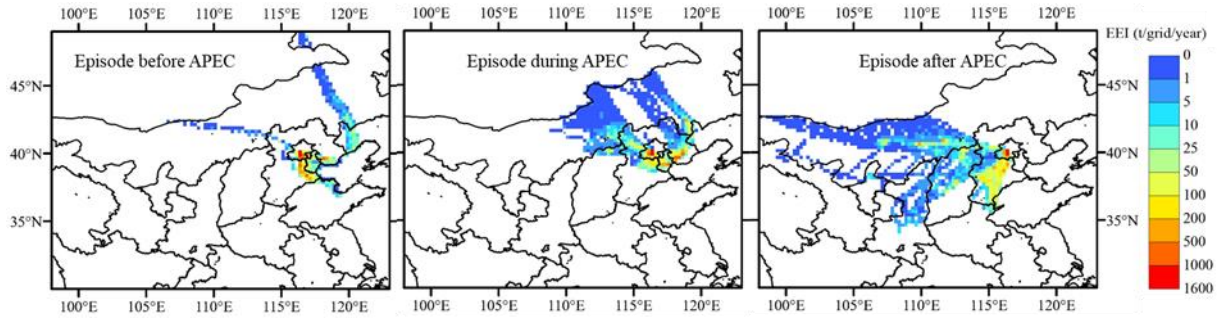

Figure R6 (Fig. S3 in the revised manuscript). Spatial distribution (0.25°×0.25°) of the effective emission intensity (EEI, defined by Lu et al. (2012)) for BC transported to the observation site (40º00'17" N, 116º19'34" E) for the pollution episodes before, during and after APEC. The EEI calculation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

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Reduction in black carbon light absorption due to multi-pollutant emission control during APEC China 2014

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Abstract. Reducing black carbon (BC) emissions has been recognized as an efficient way to simultaneously improve air quality and mitigate climate change. However, the benefits of BC emission controls are not well quantified partly due to a lack of understanding of the changes in BC light absorption as a result of emission reductions. In this work, we discussed the effects of multi-pollutant emission reductions on the BC light absorption based on a field campaign study conducted before, during and after the 2014 APEC (Asia-Pacific Economic Cooperation) meeting in Beijing, China. When emission restrictions were in place during APEC, we found that the reduction in the light absorption of BC-containing particles was driven by both the decrease in BC mass concentration and the weakened light-absorption capability of BC. Compared with that before and after APEC, the daytime light absorption of BC-containing particles during APEC reduced by ~56%, of which ~48% was contributed by the decrease in BC mass concentration and the remain ~8% was contributed by a weakening of light-absorption capability for BC. Based on single particle soot photometer (SP2) measurement and Mie calculation, we estimated that the light-absorption capability of BC-containing particles with ~80-200 nm refractory BC (rBC) cores at daytime during APEC was reduced by ~6-15% and ~10-20% compared with that before and after APEC, respectively. The decrease in BC light-absorption capability could be attributed to less coating materials on BC surfaces as a result of a decreased chemical production of secondary aerosols. Compared with that before and after APEC, the mass ratio between the coating materials and rBC core (~80-200 nm) during APEC decreased by ~10-30% and ~31-53%, respectively, due to reductions in coating precursor emissions, e.g., SO_2 and NO_2. The results revealed the benefits of emission control on BC light absorption by simultaneously reducing the mass concentration and light-absorption capability of BC, implying that synergetic reduction in multiple-pollutant emission could benefit both air quality and climate.

1 Introduction

Black carbon (BC) has drawn considerable attention due to its key role in climate and the atmospheric environment (Bond

and Sun, 2005; Jacobson et al., 2002, 2010). Because BC is the most efficient light-absorbing component in ambient aerosols (Bond and Bergstrom, 2006; Ramanathan and Carmichael, 2008), reduction measures targeting BC emissions have been recognized as a viable way to mitigate global warming (Shindell et al. 2012; Jacobson et al., 2010) and improve air quality in polluted regions (Ding et al., 2016; Wang et al., 2018). The benefits of BC emission reduction are mainly driven by more solar radiation reaching the surface due to the reduction in BC light absorption in the atmosphere.

The light absorption of ambient BC-containing particles can be reduced by decreasing the BC mass concentration, weakening the BC light-absorption capability or implementing both strategies. As primary aerosols, the mass concentration of BC particles generally decreases with emission reduction. When emission control measures were implemented, the mass concentration of the BC present in the atmosphere was proven to decrease (Han et al., 2015; Huang et al., 2010; Xu et al., 2015; Zhang et al., 2016a). In terms of the influence of emission reduction on the characteristics of BC aerosols, previous studies usually highlighted the decrease in BC mass concentration (Han et al., 2015; Wang et al., 2018; Zhang et al., 2016a). However, few studies considered the change in light-absorption capability of BC-containing particles due to emission reduction.

The light-absorption capability of ambient BC-containing particles is closely associated with their aging degree (Jacobson et al., 2001; Liu et al. 2017; Moffet et al., 2009; Peng et al., 2016; Zhang et al. 2016c, 2018), i.e., the degree to which BC is internally mixed with other species (e.g., sulfate and nitrate) (Oshima et al., 2009). When fresh BC is emitted from incomplete combustion (e.g., traffic emission) other than biomass burning (Wang et al., 2018; Pan et al., 2017), they are most likely~~most of BC particles are~~ externally mixed with other aerosol components (e.g., primary organic aerosol). These fresh BC particles exist as almost bare particles with few other species condensed on their surfaces and are named externally mixed BC particles (Jacobson et al., 2001; Chung et al. 2005). During atmospheric transport, fresh BC particles undergo aging, in which internally mixed BC particles form when other aerosol components coat the bare BC surface (Cheng et al., 2006; Bond and Bergstrom, 2006; Peng et al., 2016; Zhang et al., 2018). The internally mixed BC particles generally have a shell-and-core morphology, with the coating materials and BC as the shell and core, respectively. This shell-and-core morphology endows BC particles with a higher light-absorption capability because the coating materials act as a lens to focus more photons on BC (lensing effect, Lack and Cappa 2010). Compared with externally mixed BC particles (i.e., bare BC), the light absorption of internally mixed BC particles (i,.e, coated BC) can be enhanced by a factor of 2-3 (Fuller et al., 1999; Jacobson et al., 2001; Schnaiter et al., 2005; Zhang et al., 2016c).

Emission reduction may affect the lensing effect by changing the amount of coating materials for the BC-containing particles and consequently altering the light-absorption capability of BC. Emission control measures can reduce the concentrations of not only BC but also co-emitted gaseous pollutants (e.g., volatile organic compounds (VOCs), SO_2 and NO_x) present in the atmosphere (Tang et al., 2015; Huang et al., 2015). The reduction in these secondary aerosol precursors can lower the production of secondary components (e.g. secondary organic matter, sulfate and nitrate) in aerosol particles (Cheng et al., 2008; Huang et al., 2010; Han et al., 2015). This relationship implies that the interaction between BC and secondary aerosol components via condensation and coagulation may be impacted by the primary emission reductions of

both BC and co-emitted pollutants (e.g., VOCs, SO_2 and NO_x), namely, emission control measures may influence BC aging in the atmosphere. As mentioned above, the aging degree of BC-containing particles exerts a substantial effect on their light-absorption capability. Less aged BC is expected as emission control measures are implemented to decrease BC light-absorption capability. However, it is still unclear whether emission control measures can lower the aging degree of BC-containing particles and thus weaken their light-absorption capability.

In this work, we used the 2014 Asia-Pacific Economic Cooperation (APEC) meeting in Beijing, China, as a case study to investigate the effects of emission control measures on the light absorption of ambient BC-containing particles. This paper reported the in situ measurements before, during and after APEC and investigated how the concentrations of BC and coating precursors, the BC aging degree and the BC light-absorption capability were affected by emission reductions. Based on these results, we quantified the impact of emission reduction during APEC on the light absorption of BC-containing particles and further discuss the additional effect of emission control measures on BC light absorption due to changes in the coating materials of ambient BC particles.

2 Methods and data

2.1 Measurement location and period

The in situ measurement was carried out on the campus of Tsinghua University (40º00'17" N, 116º19'34" E, Fig. S1). The observation site is located in downtown Beijing, approximately 1 km from North 4th Ring Road, which has a high traffic density. The air quality at this site is considered typical of the Beijing urban environment. More details regarding the Tsinghua site can be found in Zheng et al., (2015) and Zhang et al. (2018).

The measurement period lasted from October 28 to November 21, 2014. A series of aggressive measures were implemented from November 3 to 12, 2014 in Beijing and the surrounding areas (i.e., Tianjin, Hebei, Shanxi, Shandong, Henan and Inner Mongolia, shown in Fig. S1) to achieve good air quality during the APEC meeting: mandatory restrictions on traffic flow in Beijing, limited or arrested production from high-emitting factories, suspended construction activities and bans on various outdoor burning practices (Gao et al., 2017; Huang et al., 2015; Tang et al., 2015; Zhang et al., 2016a; Zhang et al., 2016b). In this study, we classified the observation period into five subperiods: before APEC (October 28-November 2, 2014), which served as a reference; during APEC (November 6-12, 2014), which was characterized by the enforcement of emission control measures; after APEC (November 17-21, 2014), which served as another reference; and two transition periods (November 3-5 and 13-16, 2014), which were ~~defined considering the lifetime of BC during atmospheric transport and~~ 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).~~.~~

2.2 Instrumentation

A single-particle soot photometer (SP2) instrument (Droplet Measurement Technologies, Boulder, CO, USA) uses a 1064 nm Nd:YAG laser to measure the mass of a refractory BC (rBC) core (m_{rBC}) and the scattering cross section (C_s) of an individual BC-containing particle. As a light-absorbing component, a rBC core is gradually heated by the continuous laser beam and vaporizes at ~4000 K, where detectable incandescent light is emitted (Schwarz et al., 2006; Moteki and Kondo, 2010). The incandescence signal recorded by SP2 was used to determine the m_{rBC} of an individual BC-containing particle. The mass concentration of rBC was calculated based on the m_{rBC} and sampling flow rate (0.12 lpm (liter per minute)). On the other hand, we used the scattering signal from the SP2 measurement to retrieve the C_s of an individual BC-containing particle (including coating materials and rBC core) based on the leading-edge-only (LEO) method developed by Gao et al. (2007). The validity of the LEO method for ambient BC-containing particles observed in China has been evaluated by Zhang et al. (2016c). More details on the SP2 technique have been reported elsewhere (Gysel et al., 2011; Pan et al., 2017; Sedlacek et al., 2012; Zhang et al., 2016c).

