

# Reduced light absorption of black carbon (BC) and its influence on BC-boundary-layer interactions during “APEC Blue”

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**Abstract.** Light absorption and radiative forcing of black carbon (BC) is influenced by both BC itself and its interactions with other aerosol chemical compositions. Although the changes in BC concentrations in response to emission reduction measures have been well documented, the influence of emission reductions on the light absorption properties of BC and its influence on BC-boundary-layer interactions has been less explored. In this study, we used the online coupled WRF-Chem model to examine how emission control measures during APEC affect the mixing state/light absorption of BC, and the associated implications for BC-PBL interactions. We found that both the mass concentration of BC and the BC coating materials declined during the APEC week, which reduced the light absorption and light absorption enhancement ( $E_{ab}$ ) of BC. The reduced absorption aerosol optical depth (AAOD) during APEC were caused by both the declines in mass concentration of BC itself (52.0%), and the lensing effect of BC (48.0%). The reductions in coating materials (39.4%) dominated the influence of lensing effect, and the reduced light absorption capability ( $E_{ab}$ ) contributed 3.2% to the total reductions in AAOD. Reduced light absorption of BC due to emission control during APEC enhanced planetary boundary layer height (PBLH) by 8.2 m. Different responses of  $PM_{2.5}$  and  $O_3$  were found to the changes in light absorption of BC. Reduced light absorption of BC due to emission reductions decreased near surface  $PM_{2.5}$  concentrations but enhanced near surface  $O_3$  concentrations in the North China Plain. These results suggest that current measures to control  $SO_2$ ,  $NO_x$ , etc. would be efficient to reduce the absorption enhancement of BC, and to inhibit the feedback of BC on boundary layer. Yet

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enhanced ground O<sub>3</sub> might be a side effect of current emission control strategies. How to control emissions to offset this side effect of current emission control measures on O<sub>3</sub> should be an area of further focus.

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## 1 Introduction

Black carbon (BC) in the atmosphere is produced both naturally and by human activities, attributable to the incomplete combustion of hydrocarbons (*Bond et al., 2013; Ramanathan and Carmichael, 2008*). In addition to contributing considerably to particulate matter and degraded air quality, it is the dominant absorber of visible solar radiation, playing a unique and pivotal role in the Earth's climate system (*Bond et al., 2013; Menon et al., 2002; Ramanathan and Carmichael, 2008; Yang et al., 2019*). The absorption of BC occurs not only in the atmosphere, but when it is deposited over snow or ice, it triggers positive feedbacks and exert a positive radiative forcing (*Flanner et al., 2007; Grieshop et al., 2009*). The direct radiative forcing of atmospheric black carbon was estimated to be 0.4 W m<sup>-2</sup> (0.05-0.8 W m<sup>-2</sup>) (*IPCC, 2014*), and BC has been targeted in emission control policies to mitigate both air pollution and global warming (*Grieshop et al., 2009*).

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Before the 1950s, intense emissions of BC were concentrated in North America and Western Europe. In recent decades, South and East Asia have emerged to become major source regions (*Ramanathan and Carmichael, 2008*). BC emitted from China is responsible for a quarter of the total global emissions (*Bond et al., 2004*). Chemical transport model simulations suggest that the residential sector is the leading source for mass concentration of BC in China, followed by the industrial sector (*Li et al., 2016*). Mean BC direct radiative forcing in China is ~1.22 W m<sup>-2</sup>, more than three times the global mean forcing (*Li et al., 2016*), with two-thirds to three fourths of which contributed by local emissions of BC in China, and the rest by emissions in other countries (*Li et al., 2016; Yang et al., 2017*).

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Specific policies to address BC emissions have not been implemented in China, yet multiple measures targeting PM<sub>2.5</sub> reduction have resulted in declines in BC (*Gao et al., 2018b; Yamineva and Liu, 2019*). A number of observational studies have revealed the declining trend of BC concentrations in China in recent years (*Ji et al., 2018, 2019a, 2019b; Qin et al., 2019*). From 2013 to 2018, the annual mean BC concentrations in Beijing declined from 4.0 µg m<sup>-3</sup> to 2.6 µg m<sup>-3</sup> (*Ji et al., 2019b*). Associated changes in BC radiative forcing can be expected from declines in mass concentration of BC in China, while the radiative forcing of BC is influenced also by the changes in other aerosol components.

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BC absorption is closely connected with the aging process, which is defined as the interaction between BC and other aerosol chemical compositions (*Jacobson, 2001*). After being emitted from combustion processes, BC particles can coagulate and grow by condensation, during which both self-coagulation and hetero-coagulation happen (*Jacobson, 2001*). Although BC is mixed internally with other components, the system is impossible to be well-mixed due to the irregular shape of BC (*Jacobson, 2000*). A core-shell morphology is commonly established, with BC as the core and the coating materials (organics, sulfate, etc.) as the shell (*Jacobson, 2001; Zhang et al., 2018*). Numerous efforts have been made to explore the influence of aerosol components on internally mixed BC absorption (*Cappa et al., 2012; Chen et al., 2021; Bond*

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70 *et al.*, 2006; Fuller *et al.*, 1999; Jacobson, 2001; Liu *et al.*, 2017; Peng *et al.*, 2016). It was proposed that the coating  
components (shell) could act as a lens to focus more photons onto the core to enhance the light absorption of BC (Fuller *et al.*,  
*et al.*, 1999). Bond *et al.* (2006) estimated that this lensing effect would increase the light absorption of BC by 50-100%.  
Jacobson (2001) reported a global average BC absorption enhancement factor of 2, whereas other values, from negligible  
(Cappa *et al.*, 2012) to as high as 2.4 (Peng *et al.*, 2016) were also found previously. This lensing effect has been recognized  
75 also as an important factor affecting radiative forcing of BC (Jacobson, 2001). In the recent decade, efforts were made also  
to develop models to predict the dynamic evolution of aerosol mixing states (Ching *et al.*, 2016; Curtis *et al.*, 2017; Matsui  
*et al.*, 2013; Tian *et al.*, 2014). For example, Matsui *et al.* (2013) developed a 2-D aerosol bin scheme that can resolve BC  
mixing state and BC aging processes. However, these approaches are far too computationally expensive for use in regional  
3-D models (Barnard *et al.*, 2010).

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80 Over the past several years, the State Council of China has issued a comprehensive Air Pollution Prevention and  
Control Action Plan (APPCAP), covering major emission sectors (Liang *et al.*, 2017; Zhang *et al.*, 2019a). Long-term  
observations of aerosol chemical composition indicate that both concentrations of BC and other coating components have  
declined rapidly (Gao *et al.*, 2020b; Ji *et al.*, 2019b; Zhou *et al.*, 2019). Although the changes in BC concentrations in  
response to emission reduction measures have been documented (Ji *et al.*, 2019b; Gao *et al.*, 2020b), the influence of  
85 emission reductions on the aging processes and light absorption of BC has been less explored (Zhang *et al.*, 2019b). Zhang  
*et al.* (2018) observed that the declines in absorption of BC was mainly dominated by decreases in BC mass concentration  
(86%), and the weakening of BC light absorption capability also played a role (14%). However, this finding was formulated  
based on surface observations, little is known about the changes at upper layers. Given the importance of BC absorption in  
the upper boundary layer as to buildup of pollution (Ding *et al.*, 2016), the impact of emission reductions on the light  
90 absorption of BC, and its implications for the development of boundary layer and pollution episodes need further  
investigations. On November 5-11, 2014, Beijing, China hosted the Asia-Pacific Economic Cooperation (APEC) meeting,  
during which Beijing and surrounding regions cooperated to implement short-term emission control measures to ensure good  
air quality. This event offers a great opportunity to study physical and chemical responses of atmospheric composition to  
emission reductions.

