

Response to Referee #1

General Comments: The authors used WRF-Chem single-column model combining with available in-situ observation to quantify the impacts of some key factors on BC's dome effect. They demonstrated that the dome effect of BC aerosols strongly depends on the vertical distribution and aging process of BC, as well as the underlying land surface. The technical work appears to be competent. The paper contains important addition to existing literature, and is suited for publication to ACP. However, there are several concerns that should be addressed or considered before being accepted for publication.

Response: We would like to appreciate the referee for providing the suggestions. We have revised this article according to the comments.

Specific Comments: 1. Model configuration: In terms of mixing state of multiple aerosol chemical compositions, the authors presume that the aerosols in each bin were internally mixed, which would overestimate the light-absorbing capability of BC aerosols. In the atmosphere, a large amount of aerosol mass was partitioned onto BC-free particles rather than internally mixing with BC.

Response: Thanks for raising this important point. Yes, in this work aerosol components in each size bin are assumed to be internally mixed, which means that BC is uniformly distributed throughout the particle and its light-absorbing efficiency could be amplified by other scattering aerosols. Many existing in-situ measurements suggested that the majority of carbonaceous particles were internally mixed, especially during hazy days, and were highly influenced by secondary species in eastern China (Li et al., 2011; Wang et al., 2014b; Zhang et al., 2013). Single-particle soot photometer measurements revealed that the fraction of internally mixed BC was variable and could be as high as around 70% in the Pearl River Delta region and approximately 50% in Beijing area (Wang et al., 2014b; Huang et al., 2012). Accordingly, BC absorption enhancement with nitrate and sulfate is also frequently observed in China (Chen et al., 2017). Indeed, some studies reported that internally-mixing assumption would overestimate the light-absorbing capability of BC (Cappa et al., 2012; Bond et al., 2013), while others found that the assumption is “relatively reliable for modelling” and can promote the underestimation of modelled MAC_{BC} (Adachi and Buseck, 2008; Koch et al., 2009). Moreover, the simulations of aerosol optical properties and radiative effect conducted over China perform well with internal mixing assumption (Huang et al., 2015; Zhang et al., 2015; Wang et al., 2014a). Thus the results can be seen as an upper limit and is acceptable with the uncertainty of ~20%. Further studies on the representation of the

evolution of mixing state of BC is still needed when more field measurements data in different regions are available.

2. P5/L17: *When the abbreviation regarding “SNA” first appeared in the paper, the author should also give its full terms.*

Response: Accepted. We will give the full terms when “SNA” first appears in the revised paper .

3. *Why the author only considered BC mixing with SNA? Why not consider other aerosol compositions, especially organic species that play an important role in the mass fraction of aerosol particles.*

Response: Thanks for pointing out this issue. BC was also mixed with organic carbon (OC). There are multiple reasons for excluding OC in our current work. First, the optical properties of organic species such as refractive index and their mixing state with other aerosol components have not yet been fully explored and large uncertainties do exist (Kanakidou et al., 2004). Secondly, scattering aerosols like sulfate and nitrate magnify the light-absorbing capacity of BC not only through their own lensing effect but also through further increase in mass absorption cross section due to hygroscopic growth. Considering the uncertainties in OC’s optical properties and less notable hygroscopicity, we do not include it in this article. More quantitatively, we conducted some sensitivity simulation by assuming OC as purely scattering aerosol (default refractive index given in WRF-Chem). As shown in Fig. R1-2, when the inputted organic aerosol surface concentration exceeds $40 \mu\text{g m}^{-3}$, the changes in heating rate ($\sim 0.02 \text{ K h}^{-1}$) and temperature (less than $0.2 \text{ }^\circ\text{C}$) are not much different from those in Figure 1-2. However, we do thank the referee for raising this point. We will add a few sentences to explain the reason of excluding OC in our revised manuscript.

