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Impact of Aerosol-PBL Interaction on Haze Pollution: Multiyear Observational Evidences in North China

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Key Points:
- Aerosol-induced upper-air heating and surface dimming exist during heavy haze pollution in North China
- Light-absorbing aerosols in the atmosphere like black carbon play a vital role in aerosol-boundary layer interaction
- An index derived from radiosonde observation and reanalysis data well characterizes the intensity of such interaction

Supporting Information:
- Supporting Information S1

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Abstract Atmospheric aerosols have been found to influence the development of planetary boundary layer (PBL) and hence to enhance haze pollution in megacities. Previous works on aerosol-PBL interaction were mainly based on model simulation for short-term cases; so far, there is a lack of long-term observational evidences. In this study, based on multiyear measurements and reanalysis meteorological data, we give observational evidences on aerosol-PBL interaction and its impact on pollution aggravation. We found a significant heating in upper PBL with maximum temperature change about 0.7 °C on average and a substantial dimming near surface with a mean temperature drop of −2.2 °C under polluted condition. By integrating Eulerian forward simulation and Lagrangian backward trajectory calculation, we demonstrated that the atmospheric heating was mainly induced by light-absorbing aerosols like black carbon. Then an index representing such effect was proposed, which could well characterize aerosol-PBL interaction and its impact on air pollution.

Plain Language Summary In polluted regions like China, atmospheric aerosols can influence the meteorology by warming the upper air and blocking sunlight that would otherwise warm the surface. The opposite temperature tendencies due to aerosol, that is, atmospheric heating and surface dimming, make the air increasingly stable and stagnant. These effects then greatly weaken the diffusion and dilution of pollutants, thereby worsening air quality.

1. Introduction

Aerosols in the atmosphere could directly or indirectly influence the radiation balance of the Earth-atmosphere system and hence regional or global climate change (Ackerman et al., 2004; Bond et al., 2013; Boucher et al., 2013; Menon et al., 2002). Recent studies have found that the impact of aerosols, particularly absorption aerosols like black carbon (BC) and dusts, to solar radiation is also important to the development of planetary boundary layer (PBL) and the air quality within the PBL (Ding et al., 2013, 2016; Liu et al., 2016; Z. Wang et al., 2018; Wilcox et al., 2016; Yang, Russell, et al., 2017).

Key processes of aerosols' impact on PBL meteorology could be identified as two parts: the surface dimming and upper-level heating. First, the aerosols, including both absorption and scattering aerosols, have a bulk effect to block solar radiation reaching to the ground surface, resulting in an overall dimming effect in the ground surface and suppressed PBL development due to weakened surface heat flux (Ding et al., 2013; Z. Li, Guo, et al., 2017; Petäjä et al., 2016; Quan et al., 2013). Second, the solar absorption aerosols in the atmosphere could heat the atmosphere by trapping solar energy and result in strong heating in upper PBL or around the PBL top, which change the vertical temperature stratification and further suppress the development of PBL (Ding et al., 2016, 2017; Z. Wang et al., 2018). Ding et al. (2016) have named the aforementioned processes as the *dome effect* of aerosols and revealed that such kind of effect could substantially enhance near-surface haze pollution in megacities. Such modification in temperature stratification due to aerosol-induced upper-air heating and surface dimming effect and consequent near-surface pollution aggravation is defined as aerosol-PBL interaction and will be further investigated in this work.

Due to tremendous anthropogenic emission, the North China has been suffering from heavy haze pollution in cold seasons (Z. Wang et al., 2018; Zhang et al., 2016). Extremely high concentration of scattering and absorption aerosols have been observed frequently in this region in winter, especially in megacities like Beijing. Previous studies have shown that the aerosol-PBL interaction was very important in enhancing haze pollution in these cities (Ding et al., 2016; Gao et al., 2015; J. D. Wang et al., 2014). However, the existing works were mainly based on modeling simulation for episodes or a period about 1 or 2 months. There is still a lack of statistically meaningful observational evidences to show the roles of aerosol-PBL interaction on haze pollution. In this study, based on multiyear ground based observation of particulate matter (PM) with aerodynamic diameters 2.5 μm and smaller ($PM_{2.5}$) concentrations and radiosonde measurements of air temperature in Beijing together with global meteorological reanalysis data, we present climatological observational evidences to show how important the aerosol-PBL interaction is for haze pollution in the most polluted North China megacities, and we also define an index based on air temperature from radiosonde observations and reanalysis data to quantify the impact of aerosol-PBL interaction.