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95 In this study, we address the following questions using the APEC event as a study case: (1) how did emission  
reductions affect the aging processes and light absorption of BC during APEC; (2) what were the relative contributions of  
reduced mass concentrations of BC, aging processes of BC, and reshaped mixing state of BC to the changes in light  
absorption of BC during APEC; and (3) how did these processes affect BC-PBL interactions and formation of air pollution?  
In Sect. 2, we describe the WRF-Chem model configurations and observational datasets used in this study. Results are  
100 presented in Sect. 3, and conclusions/discussions are provided in Sect. 4.

## 2 Methods and data

### 2.1 WRF-Chem model configuration

WRF-Chem model (Grell *et al.*, 2005) version 3.8.1 was adopted in this study to simulate emission, chemical transformation and deposition of aerosols, as well as their interactions with radiation. We demonstrated in previous studies (Gao *et al.*, 2016a, 2016b, 2020b, 2020c) that the spatio-temporal variations of air pollutants over China could be reproduced well by WRF-Chem. WRF-Chem enables multiple options for gas phase chemistry and aerosol modules (Grell *et al.*, 2005). We employed the Carbon Bond Mechanism version Z (CBMZ) gas phase chemistry (Zaveri and Peters, 1999) coupled with the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) (Zaveri *et al.*, 2008) aerosol module in this study. MOSAIC treats size resolved aerosol species, and we used 8 bins version in this study, corresponding to the particle diameter ranges of 0.039-0.078, 0.078-0.156, 0.156-0.312, 0.312-0.625, 0.625-1.25, 1.25-2.5, 2.5-5.0, 5.0-10.0  $\mu\text{m}$ , respectively. Secondary organic aerosol (SOA) formation in MOSAIC was simulated with volatility basis set (VBS) (Shrivastava *et al.*, 2011). We configured two nested domains with horizontal resolutions of 81km and 27km, and 31 vertical layers up to a pressure level of 50hPa. The configured domains cover most areas of East Asia and focus on the North China region (same as Figure 1 in Gao *et al.*, 2017). Other chosen options for key physical parameterizations follow Gao *et al.* (2016b). Meteorological initial and boundary conditions were provided by the NCEP  $1^\circ \times 1^\circ$  degree final reanalysis dataset (FNL), and chemical initial and boundary conditions were obtained from the MOZART global chemistry simulations (Emmons *et al.*, 2010). [We used the YSU scheme in WRF-Chem to simulate the evolution of boundary layer, and the YSU scheme defines the top of the boundary layer using Richardson number \(Hong \*et al.\*, 2010\).](#) To allow the effects of aerosol on meteorological conditions in the model, we did not apply observational nudging or reanalysis nudging.

Anthropogenic emissions of particles and gases in China in the model were taken from the Multi-resolution Emission Inventory for China (MEIC) for year 2014 developed by Tsinghua University (Zheng *et al.*, 2018). Anthropogenic emissions for areas outside China were obtained from the MIX Asian emission inventory developed for MICS-Asia and HTAP, which combines five emission inventories for Asia (Li *et al.*, 2017). Both MEIC and MIX datasets provide monthly emissions of air pollutants at  $0.25^\circ \times 0.25^\circ$  grids, which were interpolated to WRF-Chem modeling domains in this study. We adopted the MEGAN model version 2.04 to estimate biogenic emissions of gases and particles online (Guenther *et al.*, 2006). The Global Fire Emissions Database version 4 (GFEDv4) (Giglio *et al.*, 2013) were used as open fire emissions.

We simulated the period from October 16 to November 13, and discarded the first seven days as spin-up to avoid the influences of initial conditions. To explore the influences of coordinated emission control measures on BC absorption, we conducted multiple sets of simulations, as described in Table 1. For the NOCTL experiments, simulations were conducted with no perturbations in emissions. For the CTL experiments, emissions of  $\text{SO}_2$ ,  $\text{NO}_x$ ,  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$ , VOCs, and other species in Beijing were reduced by 39.2%, 49.6%, 66.6%, 61.6%, 33.6%, and 50%, respectively, over November 3-11 period. Emissions in Inner Mongolia, Shanxi, Hebei, Tianjin, and Shandong were reduced by 35%. These perturbation

factors were taken from the BMEPB reports (Gao et al., 2017). The locations of these provinces are marked in Figure 1 in

135 Gao et al. (2017).

The influences of BC absorption under different assumptions, including external/core-shell mixing and with/without emission reductions ( $\Delta_{BC-Ext-NOCTL}$ ,  $\Delta_{BC-Ext-CTL}$ ,  $\Delta_{BC-CS-NOCTL}$ ,  $\Delta_{BC-CS-CTL}$ , and  $\Delta_{BC-CYSN-CS}$ ), can be derived with equations (1-5) below. The description of each simulation is documented in Table 1.

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$$\Delta_{BC-Ext-NOCTL} = NOCTL_{Ext} - NOCTL_{Ext-nobc} \quad (1)$$

$$\Delta_{BC-Ext-CTL} = CTL_{Ext} - CTL_{Ext-nobc} \quad (2)$$

$$\Delta_{BC-CS-NOCTL} = NOCTL_{CS} - NOCTL_{CS-nobc} \quad (3)$$

$$\Delta_{BC-CS-CTL} = CTL_{CS} - CTL_{CS-nobc} \quad (4)$$

$$\Delta_{BC-CS-CYSN} = CYSN_{CS} - CYSN_{CS-nobc} \quad (5)$$

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The influences of emission reductions during APEC on changes in light absorption of BC and associated changes in meteorological/pollution conditions under external/core-shell mixing assumptions ( $\Delta_{emission-Ext}$  and  $\Delta_{emission-CS}$ ) can be inferred with equations (6-7) below. We use equation (8) to derive the impact of changed BC aging processes by comparing the differences between core-shell simulation and external mixing simulation. The influences of reduced coating due to emission control measures during APEC are calculated with equation (9). We use equation (10) to derive the influences of changes in light absorption enhancement ( $E_{ab}$ ) of BC.

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$$\Delta_{emission-Ext} = \Delta_{BC-Ext-CTL} - \Delta_{BC-Ext-NOCTL} \quad (6)$$

$$\Delta_{emission-CS} = \Delta_{BC-CS-CTL} - \Delta_{BC-CS-NOCTL} \quad (7)$$

$$\Delta_{aging} = \Delta_{BC-CS-CTL} - \Delta_{BC-CYSN-CS} \quad (8)$$

$$\Delta_{coating} = \Delta_{BC-CS-CYSN} - \Delta_{BC-CS-CTL} \quad (9)$$

$$\Delta_{E_{ab}} = \frac{\frac{\Delta_{emission-CS}}{\Delta_{BC-CS-NOCTL}} - \frac{\Delta_{emission-Ext}}{\Delta_{BC-Ext-NOCTL}}}{\frac{\Delta_{emission-CS}}{\Delta_{BC-CS-NOCTL}}} \quad (10)$$

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## 2.2 Calculation of aerosol optical properties in WRF-Chem

WRF-Chem uses Mie theory to calculate layer aerosol optical depth (AOD), single scattering albedo (SSA), and asymmetry factor ( $g$ ). First, the size parameter and spectral refractive index are used to calculate the Mie extinction efficiency  $Q_e$ . Then, the extinction coefficient  $\sigma_e(\lambda)$  is provided by the integral of  $Q_e$  with consideration of the geometric size of the particle ( $\pi r^2$ ) and the particle number size distribution  $n(r)$  (equation (11)).  $\sigma_e(\lambda)$  is a equation of wavelength  $\lambda$ . Similarly, absorption coefficient  $\sigma_a(\lambda)$  and scattering coefficient  $\sigma_s(\lambda)$  can be obtained with Mie absorption efficiency  $Q_a$  and Mie scattering efficiency  $Q_s$ . The value of SSA can be calculated with equation (12) using  $\sigma_a(\lambda)$  and  $\sigma_s(\lambda)$ .