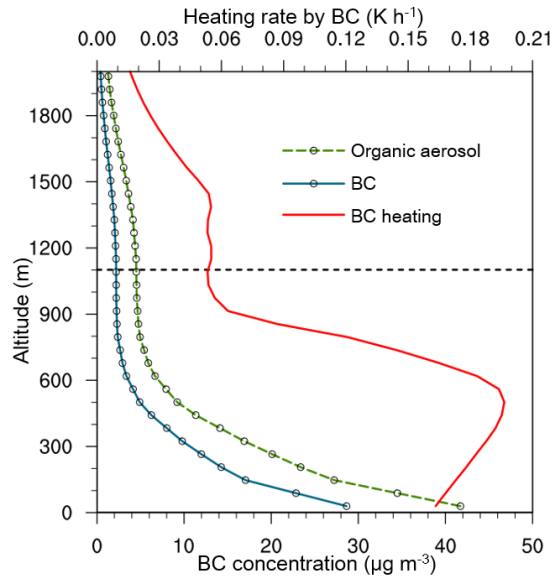


Fig. R1 Vertical profile of BC and OC along with shortwave heating rate induced by BC in the afternoon (12:00-16:00). Black dash line denotes the averaged PBL height during this period.

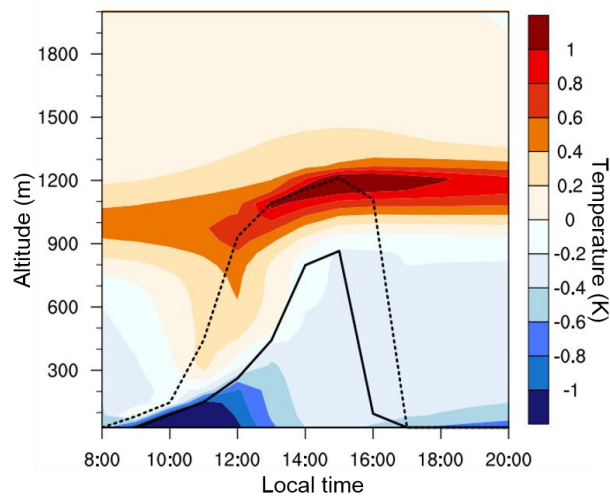


Fig. R2 Diurnal variation of the air temperature change caused by aerosols and of PBL height for runs with (solid line) and without (dash line) ARI. Other settings are the same as Figure 2 except for additional OC input.

4. Figure 1 shows the heating efficiency of BC due to shortwave radiation absorption peaked around 600-800 m. But the strongest upper-air warming exhibited at 1000-1200 m shown in Fig. 2, different with the height of the largest heating efficiency of BC. The author need to explain the reason.

Response: Although shortwave heating peaked around 600-800 m, due to turbulence

mixing and convective motion, the heated air will not stay where they are. Instead, they will rise because of small density until reaching the warmer capping inversion.

5. *According to the vertical profile of shortwave heating rate included by unit BC mass (Fig. 1), the authors discussed that BC in the upper PBL is more efficient in terms of absorbing shortwave radiation and heating surrounding air masses. To support the author's statement on the upper PBL, Figure 1 need to show the average PBL height in the afternoon (12:00-16:00 LT) during a heavy polluted episode 23rd-24th December, 2013 in Beijing.*

Response: Accepted. We re-plotted Figure 1 by adding the daytime averaged (12:00-16:00LT) PBL height during this episode.

6. *P6/L25-27: The authors state here that Figure 3a displayed the information of the incident solar radiation at the surface, decline in surface temperature and surface sensible heat flux. I did not found these information in Fig. 3a. The authors should check.*

Response: Accepted. Fig. 3a should be the whole Fig. 3, which gives all information about temperature (Fig. 3b) and radiation (Fig. 3c). Sensible heat flux is directly calculated from model output and is not shown in the figure.

7. *In Fig. 3b, what does the black dash line represent? Authors should state it in the caption of the figure 3.*

Response: Accepted. The black dash line represents the diurnal averaged PBL height, and the statement will be added to the revised manuscript.

8. *P7/L4: According to the data shown in Fig.4, the increment of increase BC concentration from 0 to 30 $\mu\text{g m}^{-3}$ should be 5 $\mu\text{g m}^{-3}$ rather than 2 $\mu\text{g m}^{-3}$.*

Response: Accepted.

9. *P8/L10-15: The calculation method of absorption coefficient amplification factors of BC was different with that in the literature. Authors calculated the absorption amplification of BC aerosols based on the BC/SNA ratio during different periods (i.e., clean period: BC/SNA=1:3, haze episode: BC/SNA=1:8). The obtained absorption amplification (i.e, 1.8, 1.7, 1.6 and 1.4 at wavelength 300, 400, 600 and 1000 nm) was then compared with previous laboratory and in-situ observational studies. However, the absorption amplification of BC aerosols in previous studies represents that the*

enhancement of light absorption of BC-containing particles due to coating materials on BC surface comparing with bare BC.