The observational data of hourly $PM_{2.5}$, SO_2, NO_2 and O_3 concentrations at the Wanliu station in urban Beijing were downloaded from the Atmospheric Environment Monitoring Network (http://www.zhb.gov.cn/). The Wanliu station is approximately 5 km away from the Tsinghua site**.**

2.3 Data analysis

2.3.1 Aging degree of BC-containing particles

The aging degree of ambient BC-containing particles was retrieved by the SP2 measurements (i.e., the m_{rBC} and the C_s of BC-containing particles) and Mie calculation. To quantify the aging degree of BC-containing particles, we assumed that a BC-containing particle was a sphere with a rBC core and a non-refractory coating material (NR-CM) shell (Moteki and Kondo, 2007; Subramanian et al., 2010; Zhang et al., 2016c). The actual shape of BC-containing particles in the atmosphere was complex (He et al., 2015; Scarnato et al. 2013; Wang et al., 2017). 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.

In this study, the diameter of the rBC core (D_c) and the whole particle diameter including the core and shell (D_p) were calculated to retrieve the aging degree of BC-containing particles. D_c was calculated from m_{rBC} and the density of the rBC core (ρ_c, here, a prescribed value of 1.8 g cm^{-3}) (Cappa et al., 2012; Pan et al., 2017; Laborde et al., 2013). D_p was determined via the Mie calculation and was related to the D_c, the C_s of the BC-containing particle, and the refractive indices of NR-CM ($RI_{NR\text{-}CM}$, 2.26-1.26i) and rBC core (RI_c, 1.5-0i). ~~The values of $RI_{NR\text{-}CM}$ and RI_e used in this study were 1.5-0i and~~

~~2.26-1.26i, respectively (Cappa et al., 2012; Taylor et al., 2015).~~ The uncertainty of size information of BC-containing particles from Mie calculation was estimated to be ~10% in our previous work (Zhang et al., 2018). More details regarding the calculation of D_p and D_c for ambient BC-containing particles observed in Tsinghua site can be found in Zhang et al. (2018).

In this study, the aging degree of a BC-containing particle was characterized by the mass ratio between NR-CM and rBC ($m_{NR\text{-}CM}/m_{rBC}$) and was calculated by Eq. (1):

$$\frac{m_{NR-CM}}{m_{rBC}} = \frac{\frac{1}{6}\times\pi\times(D_P^3-D_c^3)\times\rho_{NR-CM}}{m_{rBC}} \quad (1)$$

where $m_{NR\text{-}CM}$ is the mass of the non-refractory coating materials; $\rho_{NR\text{-}CM}$ is the density of the non-refractory coating materials, with a prescribed value of 1.4 g cm^{-3} in this study based on the composition of submicron aerosols during APEC reported by Zhang et al. (2016a) and the densities of the various components (i.e., sulfate, nitrate, ammonium and organic aerosol) (Cappa et al., 2012).

2.3.2 Light absorption of BC-containing particles

In this study, the light-absorption capability of ambient BC-containing particles was characterized by the light absorption enhancement (E_{ab}) of BC from the lensing effect caused by the coating materials. The E_{ab} of BC-containing particles was retrieved using a shell-and-core model based on Mie theory (Laborde et al., 2013; Metcalf et al., 2013; Schwarz et al., 2008), calculated by dividing the light absorption cross-section of the whole BC-containing particle ($C_{ab,p}$) by that of the bare rBC core ($C_{ab,c}$) at a certain wavelength (550 nm in this study), as expressed in Eq. (2):

$$E_{ab} = \frac{C_{ab,p}\ (D_c, D_p, RI_{NR-MC}, RI_c)}{C_{ab,c}(D_c, RI_c)} \quad (2)$$

where $C_{ab,c}$ and $C_{ab,p}$ were determined from the Mie calculation (uncertainty of ~15% estimated in our previous study (Zhang et al., 2018)). $C_{ab,c}$ is related to D_c and RI_c. For $C_{ab,p}$, we needed additional information on the whole particle, i.e., D_p and $RI_{NR\text{-}CM}$.

The light absorption coefficient (σ_{ab}) of BC-containing particles at a wavelength (550 nm and 670 nm used in this study) was determined by the light-absorption capability of BC and the rBC mass concentration (C_{rBC}), as shown in Eq. (3):

$$\sigma_{ab} = C_{rBC} \times MAC_p = C_{rBC} \times E_{ab} \times MAC_c \quad (3)$$

Where MAC_p and MAC_c ~~is~~ are the mass absorption cross-section (MAC) of BC-containing particles and rBC cores, respectively, which was ~~prescribed a value of 7.5 m^2 g^{-1} at 550 nm (Bond and Bergstrom, 2006)~~calculated based on Mie theory and SP2 measurements. In this study, the σ_{ab} at 670 nm was also obtained by a multi-Angle absorption photometer (MAAP) measurement. The MAAP data were corrected using the algorithm reported by Hyvärinen et al. (2013).

3 Results

3.1 Reduction in the concentrations of BC and coating precursors

Figure 1a shows the time series of the $PM_{2.5}$ and rBC mass concentrations during the campaign period. Three pollution episodes 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),~~In this work,~~ we focused on comparing the BC characteristics among the three 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), in which 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 (PBL), as well as high relative humidity (Sun et al. 2014; Zheng et al. 2015).

~~.~~The $PM_{2.5}$ concentration during the pollution episodes before and after APEC were ~127 μg m^{-3} and ~213 μg m^{-3}, respectively, which were larger than that (~66 μg m^{-3}) during APEC. The decrease in $PM_{2.5}$ loadings revealed that the air quality was improved during APEC. Similarly, the rBC mass concentration during APEC was also smaller than ~~that~~ those before and after APEC. However, the decreases in the rBC concentration during APEC by ~27% and ~58%, respectively, compared with that before and after APEC were smaller than the corresponding decreases in the $PM_{2.5}$ concentrations (~48% and 69%, respectively), possibly indicating that more secondary aerosols (e.g., sulfate and nitrate) than primary aerosols (e.g., rBC) were reduced during APEC, which could aid the decrease in coating materials on BC surfaces.

Fig. 2 compares the mass concentrations of both rBC and the coating precursors (i.e., NO_2 and SO_2) in the pollution episodes before, during and after APEC. Compared with that before and after APEC, the mass concentration of NO_2 during APEC was decreased by ~34% and ~45%, respectively, while the SO_2 concentration was reduced by ~35% and ~67%, respectively. These results revealed that the emission control measures implemented during APEC were a viable way to reduce not only the rBC mass concentrations but also the concentrations of secondary aerosol precursors present in the atmosphere. The emission control-caused reduction in secondary particle precursors (i.e., NO_2 and SO_2) during APEC could have reduced the secondary aerosol formation in the atmosphere. Previous studies 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)~~, indicating that the production of BC coating materials might have been affected by emission controls during APEC~~. 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.

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.

Previous studies have pointed out ~~Given~~ 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. We separated the data sets for the pollution episodes before, during and after APEC into daytime (07:00-19:00) and nighttime (19:00 to 07:00 of the following day) sets. Fig. 2 shows that while the emission controls were in place during APEC, a greater reduction in the rBC and NO_2 concentrations occurred during the day than at night. Compared with those before and after APEC, the daytime reductions in the NO_2 concentration during APEC were reduced by as much as ~40% and ~51%, respectively. By contrast, the daytime reduction (~25%) in the SO_2 concentration during APEC compared with that before APEC was smaller than that at night, which might be attributable to the high contribution of regional emissions (e.g., power generation and industrial activities in Hebei Province) to the daytime SO_2 concentration in Beijing (Guo et al., 2014; Tang et al., 2015). Meanwhile, a similar reduction (~67%) in the daytime and nighttime SO_2 concentrations during APEC compared with that after APEC was observed. In summary, the significant reductions in the daytime levels of rBC and coating precursors during APEC further indicated that the BC aging in the atmosphere might have been affected by the emission control measures.

3.2 Reductions in the aging degree of BC

Figs. 1b and 1c show time series of the number size distribution of rBC cores (D_c) and whole BC-containing particles (D_p), respectively. The rBC cores observed before, during and after APEC exhibited similar number size distributions, with a mode at ~95 nm (Fig. 1b). The similar modes of the rBC cores could have resulted from similar emission sources ~~and similar atmospheric processes (coagulation and wet removal)~~ for BC-containing particles observed before, during and after APEC. However, the whole BC-containing particles (including coating materials and rBC core) showed different number size distributions in the pollution episodes before, during and after APEC (Fig. 1c), indicating different amounts of coating materials on the BC surface during the three pollution episodes. 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 condensation and coagulation of other species (i.e., primary aerosol and secondary components)~~of secondary species~~ on the BC surface. However, the continuous size growth of the whole BC-containing particles was not observed in the pollution episode during APEC, in which the number particle size distribution with a mode no more than 280 nm (Fig. 1c), significantly smaller than ~~that~~ those before (~320 nm) and after APEC (~400 nm). These

results indicated that secondary formation during APEC was insufficient to maintain continuous BC aging.