2. Materials and Methods

2.1. $PM_{2.5}$ Observation

Available ground based $PM_{2.5}$ concentration observations in the North China were used in this study. For the entire North China, we use data measured by routine air quality monitoring stations. Since 2013, real-time air quality monitoring at more than 1,000 stations across China has been done by the Ministry of Environmental Protection, China, and the data can be openly accessed through the ambient air monitoring data center. This air quality monitoring network provides more information on the spatial distribution and its evolution characteristics across China, especially in East China (supporting information Figure S1). For Beijing, in order to obtain a more longer data record, we used hourly concentrations of $PM_{2.5}$ measured in the U.S. embassy during 2010–2016, which were acquired from U.S. Department of State air quality data files (available at http://www.stateair.net). Detailed information on this data set can be found in the supporting information S1 (Liang et al., 2016).

2.2. Radiosonde and Reanalysis Data and the Calculation of Aerosol-PBL Interaction Index

By comparing radiosonde with reanalysis data, the observation minus reanalysis (OMR) method provides a good indicator for the aerosol-PBL interaction because the model-simulation-based reanalysis data have not considered the impact of aerosols but only with limited upper atmospheric measurement data assimilated (Ding et al., 2013, 2016). To make comprehensive statistics for multiyear pollution episodes, we used radiosonde and reanalysis data since 2010 in the North China. For the radiosonde data, air temperature profiles are measured twice a day (00 universal time coordinated [UTC] and 12 UTC). We used the Integrated Global Radiosonde Archive, which compiles all these radiosonde measurements into easily accessible and quality-assured data set. The mandatory pressure levels for the measurements include 1,000, 925, 850, 700, 500, 400, 300, 250, 200, 150, 100, 70, 50, 30, 20, and 10 hPa.

For the reanalysis data, we used the ERA-Interim data, which is the latest global atmospheric reanalysis produced by the European Centre for Medium-Range Weather Forecasts (ECMWF; Dee et al., 2011). It is generated from a spectral model (ECMWF Integrated Forecast System) and has not considered the impact of aerosols on meteorology yet (Simmons, 2006). ERA-Interim data have assimilated multiple measurements through a four-dimensional variational data assimilation system in 12-hourly analysis cycles (Thepaut et al., 1996). However, prior state estimates from forecast model determine how much the observational information can be retained and indeed lead to a large drop between the total number of available data and the number of data passed to the assimilation. In fact, only less than 50% of total radiosonde measurements are assimilated in ERA-Interim data set (Poli et al., 2010). Therefore, disparities between atmospheric sounding observations and ERA-Interim reanalysis data could somehow reflect the effects induced by aerosols statistically.

Previous studies (e.g., Ding et al., 2013, 2016) suggest that the aerosol-PBL interaction is generally characterized by the opposite perturbations in temperature aloft and that near surface, which can be reflected by the gaps between direct radiosonde observations and reanalysis data (as shown in Figure 2a in Ding et al., 2016). Therefore, on the basis of these two sets of data, we propose an index representing the overall effects on upper-PBL heating and surface dimming (so called HD-index), which implies the intensity of aerosol-PBL interaction. Briefly, the HD-index can be defined as the summation of negative temperature deviation near surface and positive temperature deviation in the upper air in absolute terms. Specific to ERA-Interim data, it is calculated by summing up the absolute value of averaged temperature difference (OMR) under 950 hPa and that between 850 and 950 hPa at 12UTC. To minimize the influence of strong cold fronts, calculation was not performed if wind speed at 850 hPa is greater than 10 m per second. Here, we adopted radiosonde observations at 12UTC (20 pm Beijing time) instead of those at 00UTC (8 am Beijing time) since that temperature modification was mainly caused by aerosols' perturbation in solar radiation transfer.

$$\text{HD-}\mathit{Index} = \sum_{i=1}^{n} |\Delta T_i| / n + \sum_{j=1}^{m} |\Delta T_j| / m$$

where n and m mean the numbers of pressure levels under 950 hPa and those between 850 and 950 hPa, respectively. ΔT_i and ΔT_j are the temperature difference at level i near the ground surface and level j in the upper air.