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$$\sigma_e(\lambda) = \int_{r_{min}}^{r_{max}} Q_e \pi r^2 n(r) dr \quad (11)$$

$$SSA(\lambda) = \frac{\sigma_s(\lambda)}{\sigma_s(\lambda) + \sigma_a(\lambda)} \quad (12)$$

The calculated optical properties vary with the assumption of mixing state of aerosols. For external mixing, each particle is assumed to be a single chemical species. There are several models proposed for internal mixing, and the commonly used ones include the volume averaging model and core-shell model. In the volume averaging model, all species are assumed to be well mixed, while the core-shell model assumes that BC is coated by a well-mixed shell of other species (Jacobson, 2001). The volume-weighted refractive index  $m$  is obtained with the equation below:

$$m = \frac{\sum_i V_i m_i}{\sum_i V_i} \quad (13)$$

In equation (13),  $V_i$  denotes the volume of species  $i$  and  $m_i$  represents the refractive index of species  $i$ . For core-shell internal mixing, similar averaging processes are applied to the core and shell separately. The scattering efficiency, absorption efficiency and asymmetry parameter are then obtained using the core-shell Mie theory documented in Ackerman and Toon (1981). Core-shell Mie calculation requests core radius, shell radius, refractive index of core and refractive index to shell as inputs (Ackerman and Toon, 1981; Toon and Ackerman, 1981). The official version of WRF-Chem does not calculate optical properties of aerosols with external mixing assumption. To assess the influence of aging process mixing with coating particles on the light absorption of BC, estimated light absorption of pure BC with external mixing assumption is required. We modified the optical calculation module in WRF-Chem so that it does not mix BC with other chemical species in the calculation of optical properties, which can be used as optical properties of BC with external mixing assumption. In the calculation of optical properties with internal mixing assumption, the volume-weighted refractive index will be inputs of the Mie code. In the calculation of optical properties of BC with external mixing assumption, we only allowed BC to go through the Mie code. The mass and particle number of BC, denoted in  $M_i$  and  $N_i$  in each bin  $i$  (1 through 8) are computed first, and the volume ( $V_i$ ) are obtained then by dividing by the density of BC. During the calculation of physical diameter (equation 14), other chemical species are not considered. Accordingly, BC does not mix with other chemical species, and we named it as optical properties of BC with external mixing assumption here. It should be noted that this calculation is different from treatments of external mixing in other models that all particles are separated from each other. In our calculation, BC within each bin are internally mixed, although it does not mix with other chemical species. As the purpose of this study is to explore how coating particles on BC would affect the absorption of BC, this treatment would not be an issue.

$$D_i = 2 \left( \frac{V_i}{\frac{4}{3} \pi} \right)^{\frac{1}{3}} \quad (14)$$

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## 2.3 Observations

Both observations of meteorological variables and air pollutants were used to evaluate the performance of model over the APEC study period in *Gao et al. (2017)* and in this study. The meteorological measurements were retrieved from the National Centers for Environmental Information website (<https://gis.ncdc.noaa.gov/maps/ncei#app=cdo>), which includes near surface temperature, relative humidity (RH), wind speed, and wind direction. The hourly surface concentrations of PM<sub>2.5</sub> and daily PM<sub>2.5</sub> chemical compositions were measured at the Institute of Atmospheric Physics (IAP), Chinese Academy of Sciences (CAS) site (*Liu et al., 2017; Yang et al., 2020*). We obtained also AAOD (absorption aerosol optical depth) from the AERONET network (*Dubovik and King, 2000; Holben et al., 1998*) to evaluate model performance. We derived AAOD at 440nm based on Angstrom exponent relation (*Schuster et al., 2006*) to make it consistent with observations. Data from more than 500 sites across the world are provided online at the AERONET website (<http://aeronet.gsfc.nasa.gov>).

## 3 Results

### 3.1 Model Evaluation

Model evaluation was conducted with surface observations of meteorological variables, PM<sub>2.5</sub>, PM<sub>2.5</sub> chemical components, and AAOD. Data at two meteorological sites in urban Beijing were averaged, and were compared against the model values for the domain grid cell containing the monitoring site. Figure 1 indicates that the daily mean temperature and relative humidity (RH) are captured well by the model. Observed strong wind conditions are slightly underestimated, which is a common issue due to inaccurate land use inputs or other problems in the model (*Gao et al., 2018a*). Our previous investigation (*Gao et al., 2017*) suggested that temperature and RH were lower and northerly winds became more frequent from before APEC to during APEC periods, contributing to pleasant air quality. Figure 2(a) displays the simulated and observed hourly PM<sub>2.5</sub> concentrations in urban Beijing. Before APEC, observed high PM<sub>2.5</sub> concentration is well captured by our model. During APEC, the NOCTL case overestimates PM<sub>2.5</sub> concentrations, while the CTL case exhibits better agreement with observations. Implementing emission reductions in the model lowers the mean bias of the model from 30.8 to -4.0  $\mu\text{g m}^{-3}$ . The performance of WRF-Chem in simulating wintertime PM<sub>2.5</sub> chemical compositions was explored extensively in our previous investigations (*Gao et al., 2016b, 2018a*). Similarly, measured high concentrations of inorganic aerosols (sulfate, nitrate and ammonium) are underestimated, which could be partly due to missing sulfate formation pathways (*Cheng et al. 2016*). We used the updated version with heterogeneous sulfate formation (*Gao et al., 2016a*) to

225 reduce the underestimation of sulfate in this study. Simulated BC concentration shows high degree of consistency with  
observations, while OC is slightly underestimated due to large uncertainties in current status of SOA modeling (Figure 2f).  
In general, the temporal variations and magnitudes of air pollutants are well represented in our model. Figure 1(d) compares  
simulated AAOD with external mixing assumption and core-shell model against AERONET inferred AAOD during the  
APEC study period. AAOD simulated with external mixing assumption exhibits much lower values than observation. With  
230 the core-shell model, this underestimation is largely reduced. However, AAOD is still underestimated by the model, which  
might be caused by missing sources of absorbing particles in the model. Currently, the absorption of organics is not treated  
in the WRF-Chem model, which is likely to underestimate the light-absorbing capability of carbonaceous aerosols in the  
atmosphere (Andreae and Gelencser, 2006). Uncertainties in the aerosol size distribution in emissions may also contribute to  
this mismatch between the model and observations (Matsui, 2006).