Response: Accepted. The calculation of absorption amplification in this study indeed represents the enhancement of BC absorptive properties due to different BC/SNA ratio, or different pollution periods, and should be compared with changes of BC mass absorption cross-section at some specific wavelength. A two-week intensive measurements in the urban area in Northern China winter with both SNA and BC concentrations as well as light absorption coefficient observed are employed here to compare with the simulations (Chen et al., 2017). When BC/SNA ratio rises from 1:3 to 1:8, the light absorption coefficient observed increases by an averaged factor of ~ 1.5 , indicating that the absorption amplification obtained is reasonable. We will add this information in the revised manuscript.

10. In Fig.5, authors used BC/SNA ratio of 1:3 and 1:8 to calculate the aerosol absorption coefficient at clean period and haze episode, respectively. They should give the data sources of BC/SNA ratio at different periods. Moreover, Fig.5 also shows the solar spectral irradiance at sea level (bright yellow shadow). However, there was not any discussion on this information in the paper. Why did the authors give solar spectral irradiance at sea level in Fig. 5?

Response: The BC/SNA ratio we used in our simulation is extracted from in-situ measurements conducted in Beijing during 1-16 January 2013 (Sun et al., 2014). The detailed aerosol data of clean period and haze episode can be found in the Table 1 of this article. In shortwave radiation scheme, only 4 typical bands (300, 400, 600, 1000nm) are calculated. In order to investigate the relative importance of amplification of absorption extinction coefficient for each band, we gave solar spectral irradiance at sea level as a reference, which help the reader to understand our conclusion that the amplification of 400-600 nm is quite important because of more intensive solar irradiance. This discussion will be added to the revised manuscript.

11. Considering most of days in north China characteristic of RH lower than 50%, I suggest adding a case of RH=30% in Fig.6. Moreover, authors gave the absorption extinction coefficient at a certain wavelength (i.e., 400 nm), they should state it in the caption of the figure 6.

Response: Thanks for the suggestions. In fact, we have conducted the simulation in the case of RH=20%, 30% and 40%. However, the results are the same as RH=50% due to the RH threshold for aerosols' deliquescence. If ambient RH is lower than this threshold, also known as the deliquescence relative humidity (DRH), aerosols will not absorb water and remain solid thus no

change of optical properties occurs. Here we present the result with RH equal to 30% for your reference (Fig. R3). Moreover, the description about the wavelength will be added to the caption in revised manuscript.

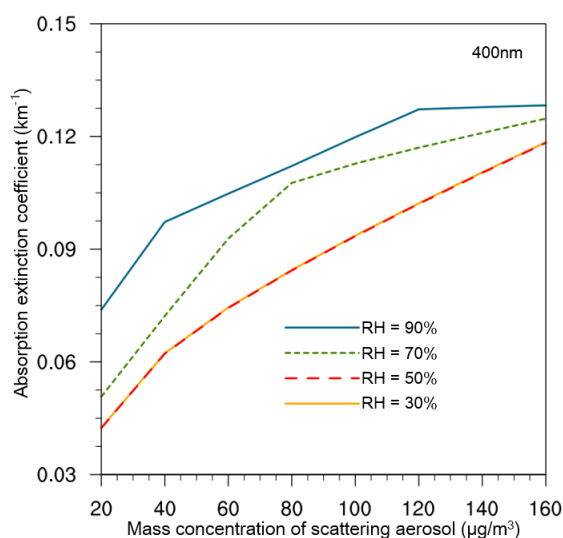


Fig. R3 Same as Figure 6 but with RH varying from 30% to 90%

12. *RH at a certain SNA influence not only aerosol particle diameter but also their reflective index (RI). Authors mentioned the change of aerosol particle diameter at different levels of RH (P8/L17-18). Did they also consider the RI change? The authors should give detail description on data process at different levels of RH in Sect. 2 (data and method), such as the diameter growth factors and RI setting in the model calculation.*

Response: Yes, RH absolutely modifies not only aerosol diameter but also the RI value. We do include the impact of RH on aerosol RI. Actually, in the WRF-Chem model, the RI value is determined by 11 aerosol species, including aerosol water which is highly dependent on RH (Barnard et al., 2010; Fast et al., 2006). The calculation of extinction coefficient, single-scattering albedo and asymmetry factor are based on a sectional approach. In each bin, the particles are assumed to be spherical and internally mixed. Chemical masses (converted to volumes after) and particle number are required to find physical diameter of each bin. Volume averaged mixing rule are employed to calculate the bulk refractive index of all the particles (including water aerosol) in each bin. The detailed information can be found in Barnard et al. 2010. Therefore, the influence of RH and hygroscopic growth on SNA optical properties technically are already considered in this study. We will add these details in Section 2 in the revised manuscript.