3. Results and Discussions

Previous studies (Ding et al., 2013, 2016) already demonstrated that the difference in air temperature between radiosonde and reanalysis data (OMR) serves as a good indicator of the anthropogenic impacts, including aerosol-PBL interaction, as such kind of processes have not been well considered in the operational global meteorological models yet. To illustrate how well this method works in North China, we investigate a heavy haze pollution episode in Beijing in mid-December 2016. As shown in Figure 1a, during 16–21 December, Beijing was suffering from a week-long severe pollution, with $PM_{2.5}$ concentration increased gradually in the first few days and then kept in severe polluted level (about 400 μg/m^3 in average) for the last 2 days. To understand the evolution of this pollution episode, we investigate the air temperature profiles and air masses trajectories for different phases during this event, that is, 12 UTC on 16, 18, and 20 December 2016. A more detailed discussion of the methodology can be found in the supporting information S1 (Draxler, 1996; Grell et al., 2005; Huang et al., 2014; M. Li, Zhang, et al., 2017). Although inversion layers existed in all 3 days in general, the PBL stratifications were quite different (Figure 1b). The intensity of temperature inversion was increasing with pollution accumulation, accompanied with decreased height of capping inversion with a well-defined one formed on 20 December 2016. Figure 1c shows the difference between radiosonde observation and reanalysis data corresponding to Figure 1b. It is indicated that the OMR differences in air temperature were all characterized as upper-PBL heating and lower-PBL dimming with intensity of both them gradually increased as the haze pollution deteriorated.

Ding et al. (2016) gave a comprehensive interpretation about the upper-PBL heating based on WRF-Chem (the Weather Research and Forecasting model coupled with Chemistry) simulation. They pointed out that the main causes for the upper-PBL heating is the solar absorption of light-absorbing aerosols like BC, which have a much stronger heating efficiency around the top of PBL (Ding et al., 2016; Z. Wang et al., 2018). Previous studies have identified BC as the most essential light-absorbing aerosol in China (Bond et al., 2013; Shen et al., 2018). Consequently, to shed more light on this process during this event, we calculated backward trajectories for air masses at an altitude of 1 km over Beijing for the 48 hr and diagnosed the BC concentration and solar radiation along the trajectories based on WRF-Chem simulation (Figures 1d and 1e; detailed methodologies and validation are given in Texts S3 and S4). The trajectories show that during this 1-week episode, the enhancement of haze pollution was partly due to gradually declined pressure gradients in the North China Plain (Figure S2). The air masses around the altitude of 1 km were mainly from northwest on 16 and 18 December but from southwest on 20 December. Similar to findings in previous studies (Yang, Wang, et al., 2017), the transport pathway indicated that heavy haze in Beijing is usually associated with anomalous southwesterly wind. Before arriving Beijing, the air masses were subject to distinct emission intensity and shortwave radiation condition. For example, on 20 December, the air masses contained high concentration of BC and had a longer time residence in the PBL, as shown in Figure 1e. If considering the shortwave radiation, the overall heating effects of BC gradually increased from 16 to 20 December 2016.

Since currently, the ECMWF model has not included such kind of effects; the heating of light-absorbing aerosols may lead to large temperature difference from radiosonde data. Of course, the aerosols-PBL interaction will also cause a substantial dimming effects in the lower PBL. In fact, such kind of upper-PBL heating and lower-PBL dimming not only existed over megacities like Beijing but also on regional scale. Figure 2 gives the spatial distributions of $PM_{2.5}$ concentration extrapolated from in situ measurements at hundreds of monitoring stations (Figure S1) and OMR temperature difference in upper air (925 hPa) and near-ground surface (1,000 hPa) for 16 and 20 December 2016. Comparatively, temperature difference was much more significant on the polluted day (20 December) than that under relatively clean condition on 16 December. Another clue is that such kind of upper-PBL heating and low-PBL dimming pattern coincided well with pollution