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### 3.2 Reductions in the concentrations of BC/coating pollutants and changes in BC aging degree

Previously, the reductions of air pollutants were estimated by comparing concentrations of air pollutants during the  
APEC period with those during other periods. Given the differences in meteorological conditions, such a comparison is not  
able to indicate the influence of emission control measures. As displayed in Figure 2(a), the concentrations during October  
24-25 can be two times of those during October 26-27, although no emission reduction measures were implemented.  
240 Previously, we concluded that the meteorological conditions during the APEC week were generally favorable for good air  
quality compared to it during the week before the APEC week (Gao *et al.*, 2017). Thus, we perturbed emissions in this  
study to examine how it would affect concentrations of air pollutants, including both BC and its coating pollutants. As  
displayed in Figure 3(a-b), mean concentrations of SO<sub>2</sub> and NO<sub>2</sub> in urban Beijing declined by 38.7% and 36.3%,  
245 respectively, in response to short-term emission control measures. Based on observations, Zhang *et al.* (2018) reported that  
SO<sub>2</sub> concentrations decreased by 35% (67%) and NO<sub>2</sub> concentrations decreased by 34% (45%) compared with that before  
(after) APEC. These declines in aerosol precursors would have modified secondary aerosol formation during the APEC  
week. Our model indicates that sulfate and nitrate declined by 40.0% and 28.2%, respectively. Given the slight  
underestimation of sulfate and nitrate, these values might have been moderately underestimated. Mass concentrations of BC  
250 declined by 34.6%, while the abundance of OC in the atmosphere exhibited a larger reduction (44.2%).

~~The changes in BC aging process is determined by both the decrease in BC and primary/secondary pollutants  
condensed on BC.~~ We used the ratio of the sum of pollutants (primary as well as secondary) to black carbon concentrations  
(*r*BC) to track the ~~changes of BC aging degree~~ relative abundance of BC and non-BC particles, which is essential in the C-S  
calculation:

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$$rBC = \frac{[sulfate]+[nitrate]+[ammonium]+[organics]+[dust]+[sodium]+[chloride]}{[BC]} \quad (14)$$



As shown in Figure 3(c), the impacts of emission reductions during APEC on  $rBC$  behave differently at different sizes. For ultrafine particles, emission reductions generally lower the aging degree of BC. This is consistent with the observational evidence that smaller BC cores show larger reductions in aging degree as a result of emission control measures during APEC (Zhang *et al.*, 2018). As most secondary aerosols are in smaller sizes, the effect of emission reduction on BC aging is more significant for smaller particles. Zhang *et al.* (2018) reported only the changes in sizes below 0.2  $\mu\text{m}$ , our modeling results suggest, however, that the aging degree of BC might be enhanced under emission reductions for relatively larger particles (Figure 3c). The impact of emission reductions on  $rBC$  behaves differently near the surface and at higher layers (Figure 3d). The aging degree is lowered in the CTL case near the surface, mainly due to reductions in coating materials. However, at layers higher than 200 meters, the aging degree of BC increases with emission reductions. In-situ near surface measurements indicate also that  $rBC$  was reduced during APEC, and the reduction was most likely caused by lower photochemical production (Zhang *et al.*, 2018).

### 3.3 Changes in AAOD and the light-absorption enhancement ( $E_{ab}$ ) of BC during APEC

$rBC$  values describe the aging degree of BC, while the exploration of how emission reductions affect light absorption of BC requires a sophisticated calculation of optical properties of BC. Mie theory is commonly used to calculate the light absorption enhancement of BC ( $E_{ab}$ ) from lensing effect with a core-shell model. Zhang *et al.* (2018) estimated  $E_{ab}$  by dividing the light-absorption cross section of the whole BC-containing particle by that of BC core at a certain wavelength. Here we follow the method in Curci *et al.* (2019), and calculate  $E_{ab}$  as the ratio of BC AAOD estimated assuming core-shell internal mixing to that calculated with external mixing assumption (enhancement due to mixing with coating non-BC particles):

$$E_{ab} = \frac{BC\_AAOD(550nm,core-shell\ mixing)}{BC\_AAOD(550nm,external\ mixing)} \quad (\text{AAOD can be either layer or column}) \quad (15)$$

Inferred vertical profiles of layer  $E_{ab}$  values in the CTL and NOCTL scenarios are displayed in Figure 3e. At the layers below 5km, mean  $E_{ab}$  values are 1.96 and 1.95 for CTL and NOCTL scenarios, respectively. Below 500 m in the troposphere, emission reductions during APEC lower  $E_{ab}$  from 2.11 to 2.06. Previous study by Jacobson (2001) suggests a global average BC absorption enhancement factor of 2, which is consistent with current study. However, a wide range of enhancement factors have been reported, from negligible (Cappa *et al.*, 2012) to as high as 2.4 (Peng *et al.*, 2016). Liu *et al.* (2017) pointed that the enhancement factors depend on the particles' mass ratio of non-black carbon to black carbon. Our model results indicate also that the reductions in light absorption enhancement of BC are concentrated at lower layers, while enhancement could happen at higher layers (Figure 3e). This is consistent with the vertical profile of  $rBC$  where it decreases due to emission reductions near the surface while increases at higher layers (Figure 3d).

Figure 4a presents the daytime mean (defined as the mean BC AAOD over 10:00-17:00 time period) BC AAOD in Beijing inferred from simulations with different mixing assumptions and emission perturbations. In the NOCTL scenarios, BC AAOD simulated with core-shell model exhibits higher values than those with external mixing assumption (0.0220 for external and 0.0427 for core-shell). Due to reductions in emissions, these values decline to 0.0145 and 0.0283, respectively.

290 Due to emission reductions (differences between CTL and NOCTL scenarios), mean daytime BC AAOD decrease by 0.0075 during the APEC week, as a result of declines in mass concentration of BC (52.0%, Table 2). However, the lensing effect of BC induces a further decline of 0.0069 (48.0%, Table 2). The influence of lensing effect is dominated by the reductions in coating materials (39.4%, equation (9), Table 2). The BC absorption enhancement ( $E_{ab}$ ) factor decreased by 0.003 due to reductions in emissions (Figure 4b). We further quantified that the reduced light absorption capability ( $E_{ab}$ ) resulting from

295 emission reductions during APEC contributed 3.2% to the total reductions in AAOD (equation (10), Table 2).

### 3.4 Influences on boundary layer process and air pollution

The vertical distribution of BC absorption plays an important role in modulating the temperature gradient and changing boundary layer meteorology (Ding *et al.*, 2016). We conducted a series of numerical experiments to understand the

300 influences of reshaped BC absorption due to emission reductions during APEC on boundary layer process and the formation of air pollution. Figure 4c illustrates the vertical profiles of BC absorption induced changes in equivalent potential temperature (EPT), which is commonly used to indicate the stability of air in the atmosphere (Obremski *et al.*, 1989). When EPT decreases with height, the atmosphere is unstable and vertical motion/convection is likely to occur. In all experiments, BC absorption induces a positive impact on EPT in the air above ground acting to enhance the stability of the atmosphere

305 (Figure 4c). The maximum enhancement occurs at layers close to 1-2km (Figure 4c). At ~2.6km, the maximum ratio of changes with core-shell model to those with external mixing reach above 2.5, indicating the important effects of mixing state of BC in the upper boundary layer (Figure 4c).

In urban Beijing, BC absorption induced mean changes of daytime planetary boundary layer height (PBLH) during the APEC week are -11.6 and -24.0 m for external mixing and core-shell model, respectively (Figure 4d). Under a relatively

310 clean condition (CTL scenarios), these values change to -8.8 and -15.6 m for external mixing and core-shell model for NOCTL emissions (Figure 4d). Due to emission reductions, the impacts of BC absorption on PBL inhibition decrease by 8.2 m on average during the APEC week (reduced emissions enhance PBLH by 8.2 m). The influences of reduced mass concentration of BC itself account for 35% of the total changes, while the lensing effect of BC explain the rest (65%, Table 2). The decreased coating due to emission reductions dominate the lensing effect of BC (47.4%, Table 2).