Fig. 3 compares the mass ratio between the coating materials and rBC cores ($m_{NR\text{-}CM}/m_{rBC}$) for BC-containing particles with size-resolved rBC cores in the pollution episodes before, during and after APEC. The $m_{NR\text{-}CM}/m_{rBC}$ ratios of BC-containing particles before, during and after APEC showed similar correlations with the rBC core size, namely, the $m_{NR\text{-}CM}/m_{rBC}$ ratio decreased with increasing rBC core size (Fig. 3a). The size-dependent $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles indicated that ~~condensational~~ particle growth was more effective for smaller particles, which followed the diffusion-controlled growth law (Seinfeld and Pandis 2006). At a certain size of rBC cores, Fig. 3a shows that $m_{NR\text{-}CM}/m_{rBC}$ ratio of ambient BC-containing particles during APEC was significantly smaller than that before and after APEC, revealing that the emission restrictions during APEC weakened the condensation of other species on the BC surface. For ambient BC-containing particles with ~80-200 nm rBC cores, the $m_{NR\text{-}CM}/m_{rBC}$ ratios observed in the pollution episodes before, during and after APEC were 4-22, 3-15, and 5-33, respectively.

Fig. 3b shows the reductions in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles for the pollution episodes during APEC compared with ~~that~~ those before and after APEC, which were also dependent on rBC core size. Smaller rBC cores exhibited greater reductions in the $m_{NR\text{-}CM}/m_{rBC}$ ratio as a result of emission control measurements during APEC~~,~~. This ~~indicating~~ indicated that in terms of BC aging, it was more sensitive to emission levels for smaller rBC cores. This could be explained by the diffusion-controlled growth law, i.e., the condensational growth of smaller BC particles was more effective (Metcalf et al., 2013; Seinfeld and Pandis, 2006), and thus, the effect of emission reduction on BC aging was more significant for smaller rBC particles. Compared with that before and after APEC, the $m_{NR\text{-}CM}/m_{rBC}$ ratio of ambient BC-containing particles with ~80-200 nm rBC cores during APEC was reduced by ~10-30% and ~31-53%, respectively. The relationship between the reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles (R_{aging}) during APEC and their rBC core size (D_c) followed an exponential function (Fig. 3b), i.e., R_{aging} = 9.1+1576.6exp(-0.055D_c) (relative to that before APEC) and R_{aging} = 30.7+169.2exp(-0.025D_c) (relative to that after APEC).

The reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles for the pollution episode during APEC relative to that before and after APEC showed pronounced diurnal cycles (Fig. 4). Compared with that before APEC, the reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles with 80-200 nm rBC cores during APEC showed maxima in the afternoon (~14:00-17:00 LT) (Fig. 4a), consistent with the peak time of the diurnal cycle of O_3 concentrations before and during APEC (Fig. 4c). This consistence indicated that the reduction in coating materials on the BC surface during APEC compared to that before APEC was most likely dominated by a lower photochemical production of secondary species. Fig. 5a1 shows that the reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles during APEC relative to that before APEC increases with the O_3 concentration during the day (7:00-19:00 LT), revealing that the effect of emission controls on BC aging is associated with photochemistry. Moreover, Fig. 4a shows the diurnal cycle of the reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles during APEC compared to that before APEC with minima during rush hour (~6:00-8:00 LT), which can be due to a larger contribution of primary emissions of fresh BC (namely, bare BC and thin coated BC particles) during rush hour than at other times for both episodes before and during APEC.

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). Fig. 4c shows that the daytime O_3 concentrations after APEC are significantly smaller than those during APEC, indicating a weakened contribution from photochemistry after APEC. The increased amount of coating materials of BC observed after APEC compared to that during APEC was mostly likely attributed to enhanced other reactions (e.g. heterogeneous chemistry) during haze episodes (Xie et al., 2015; Yang et al., 2015; Zheng et al., 2015; Mu et al., 2018). Fig. 5a2 shows that the variation in the reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles during APEC compared to that after APEC is poorly correlated with the O_3 concentration. The diurnal trend of the reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles during APEC relative to that after APEC was likely driven by the simultaneous effects of enhanced photochemistry and weakened ~~heterogeneous~~ other chemistry (e.g. heterogeneous reaction) contributions during APEC.

As discussed above, the reduction in the aging degree of ambient BC-containing particles during APEC could have been caused by a decreased chemical production (namely, weakened contributions from photochemical or ~~heterogeneous~~ other reactions) of coating materials on the BC surface. Fig. 5b shows that the reduction in the $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles during APEC relative to that before and after APEC is associated with a decrease of the concentrations of SO_2 and NO_2 due to emission reduction. A greater decrease in the concentrations of SO_2 and NO_2 corresponded to a greater reduction in the $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles during APEC. The reduction in precursor emissions of secondary species (e.g., SO_2 and NO_2) could decrease the chemical production, and therefore, lower amounts of coating materials on the BC surfaces were observed during APEC.

3.3 Reduction in the light absorption of BC-containing particles

The reduction in the BC aging degree during APEC ~~should have~~could weaken~~ed~~ the light-absorption capability of BC-containing particles owing to a decrease in the lensing effect caused by less coating materials on the BC surfaces (Fuller et al. 1999; Lack and Cappa 2010). Fig. 6 compares the E_{ab} of BC-containing particles during the day for the pollution episodes observed before, during and after APEC. The daytime E_{ab} of BC-containing particles with 80-200 nm rBC cores varied from ~1.5 to ~2.5 during APEC, values that were remarkably lower than that before and after APEC (i.e., E_{ab} of 1.7-3.0 and 1.8-3.2, respectively, Fig. 6a); these results reflected a weakened light-absorption capability of BC during APEC. The reduction in the daytime E_{ab} of BC-containing particles (R_{Eab}) during APEC compared with ~~that~~ those before and after APEC decreased with the rBC core size (D_c), and the relationship followed an exponential function (R_{Eab} = 6.3+192.9exp(-0.039D_c) (relative to that before APEC) and R_{Eab} = 9.8+148.8exp(-0.033D_c) (relative to that after APEC)) shown in Fig. 6b. Compared with that of before and after APEC, the E_{ab} of BC-containing particles with ~80-200 nm rBC cores during the day decreased by ~6-15% and ~10-20%, respectively. Our results provided evidence that emission controls could weaken the light-absorption capability of ambient BC-containing particles. This weakening would have enhanced the effects of emission control measures during APEC on BC light absorption.

~~The reduction in both the rBC mass concentration and the light absorption capability of ambient BC-containing particles during APEC revealed a decrease in the light absorption of BC aerosols caused by emission control measures. As shown in Fig. 7, we calculated the theoretical reduction in the light absorption coefficient of BC during APEC based on the simultaneous reduction in the mass concentration and light absorption capability of BC ($\sigma_{ab,with}$), and the reduction in absorption coefficient calculated without considering the weakened light absorption capability of BC-containing particles due to emission reduction ($\sigma_{ab,without}$) was also obtained. The comparison between the reductions in $\sigma_{ab,with}$ and $\sigma_{ab,without}$ could 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.~~Figure 7a shows the measured and theoretical light absorption coefficient 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 episodes 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.

Considering the reductions in both the mass concentration and light-absorption capability of BC due to the emission control measures, the daytime light absorption of BC-containing particles (i.e., $\sigma_{ab,with}$) decreased by ~41% and ~68% during APEC compared to that before and after APEC, respectively. However, the $\sigma_{ab,without}$ of BC during APEC decreased by ~34% and ~62% relative to that before and after APEC, respectively (Fig. 7b). The difference between the reductions in $\sigma_{ab,with}$ and $\sigma_{ab,without}$ indicated that the reduction in the rBC concentration contributed ~83% and ~91% of the reduction in BC light absorption during APEC compared to that before and after APEC, respectively, while the weakening of the BC light-absorption capability contributed ~17% and ~9%, respectively. On average, the light absorption of BC-containing particles at daytime during APEC decreased by ~56% compared with before and after APEC, of which ~48% was contributed by the reduction in the mass concentration of rBC and the remain ~8% was controlled by the weakening of BC light-absorption capability. These results imply that reductions in the emissions of multiple pollutants (i.e., BC and precursors of secondary species) in China could benefit air quality and climate due to significantly lowering the light absorption of BC, which was driven by the reductions in both rBC mass concentration and light-absorption capability of BC-containing particles.

4 Discussion

Based on a comparison of the observations before, during and after APEC, we found that the emission control measures successfully reduced both the rBC mass concentration and the light-absorption capability (i.e., E_{ab}) of BC-containing particles, resulting in a significant decrease in the light absorption of BC. The mechanism underlying the effect of the emission reductions during APEC on BC light absorption is summarized in Fig. 8. Emission control measures reduce the amount of both BC and co-emitted secondary aerosol precursors present in the atmosphere. The presence of lower amounts of secondary particle precursors in the atmosphere weakens the chemical formation of secondary aerosol components, suppressing the condensation of secondary species on BC surfaces. Less coating material on BC can weaken the lensing effect, which leads to a weakening of the light-absorption capability for BC-containing particles. Simultaneous reductions in the mass concentration and light-absorption capability of BC can result in a much lower light absorption of BC during APEC compared to ~~that~~ thoset before and after APEC.

