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Key Points:

- BC plays an important role in enhancing surface haze pollution in megacities in China
- Upper PBL heating and surface cooling by BC are two comparable processes in the PBL feedback
- Reducing BC emission cobenefits the mitigations of haze pollution and global warming

Supporting Information:

- Supporting Information S1

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Enhanced haze pollution by black carbon in megacities in China

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Abstract Aerosol-planetary boundary layer (PBL) interactions have been found to enhance air pollution in megacities in China. We show that black carbon (BC) aerosols play the key role in modifying the PBL meteorology and hence enhancing the haze pollution. With model simulations and data analysis from various field observations in December 2013, we demonstrate that BC induces heating in the PBL, particularly in the upper PBL, and the resulting decreased surface heat flux substantially depresses the development of PBL and consequently enhances the occurrences of extreme haze pollution episodes. We define this process as the "dome effect" of BC and suggest an urgent need for reducing BC emissions as an efficient way to mitigate the extreme haze pollution in megacities of China.

1. Introduction

Severe and persistent haze pollution has frequently occurred in China in recent years, especially in the megacities of eastern China [*Parrish and Zhu*, 2009; *Ding et al.*, 2013; *Huang et al.*, 2014; *Zhang et al.*, 2015]. Besides high emission rates and fast formation of secondary aerosols [*Huang et al.*, 2014; *Guo et al.*, 2014], the main reasons for the formation of these extremely high episodes were generally attributed to unfavorable meteorological conditions [*Ding et al.*, 2013; *Z. Wang et al.*, 2014; *Zhang et al.*, 2016]. However, current operational air quality models generally underestimate the extreme peak mass concentration of particulate matter (PM) [*Z. Wang et al.*, 2014; *J. Wang et al.*, 2014], making it difficult for the government to take quick actions or measures to tackle such extreme pollution episodes or even to issue appropriate early warnings in the case that dangerous concentrations are reached. Recent studies have found that "online" coupled models, by considering the aerosol-planetary boundary layer (PBL) feedback, could improve the forecast capabilities of such episodes [*J. Wang et al.*, 2014; *Zheng et al.*, 2015; *Wang et al.*, 2015], while quantitative understandings about the roles of key aerosol components and dominant processes in this feedback are still lacking.

Black carbon (BC), also referred to as soot, is an important particulate pollutant because of its adverse health impacts [*Bond et al.*, 2013]. Compared with the total mass of fine particles, BC only contributes a fraction of about 5–15% to the total aerosol mass concentrations in urban air [*Yang et al.*, 2011]. However, from the perspective of climate change, BC together with brown carbon (BrC) is one of the most radiatively important aerosol components in the atmosphere [*Jacobson*, 2001; *Huang et al.*, 2011; *Bond et al.*, 2013]. BC has been found to play an important role in regional and even global climate [*Menon et al.*, 2002; *Ramanathan and Carmichael*, 2008] and also extreme weather [*Fan et al.*, 2015; *Saide et al.*, 2015]. Because of the relatively short lifetime of BC in the atmosphere, reduction of BC emissions has been known to be a viable way to reduce global warming [*Jacobson*, 2001; *Bond* et *al.*, 2013].

China has been reported to have a rather high emission rate of BC [e.g., *Qin and Xie*, 2012] and consequently high BC concentration during severe haze events [*Andersson et al.*, 2015]. Studies showed that BC plays a key role in influencing regional climate and even extreme weather in China [*Yu et al.*, 2001; *Qian et al.*, 2003; *Ding et al.*, 2013; *Fan et al.*, 2015]. However, so far, there is no control measure specified for BC emissions in China,