315 The corresponding changes in daytime mean near surface concentrations of  $O_3$  and  $PM_{2.5}$  in Beijing are displayed in Figure 4e and Figure 4f, respectively. The inhibited development of PBL due to BC absorption results in higher abundance of  $PM_{2.5}$  within the PBL (Ding *et al.*, 2016; Gao *et al.*, 2016b). Previously, we quantified that the co-benefits of reduced aerosol

320 feedbacks could explain ~11% of the total decreases in  $PM_{2.5}$  in Beijing during APEC. Here we focus on light absorption of BC, and find that the lensing effect of BC decreases  $PM_{2.5}$  concentration by  $0.8 \mu\text{g m}^{-3}$  on average (Figure 4f). On average, declines in BC mass concentration itself account for 64.3% of the total impact of reduced light absorption of BC on  $PM_{2.5}$ , while 35.7% is attributed to the lensing effect of BC. However, inhibited PBL development does not necessarily lead to enhanced levels of near surface  $O_3$ , as the formation of  $O_3$  is also affected by changes in aerosols and photolysis reactions above the ground (Chen et al., 2021; Lu et al., 2019). As displayed in Figure 4e, near surface  $O_3$  concentrations in urban Beijing decrease in response to BC absorption.

325 The spatial distribution of  $\Delta_{\text{emission-ext}}$  and  $\Delta_{\text{emission-CS}}$  in Figure 5 reveal that external mixing and core-shell models estimate similar patterns of changes in AAOD, PBLH, near surface  $O_3$  and near surface  $PM_{2.5}$ . However, the responses of these variables are larger in the core-shell model due to lensing effects of coating materials. Reduced emissions of BC and its coating materials during APEC led to declined AAOD, less stabilized PBLH, decreased near surface  $PM_{2.5}$  concentrations and enhanced near surface  $O_3$  concentrations in the North China Plain (Figure 5). It was noted that  $PM_{2.5}$  concentrations was enhanced in northwest China, particularly when we used C-S model. This is related absorption-modulated natural emissions of -windblown dust.

330 Figure 6a,6b illustrates the cross sections in the northeast direction of changes in BC absorption coefficient due to emission reductions, as Beijing and polluted cities in South Hebei are covered. Pronounced declines are concentrated below 2km, and the core-shell model estimates stronger reductions due to lensing effects (Figure 6c). Emission control measures also reshaped the light absorption enhancement factor of BC, as indicated in Figure 6f. Within the lower boundary layer,  $E_{ab}$  values were reduced with emission reductions during APEC. Light absorption of BC stabilizes boundary layer to accumulate  $PM_{2.5}$ , yet this effect is inhibited at lower emission levels during APEC. These relationships are reflected in Figure 6g and 6h with negative changes in  $PM_{2.5}$  near the ground.

335 The responses of  $O_3$  to reduced light absorption of BC during APEC are in the opposite direction (Gao et al., 2018c), compared to those for  $PM_{2.5}$ . Strong absorption of BC tends to enhance photolysis above the aerosol layer, but to reduce photolysis near the ground. Figure 7d, 7g illustrate the changes in  $O_3$ 1D and  $NO_2$  photolysis rates with emission reductions inferred from an external mixing assumption. With emission control implemented, photolysis rates near the ground are enhanced due to lower light absorption of BC, while the photolysis rates above the aerosol layer are reduced. Similar patterns but with larger values are found using the core-shell model (Figure 7e, 7h). The responses of  $O_3$  are generally in line with the responses of  $O_3$ 1D and  $NO_2$  photolysis rates (Figure 7a, 7b).

#### 4 Summary and Discussions

In this study, we used the online coupled WRF-Chem model to understand how emission control measures during the APEC event would affect the mixing state/light absorption of BC, and the implications for BC-PBL interactions.

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350 Multiple observations, including surface observations of meteorological variables,  $PM_{2.5}$ ,  $PM_{2.5}$  chemical composition, and  
AAOD were used to evaluate model performance. A series of numerical experiments were conducted to address three  
questions: (1) how did emission reductions affect the aging processes and light absorption of BC during APEC; (2) what  
were the relative contributions of reduced mass concentrations of BC, aging processes of BC, and reshaped mixing state of  
BC to the changes in light absorption of BC during APEC; and (3) how did these processes affect BC-PBL interactions and  
355 formation of air pollution?

We found that both the mass concentration of BC and the BC coating materials declined during the APEC week,  
which reduced the light absorption and light absorption enhancement ( $E_{ab}$ ) of BC. Below 500 m in the troposphere, emission  
reductions during APEC lowered the absorption enhancement factor  $E_{ab}$  from 2.11 to 2.06. The column absorption  
enhancement was reduced also. The reduced AAOD during APEC is caused by both the declines in mass concentration of  
360 BC itself (52.0%) and the lensing effect of BC (48.0%). The reductions in coating materials (39.4%) dominated the influence  
of lensing effect, and the reduced light absorption capability ( $E_{ab}$ ) contributed 3.2% to the total reductions in AAOD. Our  
estimate of the contribution of reduced light absorption capability ( $E_{ab}$ ) exhibit lower values than *Zhang et al. (2016)*, which  
could be caused by the uncertainties in the assumption of the mixing state of BC in the core-shell model.

The diminished light absorption of BC during APEC promotes the development of PBL, as indicated in the changes  
365 in vertical profiles of EPT. Different responses of  $PM_{2.5}$  and  $O_3$  were found to the changes in light absorption of BC. The  
responses of  $PM_{2.5}$  follow the enhanced PBLH to decrease, while  $O_3$  concentrations increase near the ground. The enhanced  
levels of  $O_3$  were mainly caused by the influences of BC absorption on photolysis rates. As displayed in the conceptual  
scheme plot in Figure 8, reduced emissions of BC and its coating materials during APEC led to declined AAOD, less  
stabilized PBLH, decreased near surface  $PM_{2.5}$  concentrations and enhanced near surface  $O_3$  concentrations in the North  
370 China Plain.

This study with perturbations of emissions during APEC offer important implications on the potential effects of  
China's Clean Air Act. As discussed in our previous investigation (*Gao et al., 2017*), emission control measures have the co-  
benefits of reducing aerosol feedbacks to accelerate the cleaning of air, which accounts for ~11% of the decreased  $PM_{2.5}$   
concentrations during APEC. In this study, we further clarified that the ongoing measures to control  $SO_2$ ,  $NO_x$ , etc. would be  
375 efficient to reduce the absorption capability of BC to inhibit the feedback of BC on the boundary layer. Our results also show  
that near ground  $O_3$  responds differently from the changes in  $PM_{2.5}$ , which might be a side effect of current emission control  
strategies. *Ma et al. (2021)* reported that aerosol radiative effect could explain 23% of the total change in surface  
summertime  $O_3$  in China. How to control emissions to offset this side effect of current emission control measures on  $O_3$   
should be an area of further focus. In addition to the influences on air quality and weather, a sudden reduction in aerosol  
380 emissions may potentially affect climate (*Ren et al., 2020; Yang et al., 2020*), which warrants further investigation.