In China, a series of emission controls measures have been implemented in pollution regions (e.g., Jing-Jin-Ji region), aiming to increase the number of clean days and decrease the number of haze days. This comparison between periods with and without emission controls measures may illustrate the differences between clean and polluted periods. In terms of different pollution levels in China, our findings imply that a clean period is characterized by not only a lower BC mass concentration but also a weaker light-absorption capability of BC-containing particles compared to that in polluted periods. In our previous study (Zhang et al., 2018), we found that the light-absorption capability of ambient BC-containing particles observed in Beijing was enhanced by an increase in pollution levels, resulting in an amplification of BC light absorption under polluted conditions. The present work clearly demonstrates that emission control measures can reduce this amplification effect by decreasing the light-absorption capability of BC-containing particles. Moreover, this work can explain how emission control measures reduce the amplification effect, namely, by slowing the aging of BC ~~through~~ resulted from a reduction in co-emitted secondary aerosol precursors (e.g., SO_2, NO_x and VOCs).

The simultaneous reductions in the mass concentration and light-absorption capability of BC due to emission controls confirmed the suggestions of previous studies that BC emission reductions could achieve multiple benefits, i.e., simultaneously controlling air pollution and protecting the climate (Ding et al., 2016). Furthermore, our study implies that the air quality and climate co-benefits from multi-pollutant emission controls are enhanced by the weakened light-absorption capability of BC-containing particles. In terms of air quality improvement, the weakened light-absorption capability plays an important role in both the direct and indirect effects of BC. Weakened light-absorption capability can directly lower the light-absorbing efficiency of BC aerosols in the atmosphere, resulting in more solar light radiation reaching the surface~~, and~~; the weakened light-absorption capability of ambient BC-containing particles can indirectly mitigate air pollution by improving ~~the planetary boundary layer (~~PBL~~)~~ suppression driven by the dome effect of BC (Ding et al., 2016; Wang et al., 2017). On the other hand, an enhanced reduction in climate warming can be attributed to a smaller direct radiative forcing from BC aerosols due to a weaker light-absorption capability of atmospheric BC-containing particles. The importance of the

weakened light-absorption capability of BC highlighted in our study provides clues for the management of air quality and climate change. The emission controls of multiple pollutants including BC and co-emitted secondary aerosol precursors may be an efficient way to simultaneously mitigate air pollution and climate warming.

5 Concluding remarks

The effects of emission reductions on the light absorption of BC-containing particles are not only controlled by the reduction in the BC mass concentration but also dependent on the change in their light-absorption capability. The decrease in the BC mass concentration due to emission control measures is well known. However, the impact of emission reduction on the light-absorption capability of BC-containing particles remains unclear due to a lack of available observations. The 2014 APEC meeting in Beijing, China, provides an invaluable opportunity to measure the variations in the light-absorption capability of ambient BC-containing particles due to emission reductions. In this work, based on in situ measurements at an urban site in Beijing before, during and after APEC using a SP2, we explored whether and how emission control measures in China influence the light-absorption capability of ambient BC-containing particles. Note that this comparative study focused on the pollution episodes before, during and after APEC.

We found that the emission control measures successfully lowered the aging degree (i.e., $m_{NR\text{-}CM}/m_{rBC}$) of BC-containing particle. The $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles with ~80-200 nm rBC cores during APEC decreased by ~10-30% and ~31-53% compared to that before and after APEC, respectively. The reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles increased with decreasing rBC core size, following an exponential function. The size-dependent reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles indicated that emission reduction was more effective for slowing the aging of smaller rBC particles. The reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles during APEC relative to ~~that~~ thoset before and after APEC showed a pronounced diurnal cycle, with maxima at ~14:00-17:00 LT and ~10:00-12:00, respectively. The decreased ageing of BC-containing particles during APEC was mainly driven by a reduction in chemical production (i.e., oxidation products such as sulfate and nitrate) on the surface of BC due to less amounts of secondary aerosol precursors (e.g., the NO_2 concentration during APEC decreased by ~34% and ~45% compared with ~~that~~ those before and after APEC, respectively, and the corresponding SO_2 concentration decreased by ~35% and ~67% during APEC, respectively) present in the atmosphere during BC aging. The reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles during APEC relative to ~~that~~ those before and after APEC increased with the reduction in the concentrations of NO_2 and SO_2.

Due to the lower amount of coating materials on BC surfaces during APEC, the light-absorption capability (i.e., E_{ab}) of BC-containing particles with ~80-200 nm rBC cores during the day decreased by ~6-15% and ~10-20% compared to ~~that~~ those before and after APEC, respectively. The weakened light-absorption capability of BC-containing particles enhanced the reduction in BC light absorption due to the emission control measures. When considering the reduction in both the mass concentration and light-absorption capability of BC-containing particles during the day during APEC, the theoretical light absorption (i.e., σ_{ab}) decreased by ~41% and ~68% compared to ~~that~~ those before and after APEC, respectively. However, the

reduced light absorption of BC during the day caused by the decrease in the BC mass concentration during APEC compared to ~~that~~ those before and after APEC was estimated to be ~34% and ~62%, respectively. Therefore, ~10-20% of the reduction in the daytime light absorption of BC-containing particles during APEC relative to ~~that~~ those before and after APEC could be attributed to the weakened light-absorption capability. Our study revealed that reductions in the emissions of multiple pollutants (i.e., BC, NO_2 and SO_2) could reduce the light-absorption capability of BC. Weakened light-absorption capability of BC due to emission controls further confirmed the suggestions of previous studies that BC emission reductions can achieve multiple benefits, i.e., simultaneously controlling air pollution and protecting the climate (Ding et al., 2016; Peng et al., 2016; Zhang et al., 2018). Our study then implied that the air quality and climate co-benefits from multi-pollutant emission control could be enhanced by the weakened light-absorption capability of BC-containing particles.

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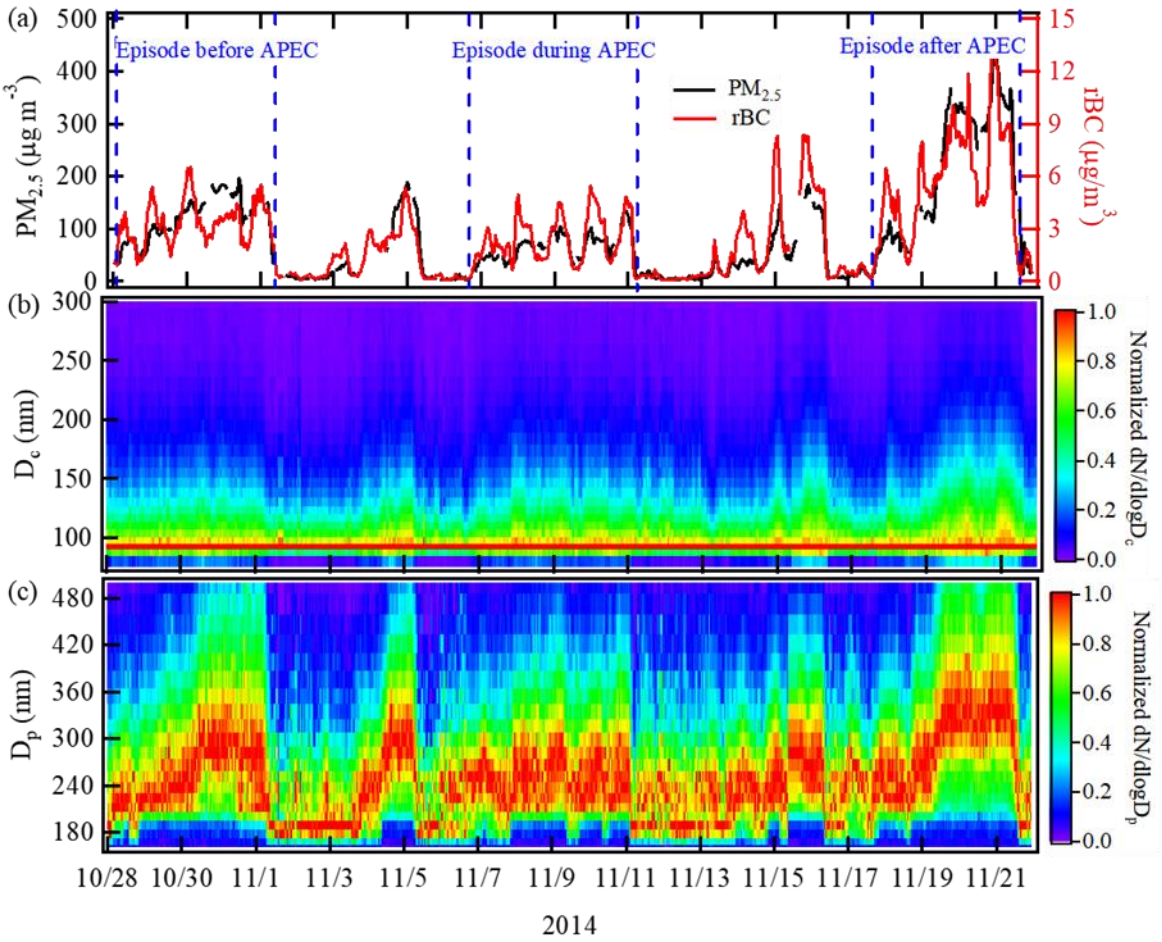


Figure 1. Time series of (a) the mass concentrations of $PM_{2.5}$ and rBC and the number size distribution of (b) rBC cores (D_c) and (c) whole BC-containing particles (D_p).