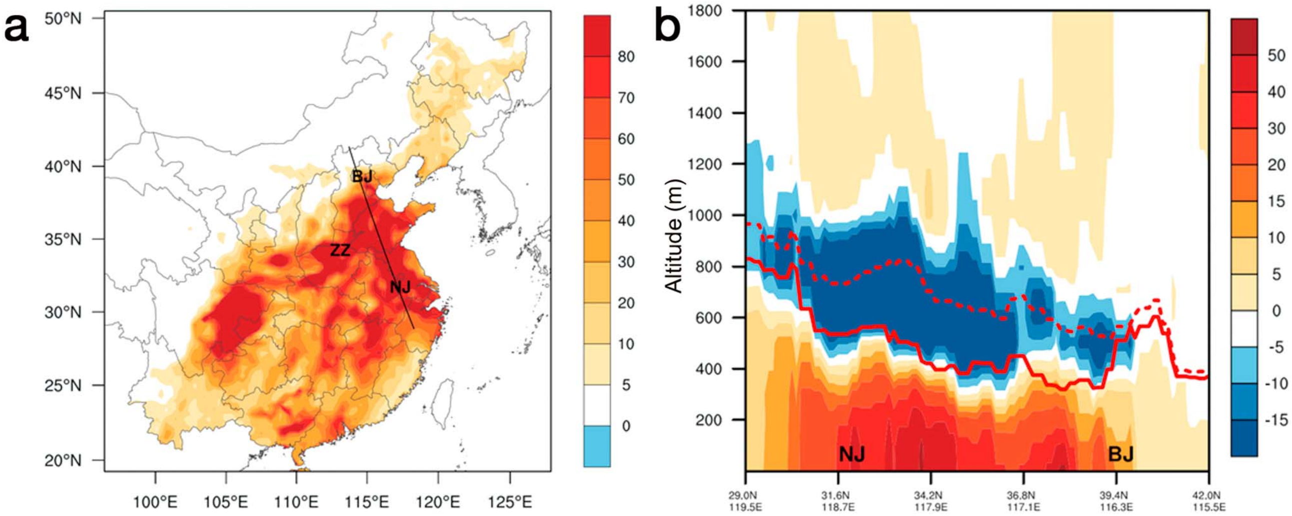

Figure 1. Influences of aerosol-PBL interactions on the horizontal distribution and vertical structure of $PM_{2.5}$ concentration and PBLH. (a) Maximum difference in the afternoon (12:00–16:00 LT) $PM_{2.5}$ mass concentration between the experiments EXP_WF and EXP_WoF. (b) Cross section of the difference in the $PM_{2.5}$ mass concentration between the experiments EXP_WF and EXP_WoF at 14:00 LT for eight haze episode days along Beijing (BJ) and Nanjing (NJ) axes. The red solid and red dashed lines are the averaged PBLH for the experiments EXP_WF and EXP_WoF, respectively. The unit for concentrations in both figures is $\mu g\, m^{-3}$.

probably because more efforts have been mainly targeted on carbon dioxide and total mass concentration of fine particle, respectively, from perspectives of climate and environment policies. In this study, we demonstrate that BC plays an important role in enhancing wintertime haze pollution in megacities in China via its interactions with PBL meteorology based on modeling simulations and data analysis of various field observations for eastern China.

2. Results

In December 2013, almost two thirds of China's territory experienced severe haze pollution (with $PM_{2.5}$ greater than the Grade II Air Quality National Standard in China, i.e., 75 $\mu g\, m^{-3}$ for daily average), especially in the megacities of eastern China (Figure S1a in the supporting information). To understand the impact of BC on the haze pollution via aerosol-PBL interactions, we conducted numerical simulations using the Weather Research and Forecasting (WRF) model coupled with Chemistry (WRF-Chem), an online-coupled three-dimension Eulerian chemical transport model considering complex physical and chemical processes [*Grell et al.*, 2005]. We run the model with three parallel experiments: (1) without any aerosol feedback (EXP_WoF), (2) with aerosol feedback considering total (direct and indirect) radiative effects from all chemical constituents (EXP_WF), and (3) with the aerosol feedback except BC (EXP_WFexBC). The detailed model configurations are given in the methodology section and Table S1 in the supporting information. We evaluated the model performance for the three experiments by comparing the simulation results with surface observed $PM_{2.5}$ and BC concentrations, upwelling and downward shortwave radiation, latent and sensible heat fluxes, radiosonde and aircraft-measured meteorological parameters, and chemical components.