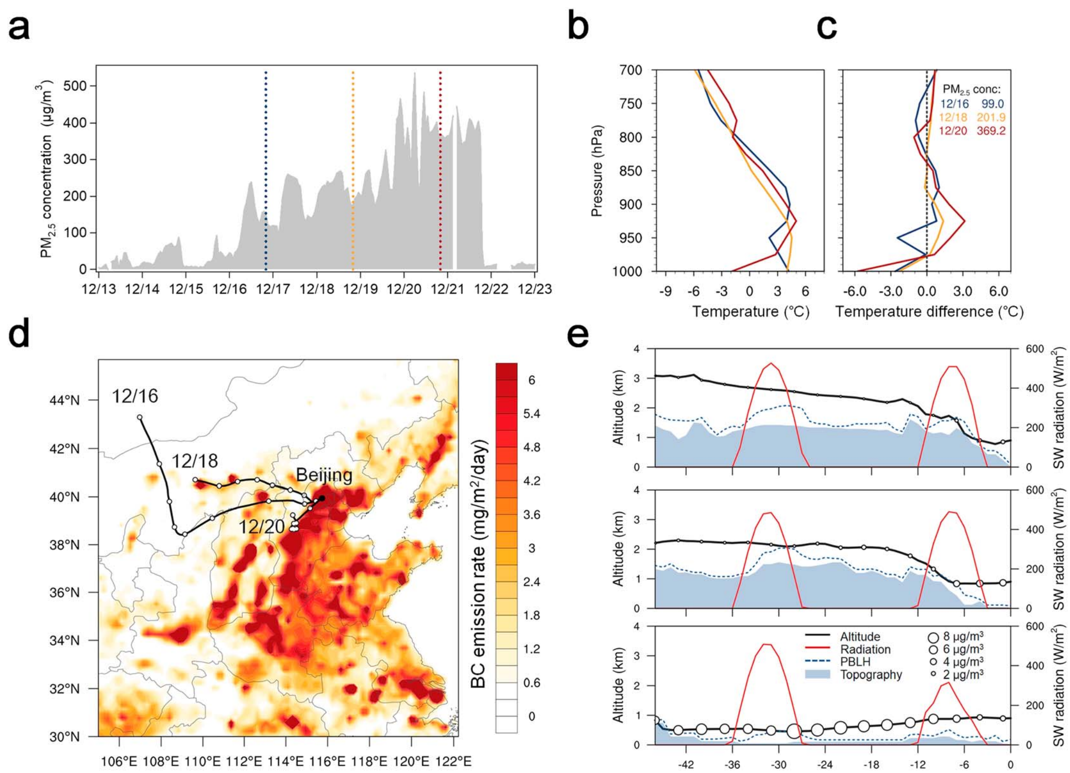

Figure 1. Time series of $PM_{2.5}$ concentration evolution during typical haze event in December 2016 (a). Vertical profiles of radiosonde-observed air temperature (b) and their difference from ERA reanalysis data (c) at 12 universal time coordinated on 16, 18, and 20 December. Daytime $PM_{2.5}$ concentration are labeled in the same color order at the top-right corner. Dash solid zero lines are shown for reference. Spatial distribution of BC emission rate in December and 48-hr backward trajectories at 12 universal time coordinated on 16, 18, and 20 December 2016, respectively (d). Air mass altitude (black lines), BC (black circles), PBL height (blue dash lines), and topography (blue areas) along the backward trajectories on 16, 18, and 20 December 2016 (e). BC = black carbon; PBL = planetary boundary layer.

distribution, with much stronger signals in regions with higher PM concentrations. As revealed by Z. Wang et al. (2018), the dome effect not only influences in megacities but also in the downwind countryside or rural area, resulting in regional-scale air pollution deterioration.

Continuous observations on temperature profile and $PM_{2.5}$ concentration in Beijing since 2010 make it possible to provide a climatological perspective on how significant the aerosol-PBL interaction is during hazy days. Similar method (i.e., OMR) was applied to get statistic result of temperature difference during polluted and nonpolluted conditions in winter during 2010–2016. Given that surface pollution is well correlated with PBL pollution in general (Figure S3), here surface $PM_{2.5}$ concentrations were employed to identify air pollution condition. Figures 3a and 3b give the overall vertical profiles of air temperature difference between radiosonde observation and the ERA reanalysis data for polluted and *clean* days, when daily $PM_{2.5}$ concentrations were greater than the 75th percentile and less than the 25th percentile, respectively. A comparison of these two clearly indicates that the air temperature profile was substantially changed by aerosol pollution. Under clean conditions, radiosonde-observed temperature and that in reanalysis data were consistent with each other in the entire low troposphere (Figure 3b). However, during polluted days, temperature stratification was significantly modified in the lower troposphere, particularly under 800 hPa. Specifically, positive temperature difference existed between 925 and 800 hPa with a maximum around +0.7 °C, and the cooling bias was increasingly substantial while getting closer to the ground surface (at 1,000 hPa) and reached up to approximately −2.2 °C. The warming tendency in the upper PBL was always smaller than the dimming effect near surface. It is expectable because that surface cooling somehow reflected the overall effect of aerosol extinction including both shortwave radiation absorption and scattering, while the atmospheric heating was merely related to the light absorption (Huang et al., 2015). Moreover, part of the heating effect in the