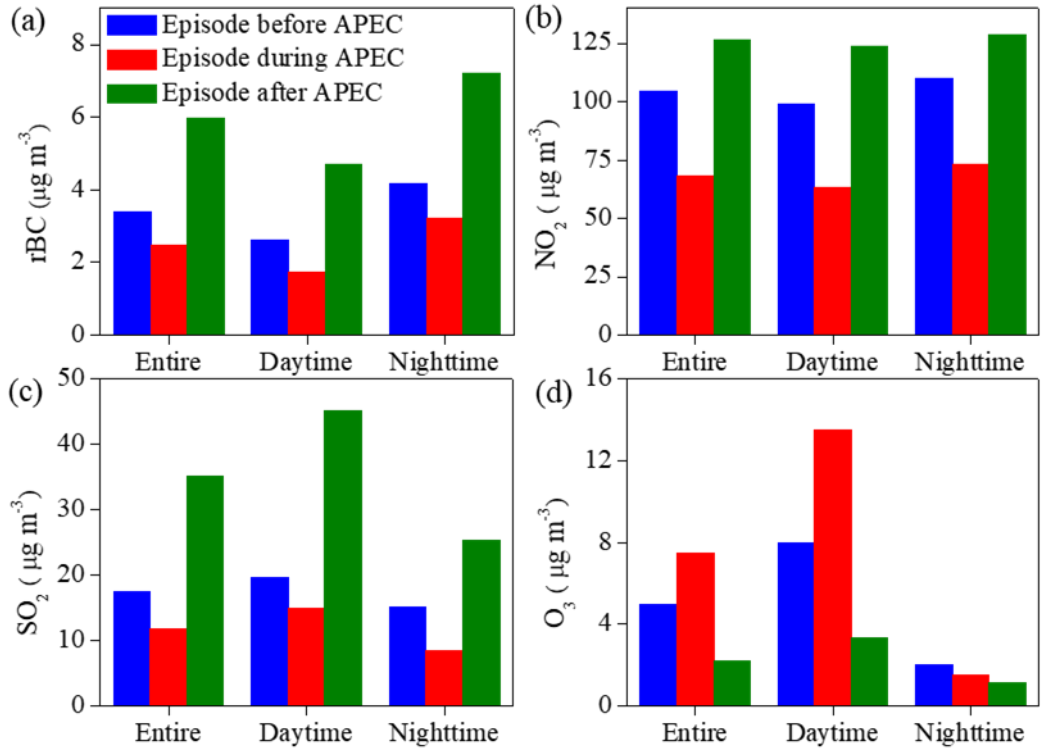
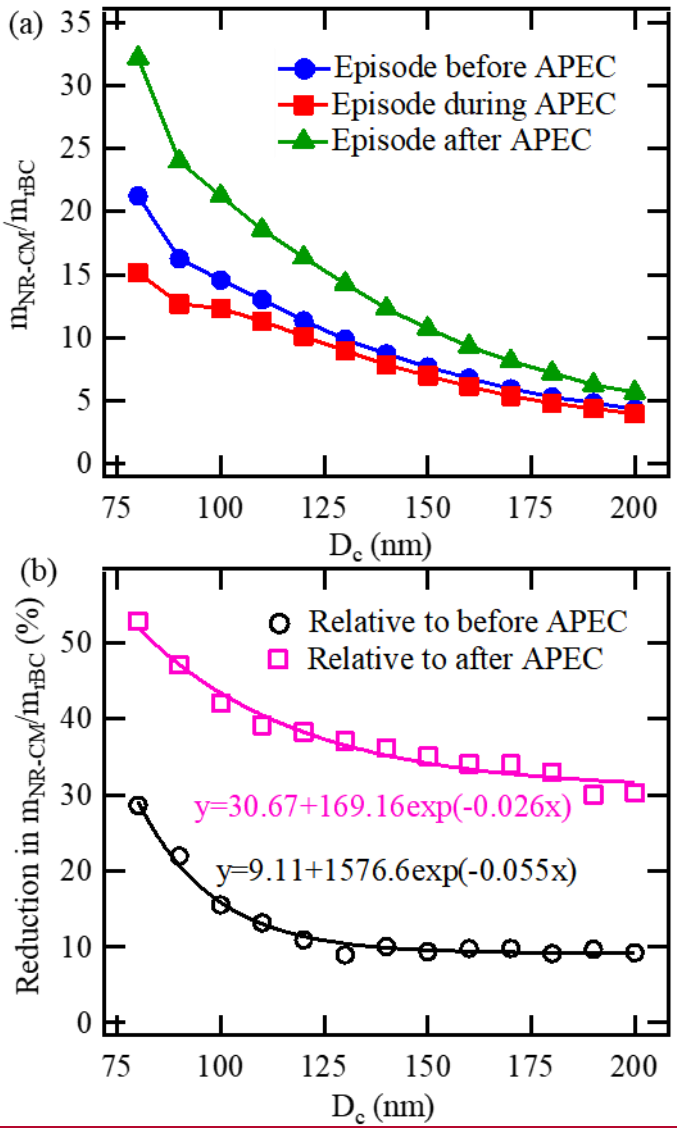


Figure 2. The mass concentrations of (a) rBC, (b) NO_2, (c) SO_2 and (d) O_3 for the pollution episodes before, during and after APEC. We separated the entire data sets into daytime (7:00 LT to 19:00 LT) and nighttime (19:00 LT to 7:00 LT of the following day) sets.


(a)
Episode before APEC
Episode during APEC
Episode after APEC
m_{NR-CM}/m_{rBC}
D_c (nm)
(b)
Relative to before APEC
Relative to after APEC
y=30.67+169.16exp(-0.026x)
y=9.11+1576.6exp(-0.055x)
Reduction in m_{NR-CM}/m_{rBC} (%)
D_c (nm)

1

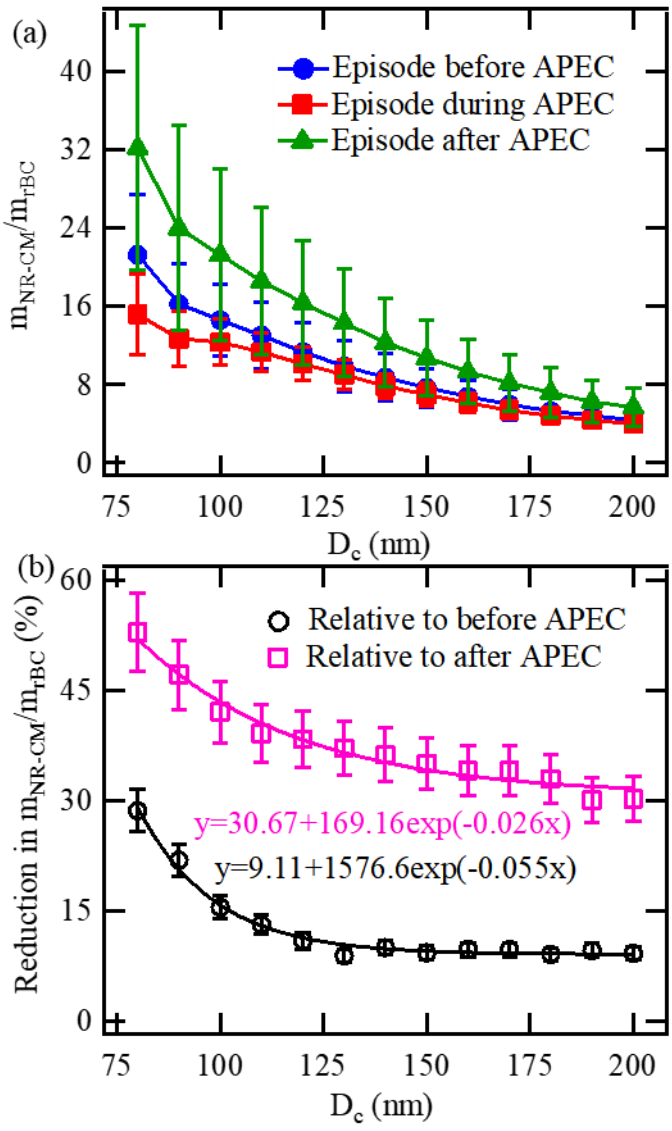


Figure 3. Comparison of the aging degree of BC-containing particles for the pollution episodes before, during and after APEC: (a) m_{NR-CM}/m_{rBC} ratio of BC-containing particles and (b) the reduction in m_{NR-CM}/m_{rBC} ratio of BC-containing particles during APEC relative to ~~that~~ those before and after APEC. The error bar shown in (b) represents the uncertainties (~15%) from Mie calculation.