The simulated monthly averaged $PM_{2.5}$ concentration in China by the run EXP_WF (Figure S1b) well reproduced the spatial variations of $PM_{2.5}$ in the eastern China (Figure S1a). Evaluation with various measurements demonstrates that the EXP_WF run with non-BC and BC aerosols had the best performance in capturing the temporal variations of vertical structure of BC, absorption aerosol optical depth, solar radiation, and surface energy budget in the main megacities in eastern China during the studied period (Figures S2–S4 and Table S2). On the other hand, without aerosol feedback, the model (EXP_WoF run) significantly underestimated $PM_{2.5}$ concentration by up to 100 $\mu g\, m^{-3}$ in many regions, such as eastern China and the Sichuan Basin (Figure 1a), accompanied with overestimations of PBL height (PBLH) and $PM_{2.5}$ concentration around the top of PBL (Figure 1b).

To explore the role of BC in the aerosol-PBL feedback, we selected haze episode days (with the maximum hourly $PM_{2.5}$ concentration exceeding 200 $\mu g\, m^{-3}$) at three megacities, Beijing (BJ) in the north, Nanjing

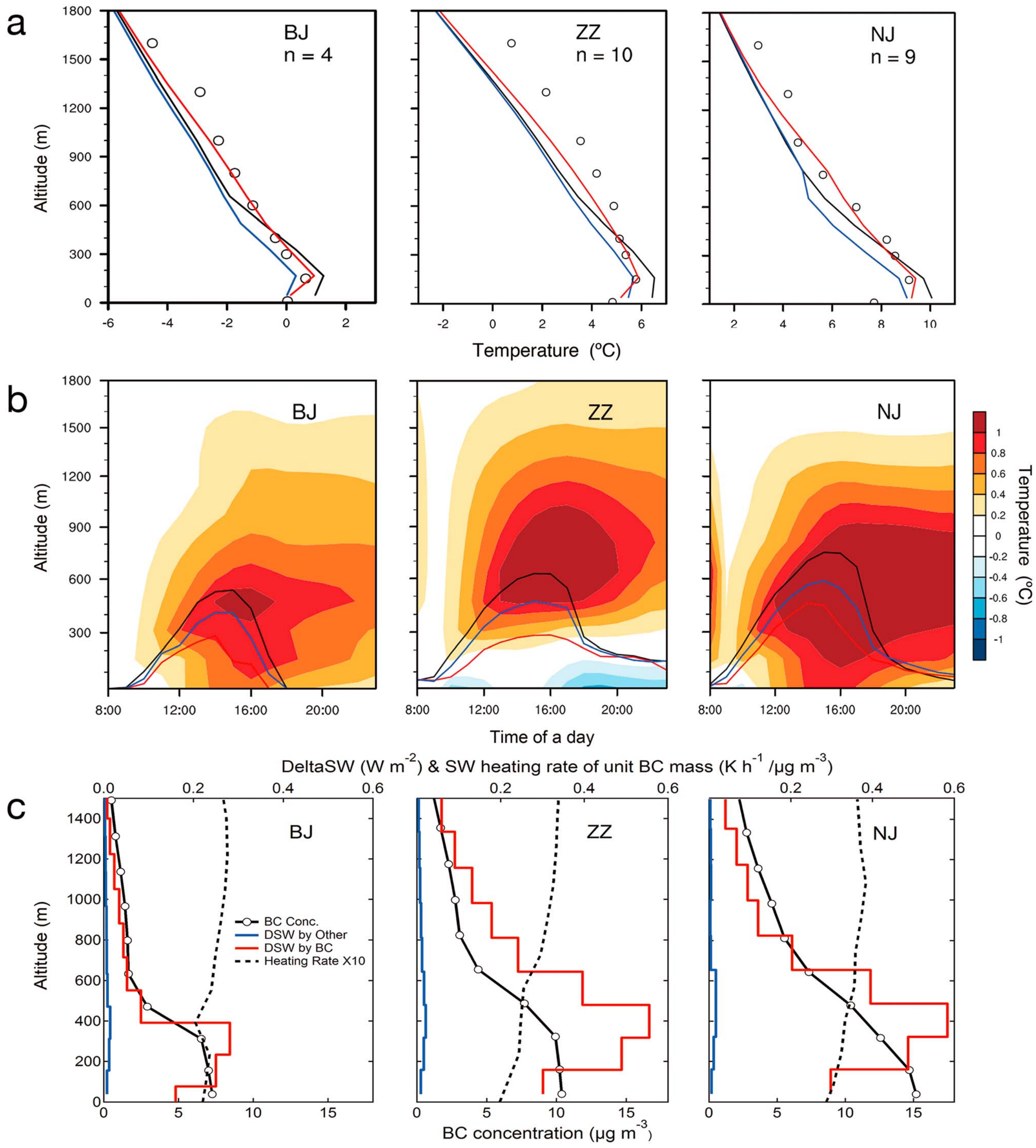

Figure 2. (a) A comparison between the observed and modeled air temperature profiles for the three WRF-Chem experiments at 20:00 LT during the haze episode days (*n* means number of haze days). Black circles denote sounding observations. Black, blue, and red solid lines are for EXP_WoF, EXP_WFexBC, and EXP_WF, respectively. (b) Modeled diurnal variations of the air temperature change due to BC during the haze days and of PBLH for the three runs (solid black, blue, and red lines). (c) Vertical distributions of the averaged BC concentrations, atmospheric attenuation of incident shortwave radiation induced by BC (red) and other aerosols (blue), and shortwave heating rate induced by unit mass of BC (black dashed line) for the haze days.