Although careful validation was conducted in this study, uncertainties still remain in the current study. We  
concluded that the core-shell model captures the variation of AAOD better than external mixing assumption. However, the  
core-shell model is an ideal scenario that assumes all non-BC materials are internally mixed and coated on BC (*Miyakawa et*

*al., 2014*). Zhang *et al.* (2016) observed that BC particles are heavily coated and are in a near-spherical shape in the North  
385 China Plain. The usage of core-shell model seems to be reasonable in this study, whereas the assumption that all non-BC  
materials are coated on BC might not be true in real atmosphere. The observed ratio of coatings to PM<sub>1</sub> was ~25-70% in  
summer in Beijing (Xu *et al.*, 2019), and the observed ratio of coatings to PM<sub>2.5</sub> was ~10-40% in winter in Beijing (Wang  
*et al.*, 2019). ~~Thus, the assumption of BC coating in the model might have overestimated  $r_{BC}$  in this study, leading to~~  
390 ~~uncertainties in the results. In the near future, we would examine how different assumptions of BC coating would affect the~~  
~~light absorption properties of BC. Additionally, the simulated feedbacks of BC absorption on boundary layer processes are~~  
~~not well constrained. We used multiple coupled models to examine how these processes are represented, and we calculated~~  
~~ensemble mean to obtain the best current understanding (Gao *et al.*, 2018a, 2020a). In the future, further efforts are needed~~  
~~also to constrain the uncertainties of these processes in the model. We conducted additional simulations assuming 10%,~~  
395 ~~20%, 40%, 60%, and 80% mixing fractions of coating aerosols, and explored how these assumptions would affect estimated~~  
BC AAOD, and its feedbacks on radiation, boundary layer and air pollutants. As suggested in Figure 9, modelled BC AAOD  
increases gradually when mixing fraction rises from 0.1 to 0.4, but it keeps relatively stable when mixing fraction is between  
0.4 and 1. The responses of near surface shortwave radiation and PBLH are in line with it, exhibiting relatively constant  
reductions when mixing fraction is higher than 0.4 (Figure 9). However, the no significant relationships are found for BC  
absorption induced changes in O<sub>3</sub> (<10% difference between fractions of 0.1 and 1) and PM<sub>2.5</sub> (<22% difference between  
400 fractions of 0.1 and 1) in Beijing. These results suggest that the findings demonstrated in this study might not be largely  
affected by the assumptions in mixing fractions of coating particles. Additionally, the simulated feedbacks of BC absorption  
on boundary layer processes are not well constrained. We used multiple coupled models to examine how these processes are  
represented, and we calculated ensemble mean to obtain the best current understanding (Gao *et al.*, 2018a, 2020a). In the  
future, further efforts are needed also to constrain the uncertainties of these processes in the model.

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Data availability. The measurements and model simulations data can be accessed through contacting the  
corresponding authors.

410 Author contributions. MG and JH designed the study. MG performed model simulations and analyzed the data with  
help from YY, HL, BZ, YZ, XL, CW, QZ and GRC. QZ provided the emission inventory. YW and ZL provided  
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**Table 1: Descriptions of model simulations**

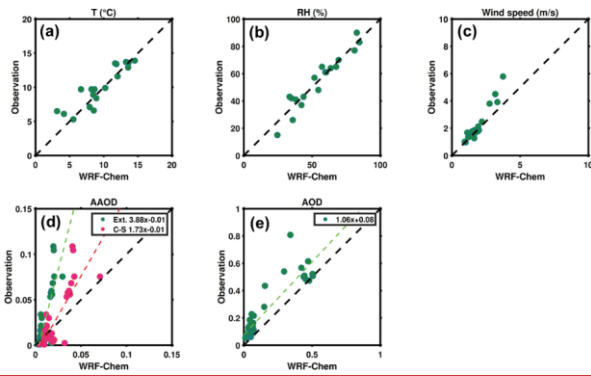
Experiments	Descriptions
<i>NOCTL<sub>Ext</sub></i>	No perturbations in emissions; assuming external mixing of BC.
<i>NOCTL<sub>Ext-nobc</sub></i>	No perturbations in emissions; assuming external mixing of BC; assuming no absorption of BC.
<i>NOCTL<sub>CS</sub></i>	No perturbations in emissions; calculating optical properties using core-shell assumption.
<i>NOCTL<sub>CS-nobc</sub></i>	No perturbations in emissions; calculation of optical properties using core-shell assumption; assuming no absorption of BC.
<i>CTL<sub>Ext</sub></i>	emissions are reduced during APEC; assuming external mixing of BC.
<i>CTL<sub>Ext-nobc</sub></i>	emissions are reduced during APEC; assuming external mixing of BC; assuming no absorption of BC.
<i>CTL<sub>CS</sub></i>	emissions are reduced during APEC; calculation of optical properties using core-shell assumption.
<i>CTL<sub>CS-nobc</sub></i>	emissions are reduced during APEC; calculation of optical properties using core-shell assumption; assuming no absorption of BC.
<i>CYSN<sub>CS</sub></i>	emissions of BC are reduced while emissions of other species are not during APEC; calculation of optical properties using core-shell assumption.
<i>CYSN<sub>CS-nobc</sub></i>	emissions of BC are reduced while emissions of other species are not during APEC; calculation of optical properties using core-shell assumption; assuming no absorption of BC.

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**Table 2: The division of the impact of BC absorption into the impact of BC mass itself and BC mixing state**

Influenced variables	BC mass itself	BC lensing effect	Reduced coating	Reduced $E_{ab}$
AAOD	52.0%	48.0%	39.4%	3.2%
PBLH	34.9%	65.1%	47.4%	-
PM <sub>2.5</sub>	64.3%	35.7%	-	-
O <sub>3</sub>	49.1%	50.9%	-	-

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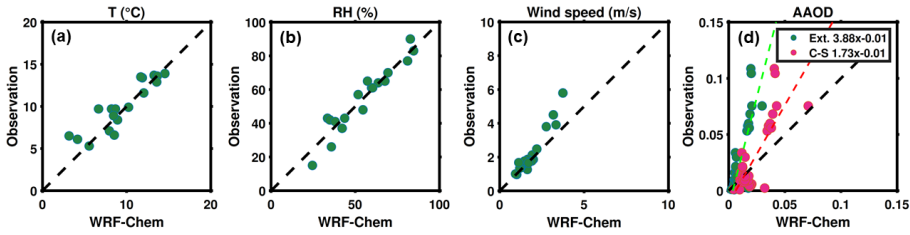


Figure 1: Scatter plots of modeled and observed near surface meteorological variables (a: T, b: RH, c: wind speed); modelled AAOD with core-shell model/external mixing assumption, and the comparisons against observations (d); modelled and observed AOD (e).

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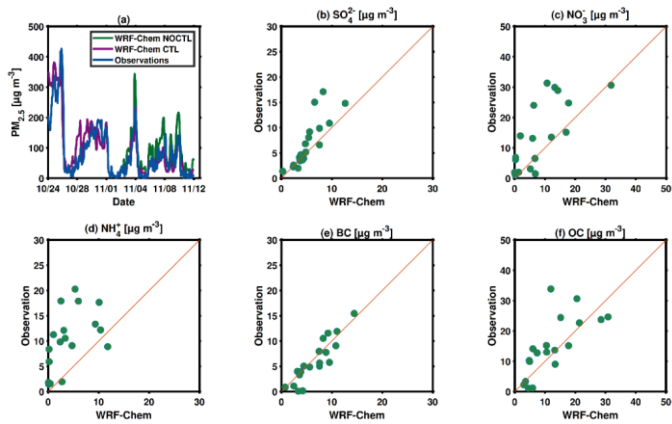


Figure 2: Modeled and observed time series of  $PM_{2.5}$  concentrations in urban Beijing (a); Scatter plots of modeled and observed near surface daily mean concentrations of sulfate, nitrate, ammonium, BC (black carbon) and OC (organics) in Beijing (b-f).