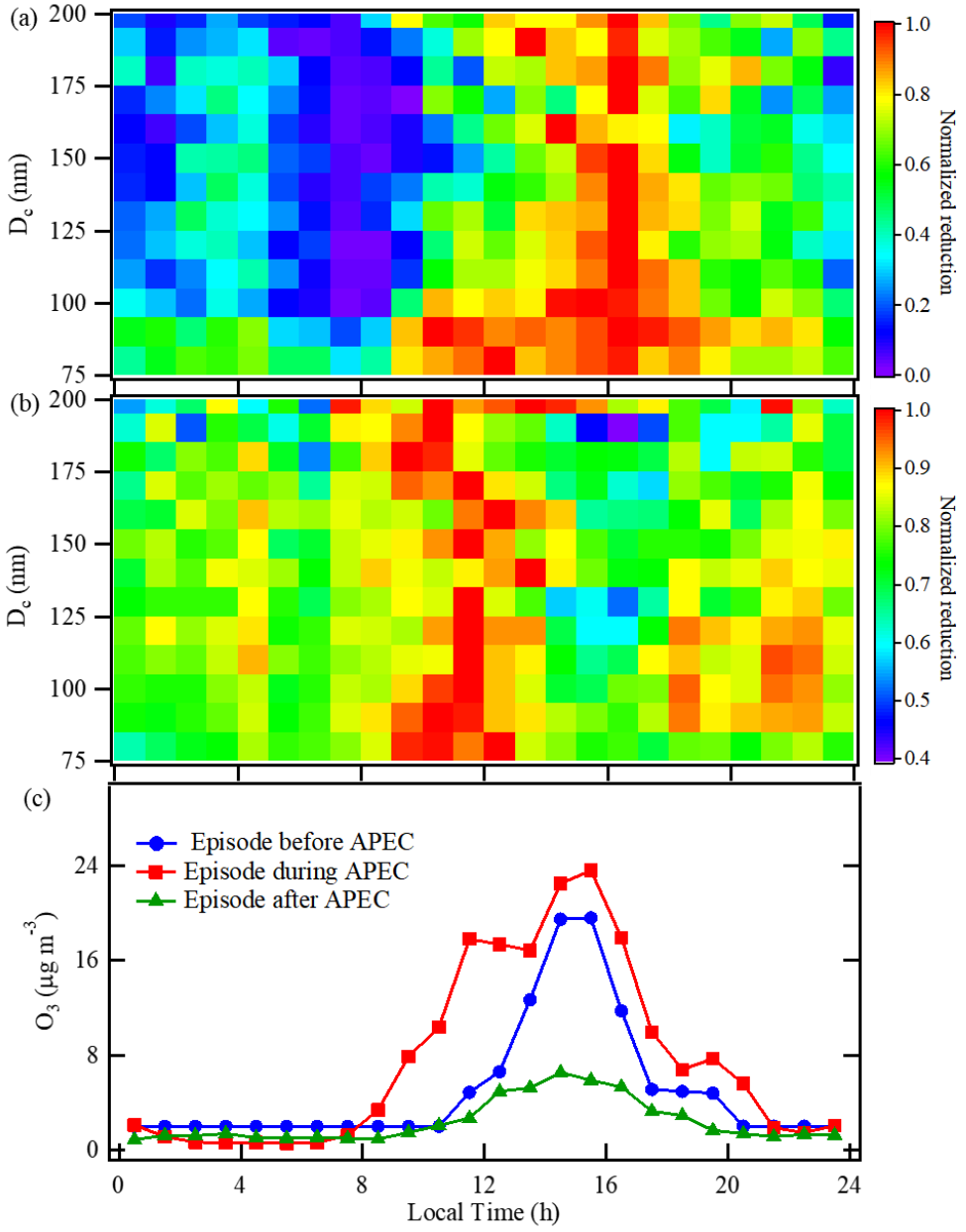


Figure 4. Diurnal cycle of the normalized reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles for the pollution episode during APEC relative to ~~that~~ those (a) before and (b) after APEC. (c) Diurnal cycle of O_3 concentration for the pollution episodes before, during and after APEC

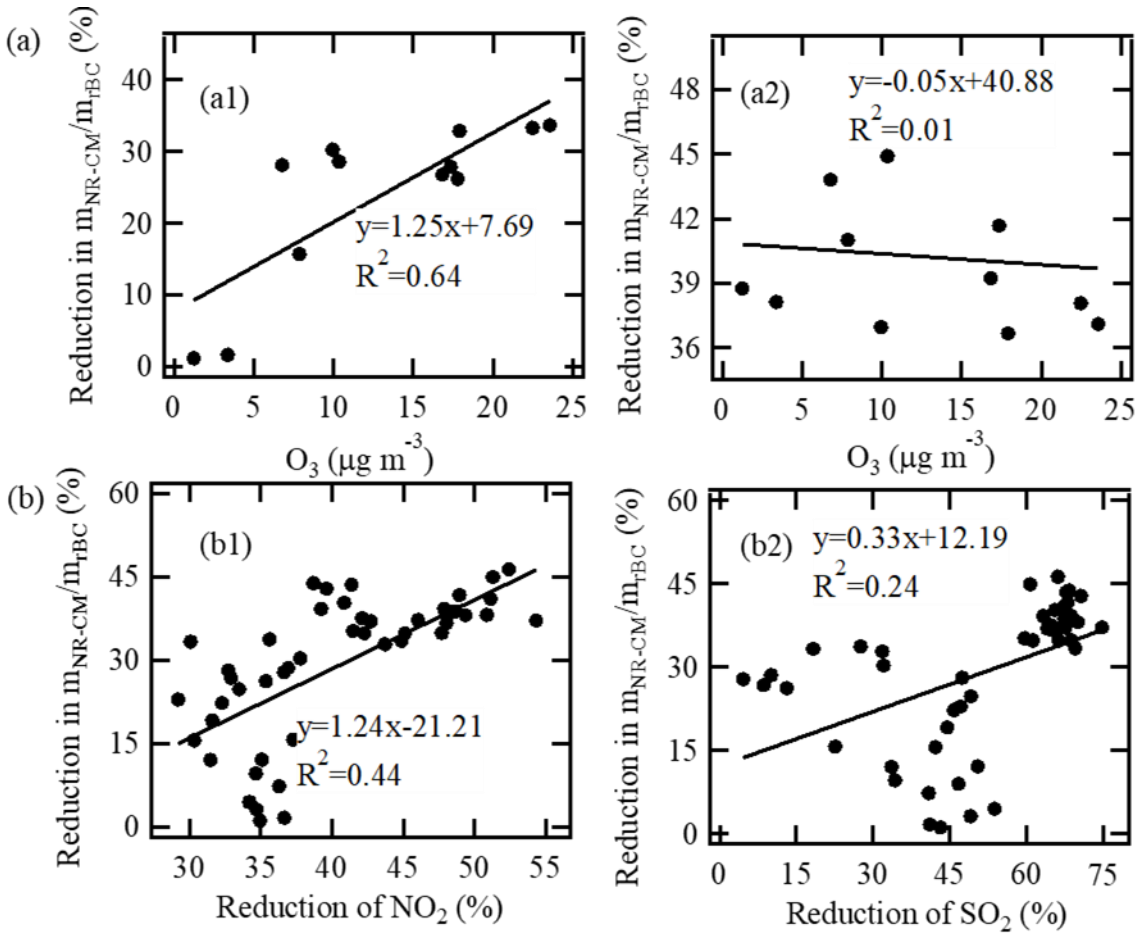
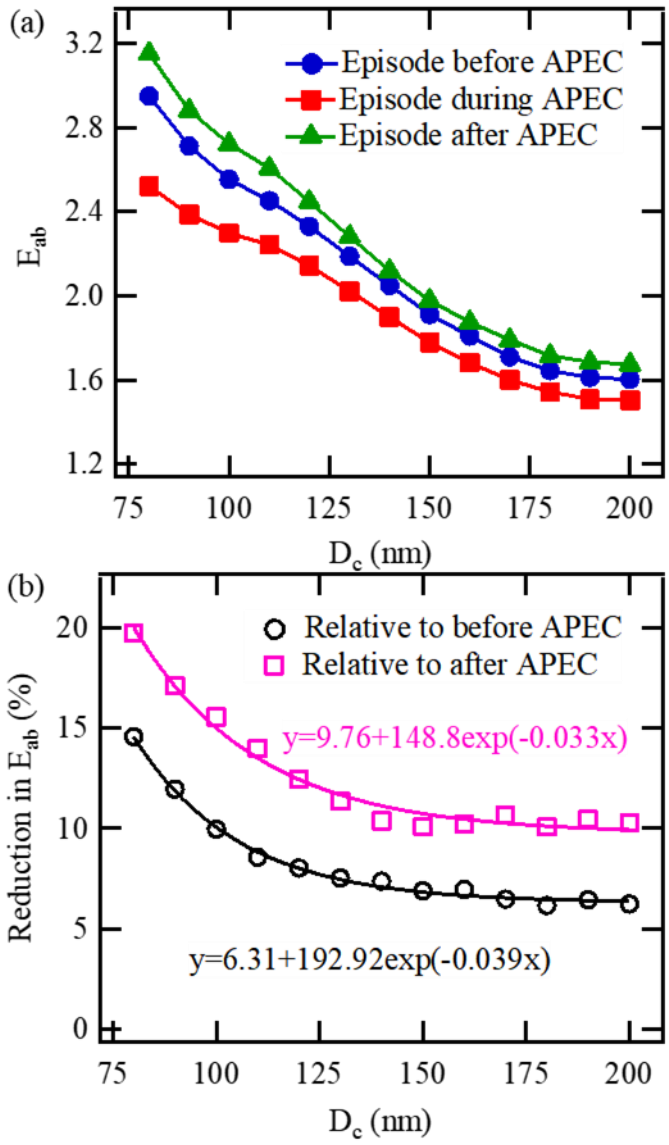


Figure 5. (a) Correlation between the reduction in $m_{NR\text{-}CM}/m_{rBC}$ ratio of BC-containing particles for the pollution episode during APEC relative to ~~that~~ those (a1) before and (a2) after APEC and the daytime (7:00-19:00) O_3 concentration during APEC. (b) Correlation between the reduction in $m_{NR\text{-}CM}/m_{rBC}$ during APEC relative to ~~that~~ those before and after APEC and the corresponding reduction in the concentrations of (b1) NO_2 and (b2) SO_2.