(NJ) in the south, and Zhengzhou in the west of the North China Plain (Figure S1b), and compared the simulated vertical profiles of air temperature with sounding observations, diurnal variations of air temperature changes, and vertical heating induced by BC in Figure 2. The EXP_WF run, i.e., simulations with the full aerosol feedback, had the best performance in reproducing the observed vertical structure of air temperature, especially that between the altitudes of 600 and 1200 m. For the EXP_WFexBC, a substantial underestimation (1 ~ 1.5°C) was apparent between 300 and 1200 m compared with EXP_WF, indicating an important role of BC in changing the air temperature in the upper PBL. The averaged vertical profiles of atmospheric attenuation of solar radiation, defined as the gradual loss in intensity of solar radiation in the atmosphere [*Idso*, 1969], suggest that although BC generally reaches its maximum concentration near the ground surface, the largest change in shortwave radiation takes place in the upper PBL because of more efficient shortwave absorption by unit mass of BC than that in the lower PBL (Figure 2c). These results are also consistent with previous modeling and observation studies [*Samset and Myhre*, 2011; *Cappa et al.*, 2012; *Ferrero et al.*, 2014; *Samset et al.*, 2014]. As a consequence, the daytime continuous heating resulted in a maximum air temperature change during the late afternoon (around 16:00, Figure 2b), which favored the formation of an inversion layer in late afternoon and at night.