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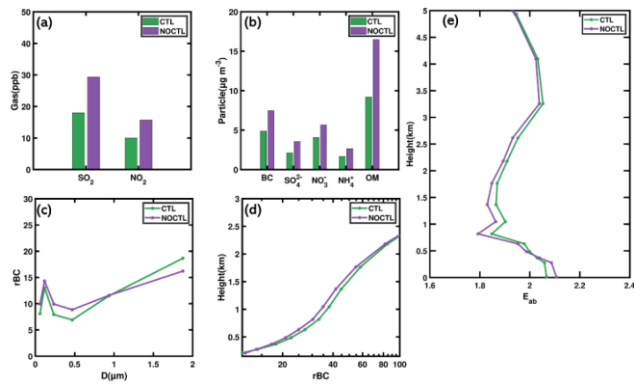


Figure 3: Mass concentrations of gaseous and condensed pollutants in the CTL and NOCTL cases (a-b); the distribution of  $rBC$  with sizes in the CTL and NOCTL cases (c); the distribution of  $rBC$  with height in the CTL and NOCTL cases (d) and the distribution of  $E_{ab}$  values with height in the CTL and NOCTL cases (e).

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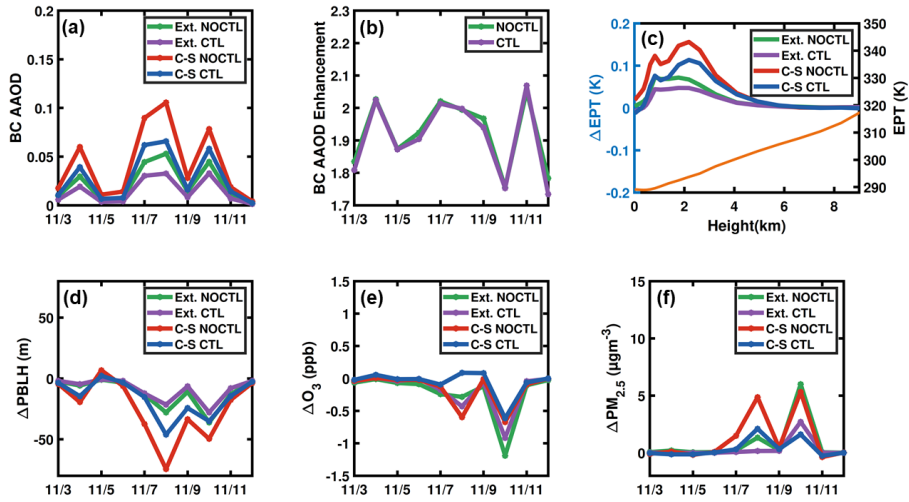
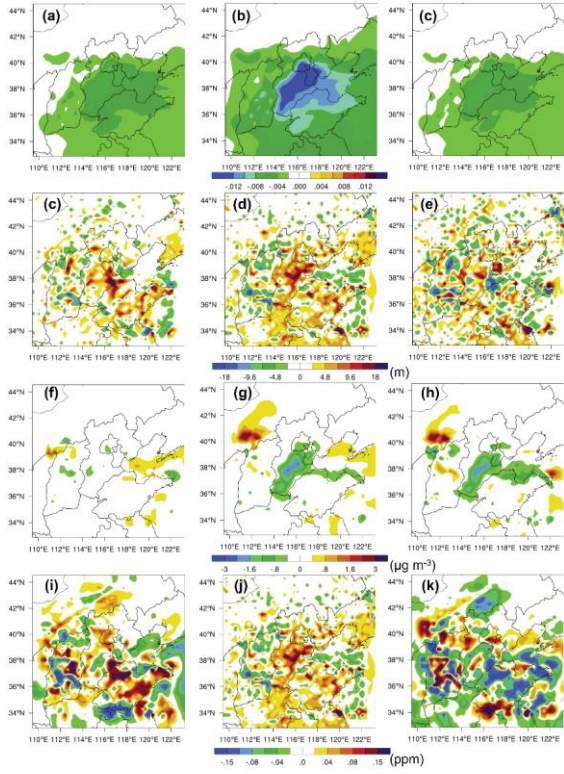
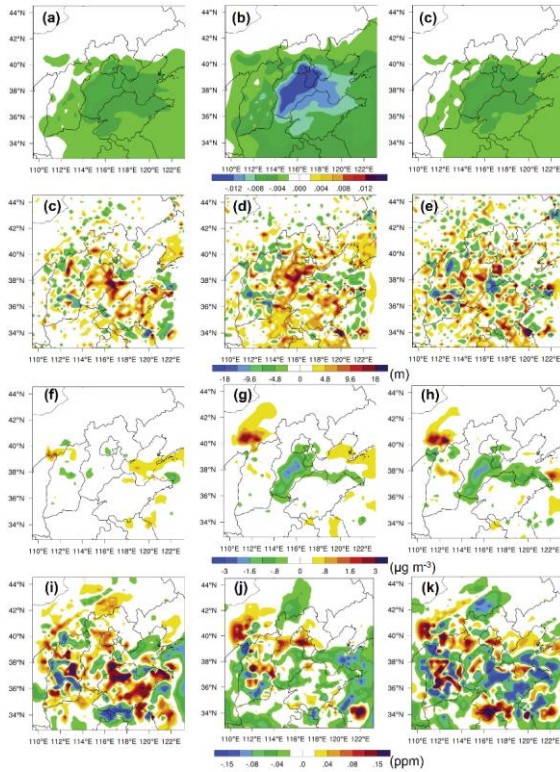


Figure 4: Daytime (10:00-17:00 local time) mean BC AAOD in Beijing inferred from different simulations (a) and the BC AAOD enhancement in the NOCTL and CTL scenarios (b); BC absorption induced changes in EPT (orange line indicates the vertical profile of EPT) (c), PBLH (d),  $O_3$  (e), and  $PM_{2.5}$  (f).