(a)
Episode before APEC
Episode during APEC
Episode after APEC
E_{ab}
3.2
2.8
2.4
2.0
1.6
1.2
75
100
125
150
175
200
D_c (nm)
(b)
Relative to before APEC
Relative to after APEC
y=9.76+148.8exp(-0.033x)
y=6.31+192.92exp(-0.039x)
Reduction in E_{ab} (%)
20
15
10
5
0
75
100
125
150
175
200
D_c (nm)

1

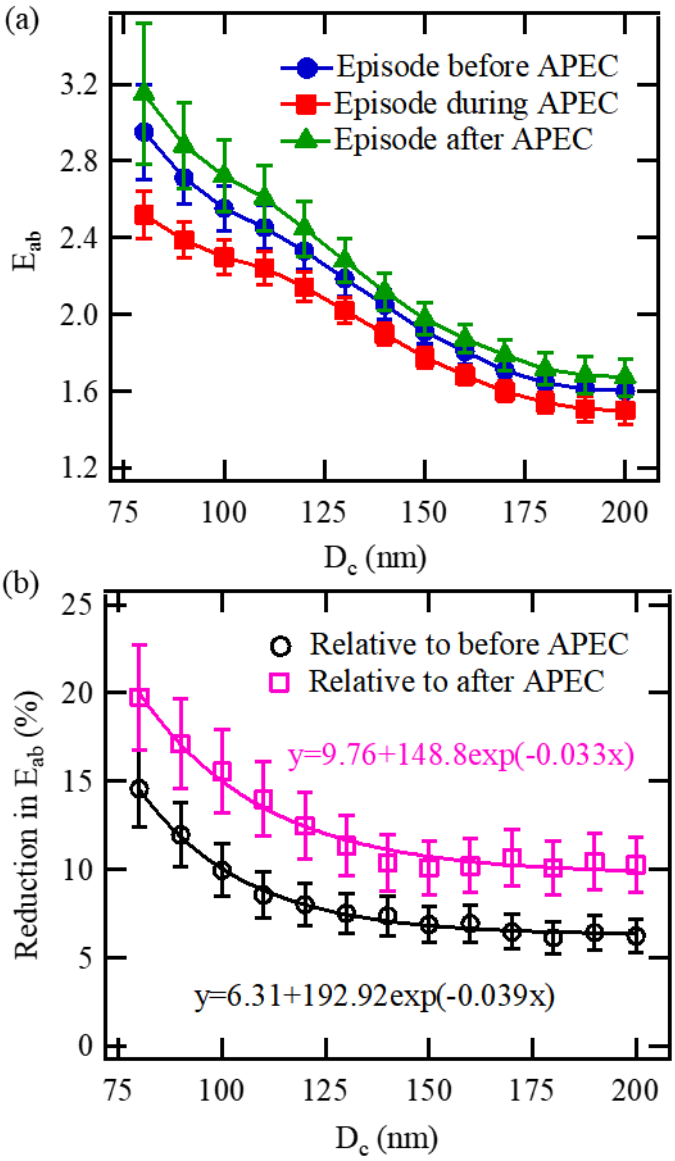
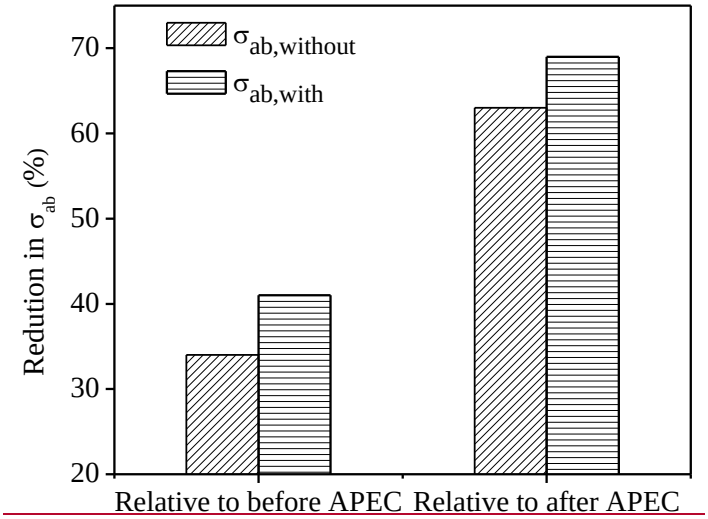
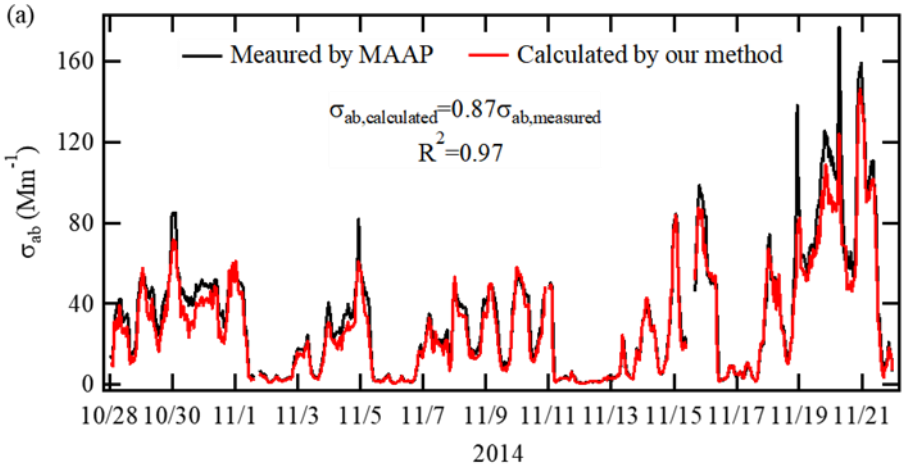
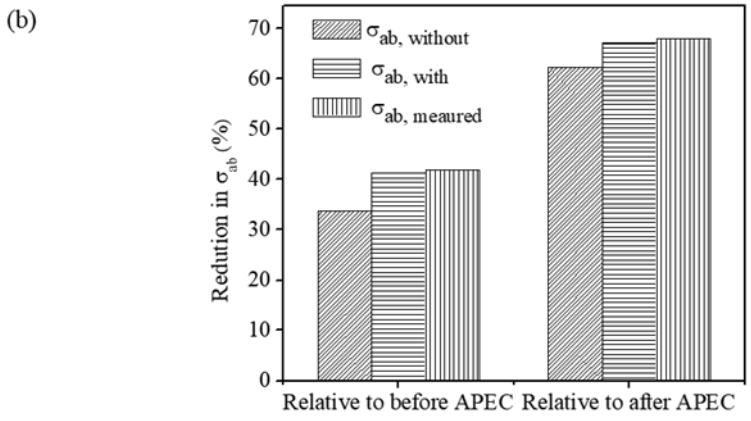


Figure 6. Comparison of the light-absorption capability of BC-containing particles during the day for the pollution episodes before, during and after APEC: (a) light absorption enhancement (E_{ab}) of BC-containing particles and (b) the reduction in E_{ab} of BC-containing particles during APEC relative to that before and after APEC. The error bar shown in (b) represents the uncertainties (~15%) from Mie calculation.


σab,without
σab,with
Redution in σab (%)
70
60
50
40
30
20
Relative to before APEC
Relative to after APEC

1


(a)
Meaured by MAAP
Calculated by our method
σab,calculated=0.87σab,measured
R2=0.97
σab (Mm-1)
160
120
80
40
0
10/28 10/30 11/1 11/3 11/5 11/7 11/9 11/11 11/13 11/15 11/17 11/19 11/21
2014


(b)
σab, without
σab, with
σab, meaured
Redution in σab (%)
70
60
50
40
30
20
10
0
Relative to before APEC
Relative to after APEC

2

Figure 7. (a) The light absorption coefficient (σ_{ab}) at 670 nm. (b) Reduction in the absorption coefficients (σ_{ab}) ~~at 550 nm~~ of BC-containing particles observed in the pollution episode during APEC relative to ~~that~~ those 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 $\sigma_{ab,\ calculated}$ ~~σ_{ab}~~ values with/without, respectively, considering the differences of light-absorption capability of ambient BC-containing particles among the episodes before, during and after APEC.

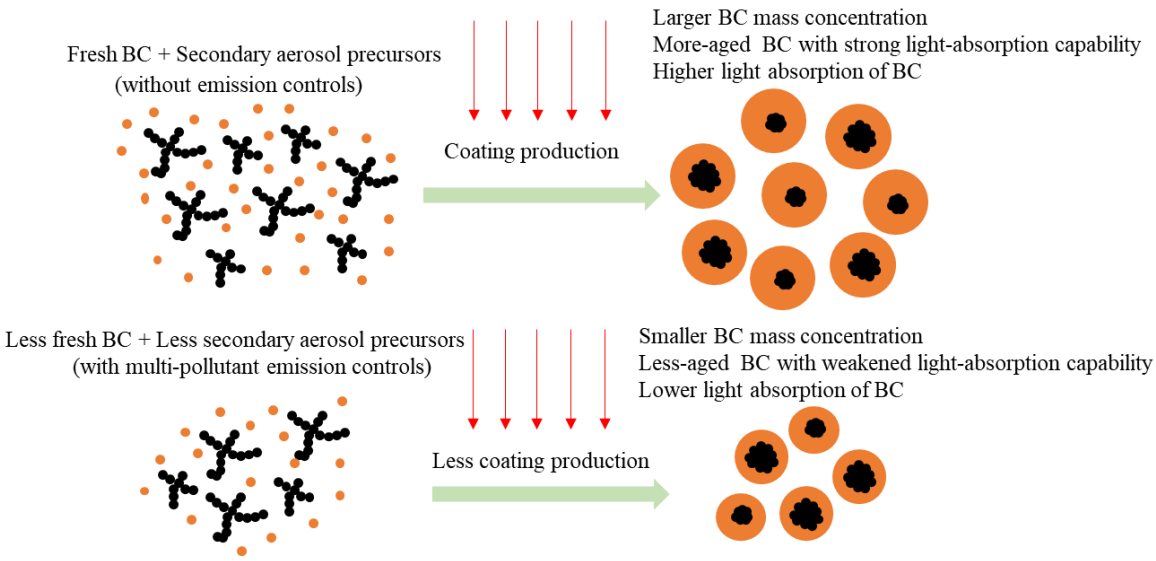


Figure 8. Conceptual scheme of the reduction in light absorption of BC-containing particles due to multi-pollutant emission controls.