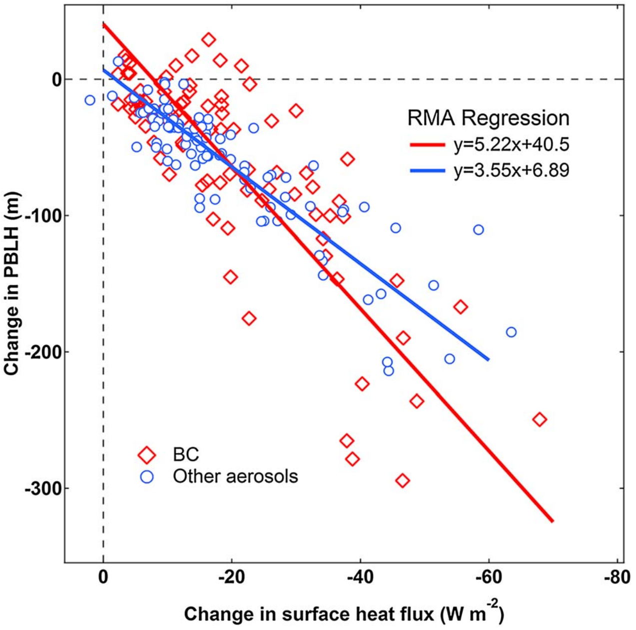

Figure 3. Scatterplot of PBLH change as a function of change in surface heat flux (the sum of sensible heat and latent heat) induced by BC and other aerosols in the Beijing-Tianjin-Hebei, Henan-Shandong, and Yangtze River Delta regions. The influence of BC was calculated as the difference between EXP_WF and EXP_WFexBC and influence of other aerosols as the difference between EXP_WFexBC and EXP_WoF. The red and blue solid lines give the reduced major axis (RMA) regression for the two data sets. The RMA regression software was provided by Andrew J. Bohonak at San Diego State University.

The change in the vertical heating rate, together with that in the surface sensible and latent heat fluxes induced by PBL aerosols, caused opposite effects in the upper and lower PBL: a net heating effect in the upper PBL and cooling effect close to the surface. This change increased the atmospheric stratification and caused a substantial drop in the PBLH. Among the three cites, Zhengzhou showed the strongest PBL depression because of the highest BC column concentration (Figure 2b). The situation occurring on 24 December gave the best demonstration for the roles of BC's heating on the evolution of the vertical temperature profile in the upper PBL. Figure S5a shows that aerosol particles substantially increased the radiative heating in the atmosphere at Zhengzhou on that day, particularly in the upper PBL around 12:00 LT. The vertical temperature showed obvious responses, with a significant warming in the upper air but a notable cooling near the surface (Figure S5b). Distinct discrepancies exist between the atmospheric sounding measurements and the simulation results when the BC's role was neglected either in EXP_WoF or EXP_WFexBC (Figure S5c). Only the run with the BC's radiative heating effects, i.e., EXP_WF, could remarkably narrow the gaps toward the observation, especially the warming features in the upper part of the PBL.

The development of a convective PBL is mainly driven by thermal processes [*Yu et al.*, 2002; *Ding et al.*, 2013; *Sühring et al.*, 2014; *Petäjä et al.*, 2016]. Air pollution alters the growth of the PBL both by changing the surface buoyancy flux and by favoring a capping inversion in the upper PBL [*Sühring et al.*, 2014], which are influenced by either surface longwave radiation or absorption of solar radiation in the atmosphere [*Yu et al.*, 2002]. The radiative forcing analysis (Figures S6 and S7) suggests that BC, as the most radiatively efficient species, plays a key role in heating the atmosphere and cooling the surface, which is quantitatively consistent with previous works [e.g., *Ramanathan and Carmichael*, 2008]. Figure 3 shows the drop of PBLH as a function of the surface heat flux change induced by BC and non-BC aerosols. Given that the same change in the surface flux should contribute to an equal PBLH change if only considering the effect of surface buoyancy flux alone [*Yu et al.*, 2002; *Sühring et al.*, 2014], the different slopes for BC and non-BC aerosols here reveal that the stratification change induced by vertical heating of BC gave a significant contribution to the PBL decrease. Further 1-D single column modeling confirms that the upper PBL BC, even with a burden ratio lower than

30% of the total column, could cause a drop of the daytime PBLH almost comparable to that by the total column BC (Figure S9). Figure 3 also indicates that PBLH could be increased by BC under conditions of a low surface flux change (see the left-hand side of the figure). Further investigations suggest that this phenomenon occured mainly in regions having relatively low BC emissions and high surface albedo (Figures S8b and 8c), but in the regions having high BC emissions and strong solar radiation intensity, such as in the megacities of the eastern China, the Sichuan Basin, and South China, a high probability of a PBLH decrease prevailed (Figure S8d).