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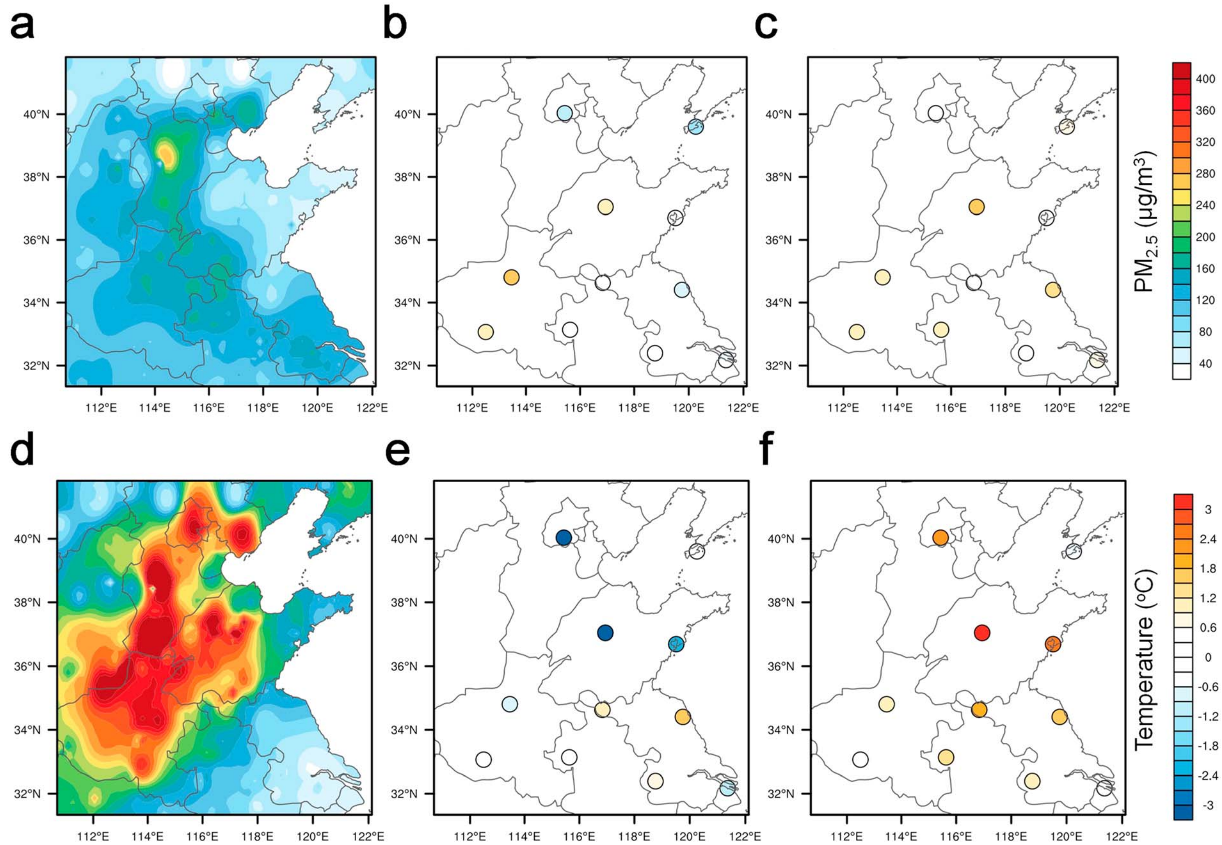

Figure 2. Spatial distribution of $PM_{2.5}$ concentration (a, d) and air temperature difference between radiosonde observation and ERA reanalysis data near surface (b, e) and in the upper air (c, f) on 16 and 20 December 2016.

upper air could be offset by the surface dimming through turbulence and convective mixing in the PBL (Z. Wang et al., 2018).