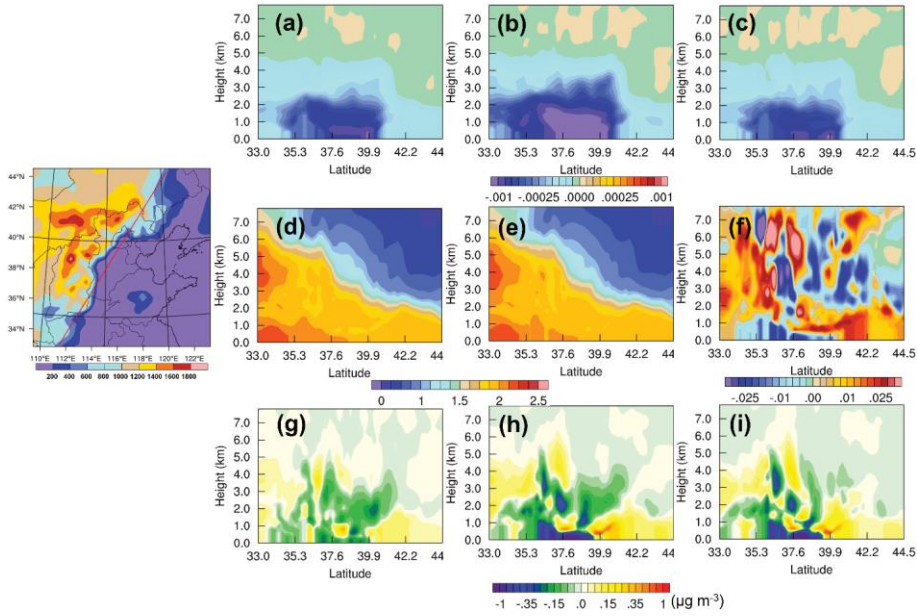
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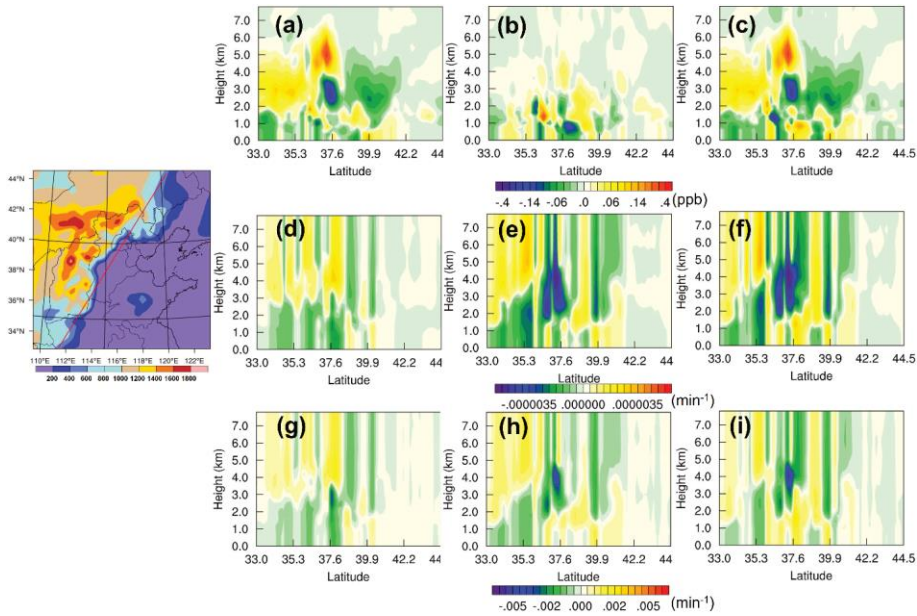




655 **Figure 5: Spatial distribution of daytime (10:00-17:00 local time) mean BC AAOD (first row), and mean BC absorption induced changes after emission reductions (CTL minus NOCTL) in PBLH (second row), PM<sub>2.5</sub> (third row), and O<sub>3</sub> (fourth row); first, second and third columns represent  $\Delta_{emission-Ext}$ ,  $\Delta_{emission-CS}$ , and  $\Delta_{aging}$ .**

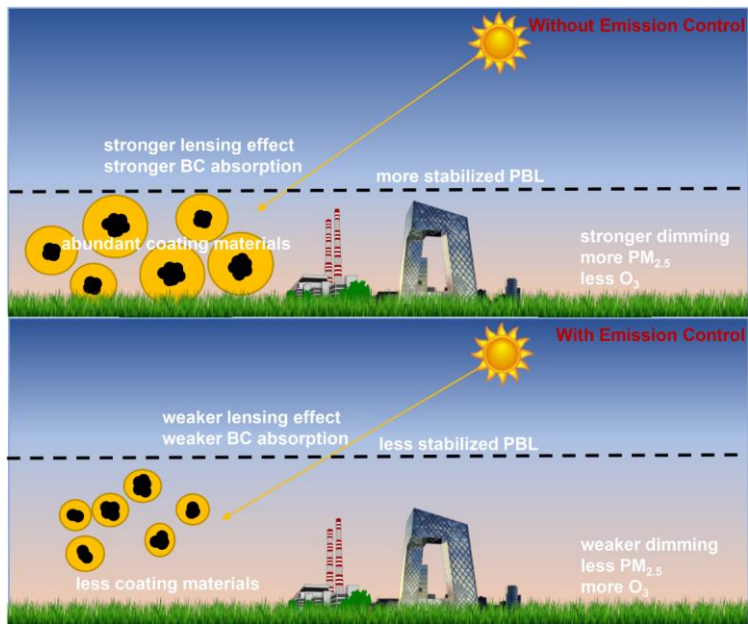


660 **Figure 6:** Cross sections of daytime (10:00-17:00 local time) mean changes in BC absorption coefficient (first row, CTL minus NOCTL),  $E_{ab}$  (second row), BC absorption induced changes after emission reductions (CTL minus NOCTL) in  $PM_{2.5}$  (third row); first, second and third columns represent  $\Delta_{emission-Ext}$ ,  $\Delta_{emission-CS}$ , and  $\Delta_{aging}$ .

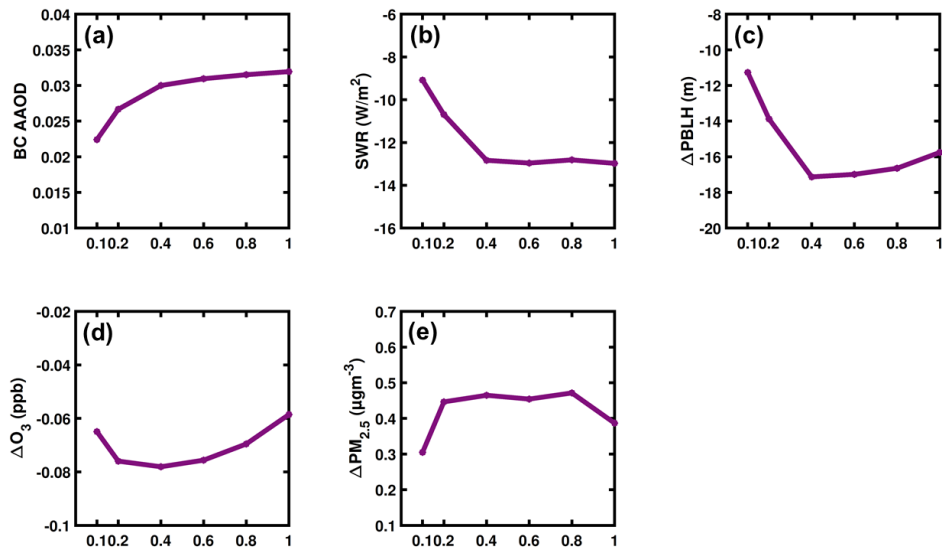


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**Figure 7: Cross sections of daytime (10:00-17:00 local time) mean changes in BC absorption induced changes after emission reductions (CTL minus NOCTL) in O<sub>3</sub> (first row), O<sub>3</sub>D photolysis rate (second row), and NO<sub>2</sub> photolysis rate (third row); first, second and third columns represent  $\Delta_{emission-Ext}$ ,  $\Delta_{emission-CS}$ , and  $\Delta_{aging}$ .**



670 **Figure 8: Conceptual scheme of the effects of emission control during APEC on light absorption capability of BC, PBL and air quality.**



675 **Figure 9: Daytime (10:00-17:00 local time) mean BC AAOD in Beijing (a) BC absorption induced changes in shortwave radiation (b), PBLH (c), O<sub>3</sub> (d), and PM<sub>2.5</sub> (e) as a function of fraction of non-BC particles mixed with BC.**

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