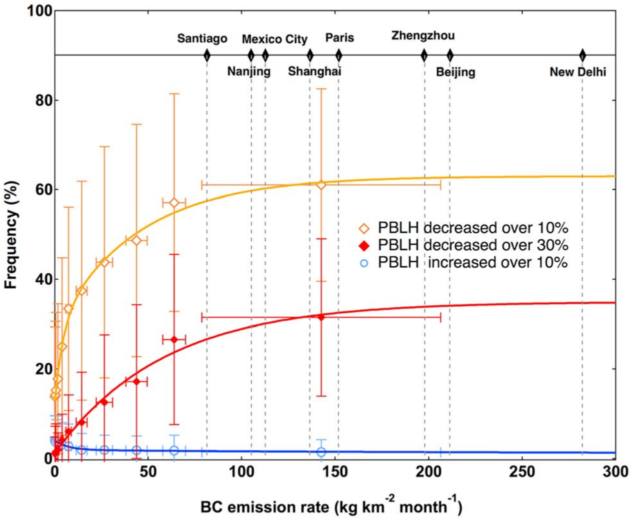

Figure 4. Frequency of days with the afternoon (12:00–16:00 LT) PBLH change (decreased and increased over 10% and decreased over 30%) as a function of BC emission rate, based on all grids in the simulation domain during December 2013. The whiskers show the standard deviation. Bold solid lines give double-exponential fit for the three data sets. Black diamond symbols show the corresponding emission rate of BC in megacities in China and other countries. Emission data of the Chinese cities are from MEIC database provided by Tsinghua University, and those of other cities are from the MACCity inventory from ECCAD website (http://eccad.sedoo.fr).

3. Discussions

Our simulations show that the main megacities, including Beijing, Zhengzhou, Nanjing, and Shanghai in eastern China, had a high frequency of decreased PBLH during the 1 month period (Figure 4). In general, the probability of a decreased PBLH is expected to first increase and then saturate as a function of the BC emission rate in a megacity. The nonlinear relationship probably suggests that the aerosol-PBL feedback to unit quantity of BC will be lower in higher aerosol loading case as solar radiation weakened. This means that if BC emissions were reduced down to a certain threshold, e.g., lower than 80 kg km^{-2} month^{-1}, a "nonlinearly" quick improvement of air quality will be achieved. In China, although the central government has stepped up efforts to control PM pollution after realizing the negative impacts from $PM_{2.5}$ on human health, the main control efforts have been made for the precursors of non-BC aerosols, such as SO_2 and NO_x, while a continuously increased BC emission has been reported in the past decades with an even accelerated trend during the latest years [*Qin and Xie*, 2012] (Figure S10). Our analysis indicates that lacking an efficient control of BC emissions may cancel out other efforts of air pollution measures through the aerosol-PBL interactions induced suppression of PBL development and even probably increase the frequency of extreme haze episodes. Furthermore, Figure 4 shows that the feedback mechanism introduced here not only concerns megacities in China but is also likely to operate in other well-known polluted megacities in the world, such as New Delhi, Paris, Mexico City, and Santiago.

The results clearly demonstrate that BC aerosols could substantially decrease the height of the PBL and hence enhance the surface haze pollution by accumulating air pollutants into a lower PBL. BC's key role in the aerosol-PBL feedback includes its cooling effect around surface, with a contribution almost half of the surface flux reduction induced by all aerosols, and its upper PBL heating, which constrains the daytime development of PBL in a comparable magnitude with the surface cooling effect. Figure 5 provides a conceptual scheme to show how BC and other absorbing aerosols influence the PBL in a megacity through the aerosol-radiation-PBL feedback loop. Considering that the polluted PBL often looks like a dome covering a city, we name the effect of BC on PBL development as its "dome effect." This kind of dome effect not only occurs in a city but also influences PBL meteorology in rural areas surrounding or downwind the city by the long-range transport of BC.