Supplement of

Reduction in black carbon light absorption due to multi-pollutant emission control during APEC China 2014

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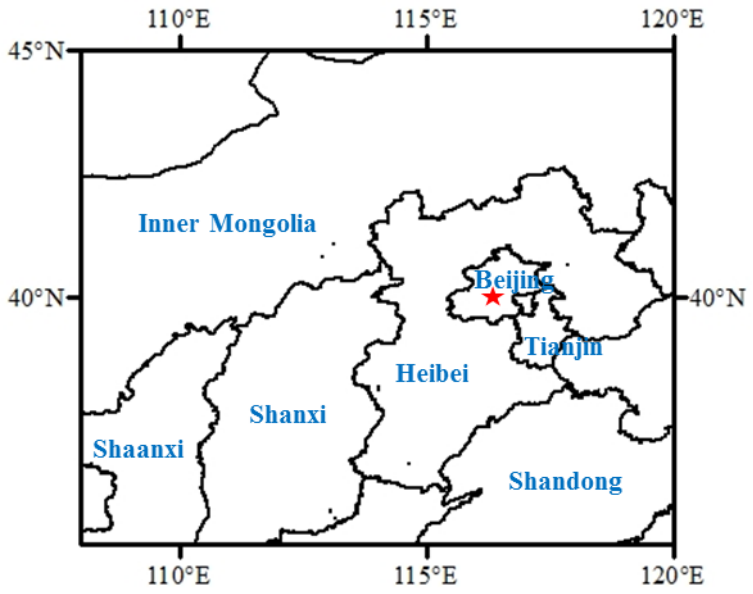

Figure S1. Location of the observation site (red star).

Figure S2 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. For "APEC blue", the emission control measures were implement on November 3-12, 2014. Considering BC over the site mainly from emission within 3 days, 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.

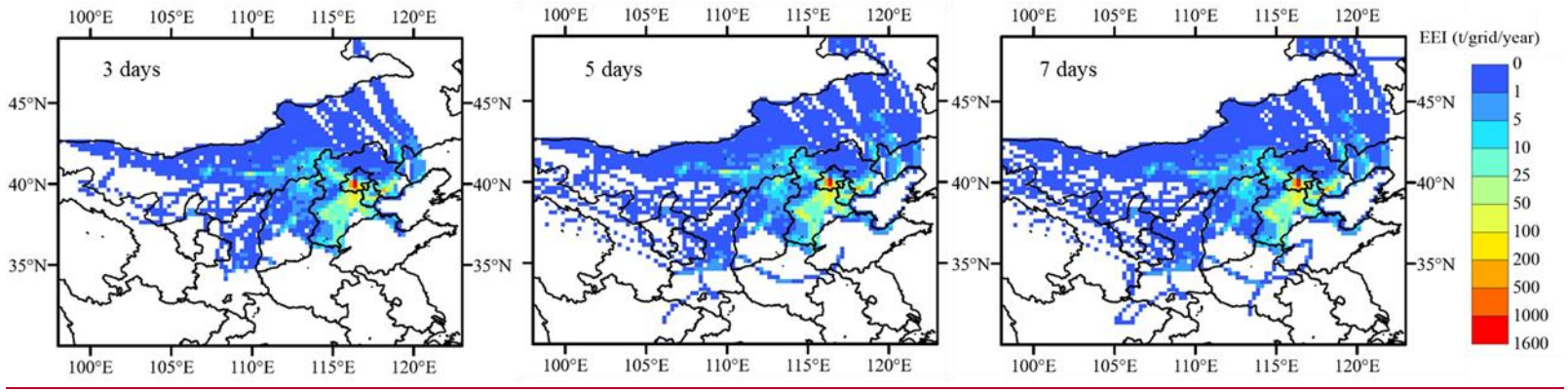

Figure S2. Spatial distribution (0.25°×0.25°) of the effective emission intensity (EEI, defined by Lu et al. (2012)) for BC transported to the site (40º00'17" N, 116º19'34" E) based on 3, 5 and 7 days back-trajectory. The EEI calculation was stated in our previous study (Zhang et al. 2018).

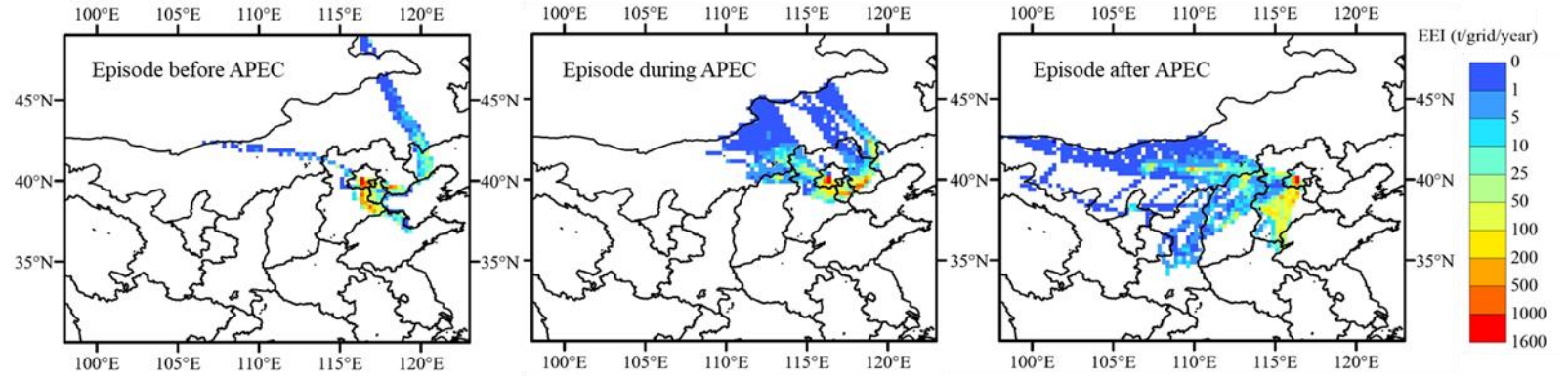

Figure S3. Spatial distribution (0.25°×0.25°) of effective emission intensity (EEI) for BC transported to the site (40º00'17" N, 116º19'34" E) for the pollution episodes before, during and after APEC. The EEI was obtained based on a novel back-trajectory analysis (Lu et al. 2012).

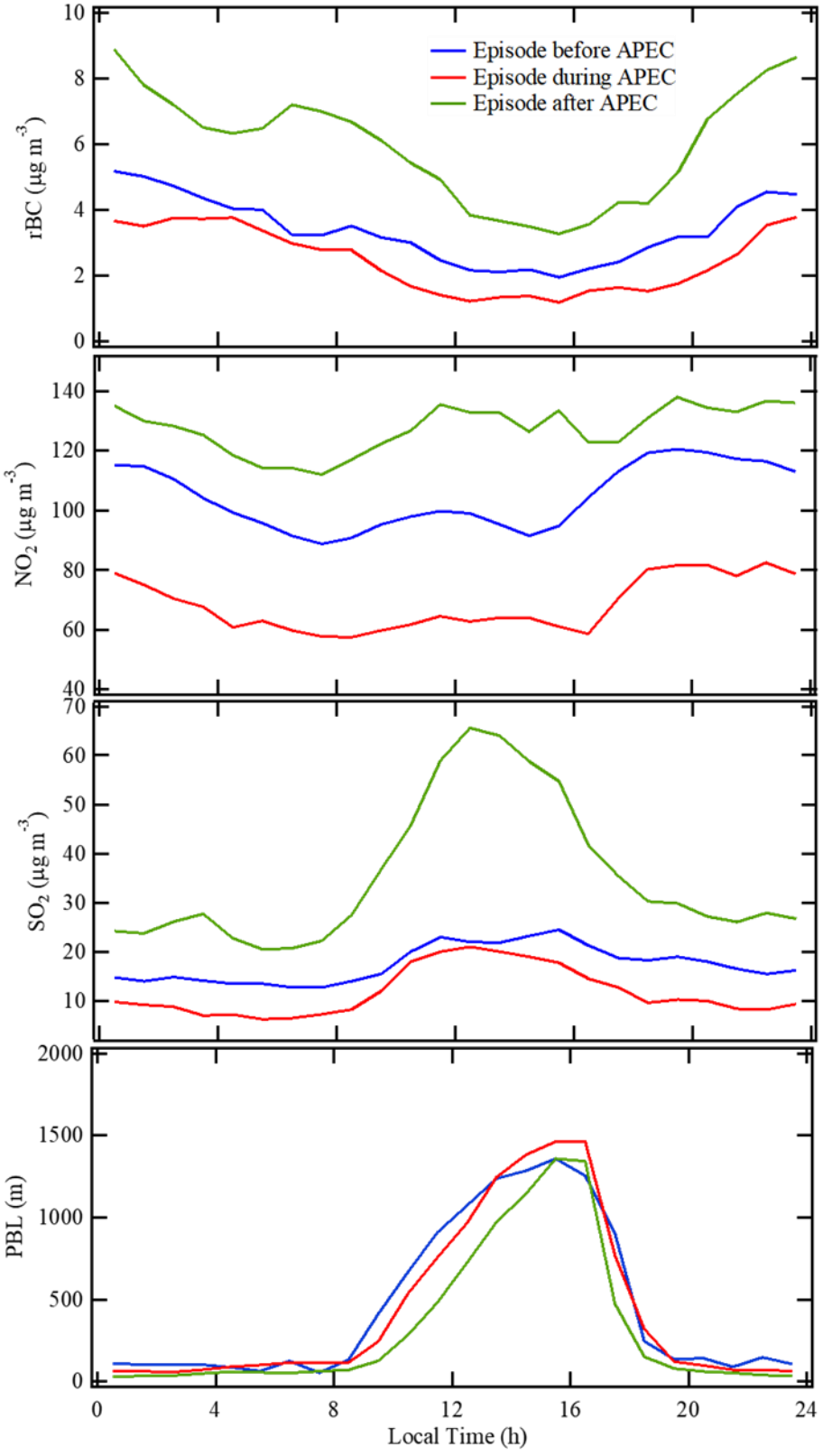

Figure S4. Diurnal variations of the rBC, NO_2 and SO_2 concentrations and the PBL for the pollution episodes before, during and after APEC.

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