It has been demonstrated that the key feature of aerosol-PBL interaction is the synchronal upper-air heating and surface dimming. Therefore, an index (HD-index) representing the intensity of aerosol-PBL interaction was proposed based on temperature difference between radiosonde observation and reanalysis data, as described in section 2. First, we checked if this index can capture aggravating aerosol-PBL interaction as pollution deteriorates. Theoretically, both atmospheric heating and near-surface dimming tend to accumulate and then resulted in greater HD-index just as illustrated by using one-dimensional model which excluded the influences from synoptic weather (Figure S4). On the basis of air quality measurements, six haze pollution episodes during past years that lasted 5 days was collected, which were identified by monotonically increasing daily $PM_{2.5}$ concentrations and the maximum over 200 $\mu g/m^3$. As displayed in Figure S5, HD-index increased progressively with $PM_{2.5}$ concentrations during these long-lasting haze pollution episodes though under different synoptic conditions, implying intensified aerosol-PBL interaction during the evolution of haze pollution event.

The HD-index was correlated well with particle pollution levels, not only during pollution events but also in long-term period. The relationship between HD-index and daytime $PM_{2.5}$ concentration in winter during 2010–2016 is illustrated in Figure 4a. Here instead of daily mean concentration, daytime mean value was utilized since that aerosol-PBL interaction builds up only when there exists solar radiation. As shown, the index values were correlated very well with daytime $PM_{2.5}$ concentration with the Pearson's correlation coefficient (R) as high as 0.56, indicating that higher concentration of $PM_{2.5}$ coincided with more substantial temperature bias in reanalysis data. Some existing studies suspected that extremely high loading of scattering aerosols may play the dominate role in enhancing PBL stability through backscattering solar radiation (Qiu et al., 2017). However, quite good linear negative relationship of surface dimming and

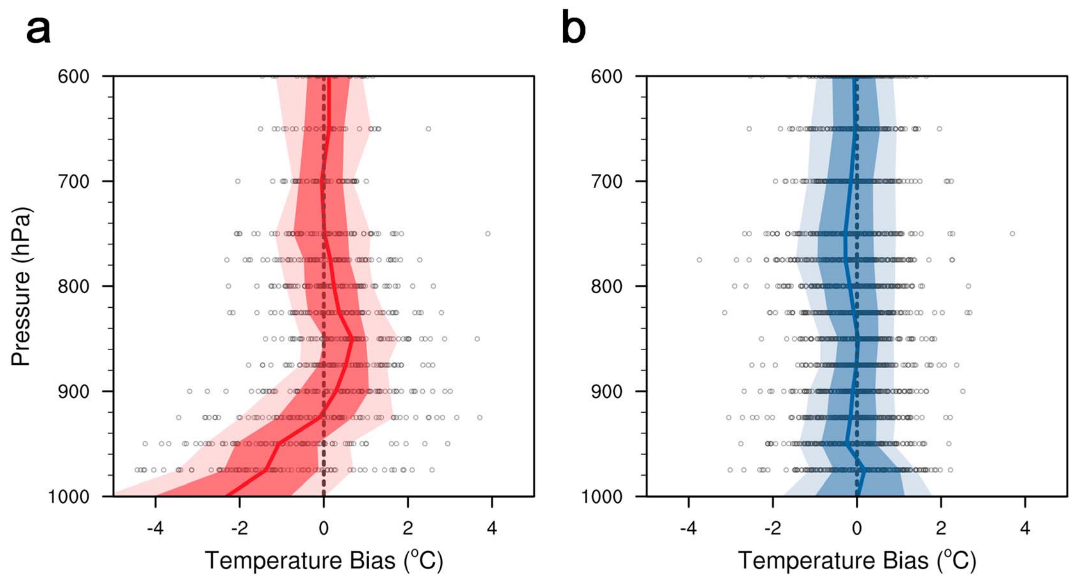

Figure 3. Vertical profiles of temperature bias between radiosonde observation and ERA reanalysis in Beijing under polluted (a, $PM_{2.5}$ concentrations greater than the 75th percentile) and clean (b, $PM_{2.5}$ concentrations less than the 25th percentile) conditions in winter during 2010–2016. Solid lines and deep and light shadows represent average, 25–75th percentile, and 10–90th percentile, respectively. Dash solid zero lines are shown for reference.

overall upper-level heating shown in Figure S6 provided an important observational clue to the significance of light-absorbing aerosols, so did previous shortwave radiation budget studies (Huang et al., 2015; Ramanathan & Carmichael, 2008).