13. *When investigating the impacts of BC aging process on light absorption, authors discussed the difference of absorption under difference BC/SNA ratio, shown in Fig.5*

and Fig.6. However, why they fixed a value of BC/SNA ratio (i.e., 1:6 shown in Fig. 7) rather than using different BC/SNA ratio as above two figures to further discuss the impacts of BC aging process on PBL height? Figure 5 shows different BC/SNA ratio under different pollution levels (i.e., clean period: BC/SNA=1:3, haze episode: BC/SNA=1:8) based on in-situ observation from literature, indicating that BC/SNA ratio will change with BC concentrations. Therefore, it seems to be unreasonable to assume same BC/SNA ratio under different BC concentrations.

Response: Accepted. There is no need to use a fixed value of BC/SNA ratio since it varies a lot in actual haze episode. Here, according to the comment, we conducted another set of parallel experiments with BC concentration fixed at $5 \mu\text{g m}^{-3}$ and SNA concentration ranges from 20 to $160 \mu\text{g m}^{-3}$, which shares the same chemical components with Figure 6. The PBL response of increasing by lower-level BC and decreasing by upper-level BC are both magnified. As the pollution gets severe which characterized by increasing level of scattering aerosols, the amplification of BC induced PBL height changes also increases, as shown in the following figure. We will revise Figure 7 and the relevant discussions.

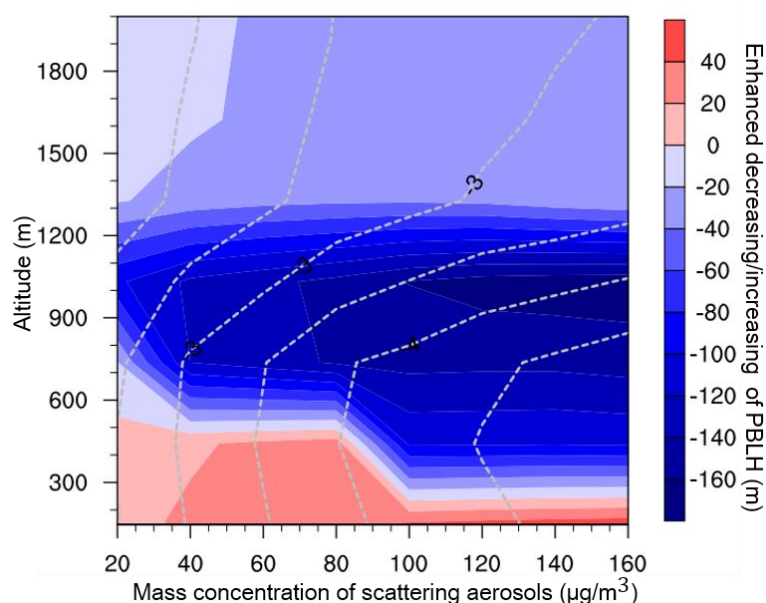


Fig .R4 Enhanced decreasing/increasing of PBL height (contour map) and enhanced reduction of sensible heat flux at surface (dash isoline) caused by amplified absorption of BC internally mixed with scattering aerosols.

14. When BC concentration is lower than $2 \mu\text{g m}^{-3}$, the PBL height variations due to specific BC distribution shown in Fig. 9 exhibited significant difference with that shown in Fig.4. Why?

Response: After looking through the modelling results, it turns out that the difference is attributed to different criteria of comparison. In Fig. 4, the positive

variations of PBL height is compared to the scenario without aerosol radiation interaction (ARI), while in Fig. 9 the negative variations of the first two layers result from the comparison to the scenario with ARI but no manually inputted aerosols. Therefore, the difference can be viewed as the effect of back ground aerosols. For convenience, the comparison standard will be unified to the difference between the scenarios with/without ARI in the revised version.

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