Our results presented in this study call for more restricted control measures for emissions of BC and other light-absorbing fine particles in order to tackle the dome effect of BC on extreme haze pollution in megacities. In fact, Chinese government has made many efforts to improve haze pollution in recent years. For example, the Action Plan for Air Pollution Prevention and Control (2013–2017) released in 2013 has set a strict target for the three developed regions: the Beijing-Tianjin-Hebei region, the Yangtze River Delta (YRD) region,

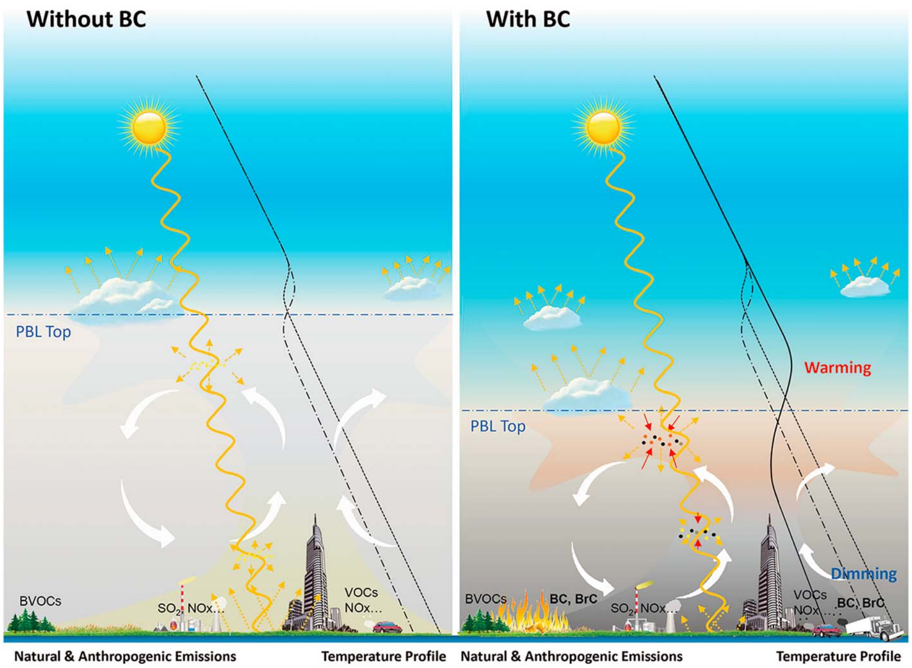

Figure 5. Aerosol-PBL feedback loop for the scenarios without and with BC emission in a megacity. Black lines show the vertical profile of air temperature (solid, dash-dotted and dotted lines for the scenarios with BC, with aerosols except for BC and without aerosols, respectively). Yellow dashed arrows show the reflection of solar ration by the ground surface, clouds, and scattering aerosols. Red arrows show the absorption of solar radiation by absorbing aerosols like BC and BrC. The blue dash-dotted line indicates the top of PBL. White arrows indicate the vertical ventilation of urban plumes induced by circulations or large eddies induced by the urban heat island effect. The difference in the color of urban plumes means different chemical compositions: the case on the right has more BC (or BrC) from sources like biomass burning, industry, and diesel vehicles, along with more aged air in the upper PBL.

and the Pearl River Delta (PRD), which all consist of megacities and city clusters [*Sheehan et al*., 2014]. In this action plan, a mandatory reduction of $PM_{2.5}$ annual mean concentration has been planned for the five years to 2017: with 25% for Beijing, 20% for YRD cities, 15% for PRD cities, and 10% for other megacities. Many efforts and huge economic cost have been devoted to the control measures to meet this target. Our study suggests that a specified control policy for BC other than $PM_{2.5}$ control alone will be a more efficient way to mitigate the current haze pollution. According to *Qin and Xie* [2012], more efforts to reduce BC emission could be realized by concentrating on industry, biomass/biofuel burning, diesel vehicle, and coal burning.

At the same time, as one of the short-lived climate forcers, the reduction of BC is a viable way to mitigate global warming [*Jacobson*, 2001; *Ramanathan et al*., 2007; *Andreae and Ramanathan*, 2013; *Bond et al*., 2013]. Even though more and more lines of scientific evidence demonstrated this fact, there is no direct motivation from municipal even national governments to make special control measures to reduce BC emissions as main concerns have been put on reducing CO_2 emission. Since mitigation of air pollution is a current main concern of megacities, especially in China, implementing such kind of policy at a municipal level is expected to cause a substantial reduction of BC concentrations, which hence will certainly cobenefit the mitigation of climate change from regional to global scales.

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