Greater HD-index corresponded to increasing PBL stability, whereby weakening the vertical diffusion of locally emitted air pollutants and suppressing the air pollutants in a shallower PBL. The resultant enhancement in near-surface air pollution has been clearly analyzed in Ding et al. (2016). Based on long-term observations, we also found that the increment in daily $PM_{2.5}$ correlated well with HD-index increase (Figure 4b). One-dimensional meteorology and chemistry online coupled simulation confirmed the similar signal, that is, near-surface $PM_{2.5}$ concentration growth accelerated as HD-index increase. As demonstrated in Figure S4,

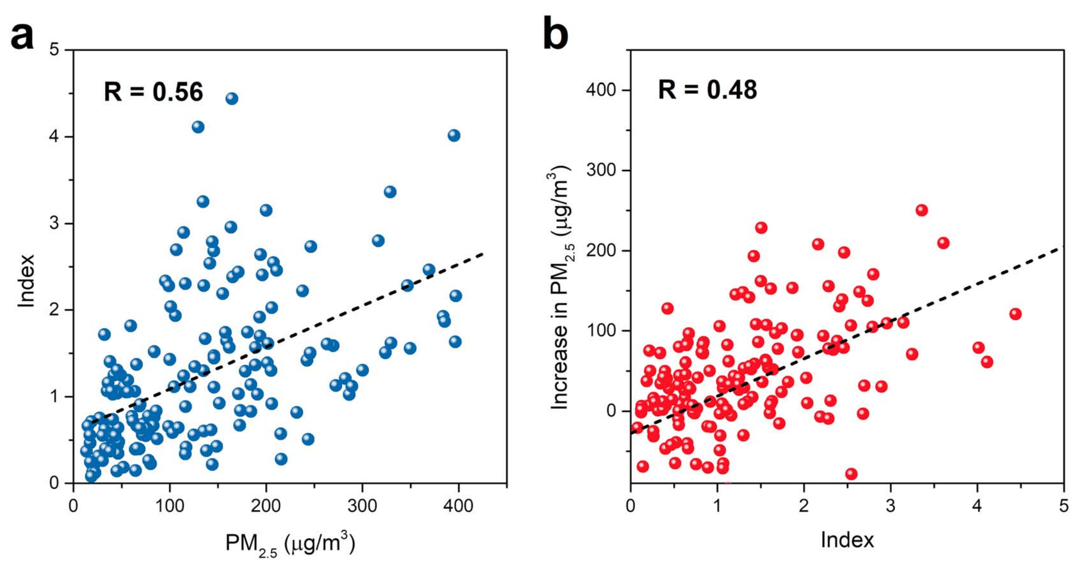

Figure 4. (a) Scatter plots of HD-index versus $PM_{2.5}$ daytime mean concentration. (b) Daily increase in $PM_{2.5}$ concentration versus HD-index in winter (December-January-February) during 2010–2016.

the atmospheric heating and surface cooling actually accumulate over time and weaken the turbulent mixing gradually. Subsequently, suppressed vertical diffusion makes the locally emitted pollutants concentrate in a much shallower PBL and accumulate near the surface. Both observations and simulations imply that increased stability caused by aerosol-PBL interaction may continue to influence the atmospheric stratification and deteriorate the pollution on the next day.

4. Summary

In this study, multiyear air quality and meteorological measurements together with reanalysis data were combined to provide observational evidences on aerosols' effect on PBL evolution. By comparing the in situ measured temperature and reanalysis data, we found that a significant heating in upper PBL and a substantial dimming near surface under polluted condition. Such modification on temperature stratification aggregated surface air pollution, which closely related to pollution exposure and human health. The HD-index derived from the difference between radiosonde observation and reanalysis data can well characterize the intensity of aerosol-PBL interaction and also its evolution during haze events. Both observations and simulations using multiple models suggested that light-absorbing aerosols exert crucial parts on such interaction, indicating that emission reduction on carbonaceous aerosols may serve as a cost-effective way to mitigate air pollution. Given the vigorous regional transport of air pollution in China, such emission control may not only mitigate local air pollution but also benefit air quality on a regional scale.

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