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Linkages between aerosol pollution and planetary boundary layer structure in China

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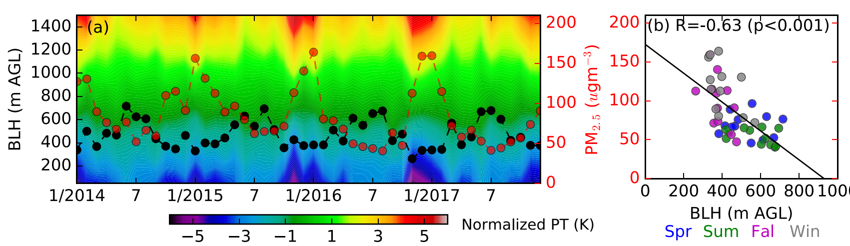
HIGHLIGHTS

- PBL-aerosol linkages in China are elucidated using 4-year sounding data.
- Seasonal variation in the PBL modulates seasonal variation of pollution.
- Strong thermal stratification and a shallow PBL are responsible for high pollution levels in winter.

GRAPHICAL ABSTRACT



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ABSTRACT

China suffers from high levels of $PM_{2.5}$ pollution, which is often exacerbated by unfavorable planetary boundary layer (PBL) structures. Partly due to a lack of appropriate observations, the PBL-aerosol linkages in China are not clearly understood. Thus, we investigated these linkages from a national perspective using sounding data collected from 2014 to 2017. Correlation analyses revealed a significant anti-correlation between monthly boundary layer height (BLH) and aerosol pollution that was ubiquitous across China, indicating the important role of the PBL in regulating the seasonal variations of pollution in China. Besides, the day-to-day variations in pollution were modulated by the daily variabilities in the PBL structure. During winter, highly polluted days in most of the Chinese cities studied were associated with a low BLH, strong thermal stability, and weak PBL winds. In the North China Plain and Northeast China, the wintertime heavy pollution was often related to southerly winds and moister PBL. This study has important implications for understanding the crucial role that the PBL plays in modulating aerosol pollution in China.

1. Introduction

Rapid urbanization and industrialization during the past decades has led to a deterioration in the air quality in China (Chan and Yao, 2008; J. Li et al., 2016; San Martini et al., 2015). Consequently, air pollution that is characterized by high concentrations of aerosol particles having an aerodynamic diameter <2.5 μm ($PM_{2.5}$) has become a serious environmental problem in China, which is influencing air quality (Huang et al., 2014; Li et al., 2017; Yang et al., 2016), human health (Pope and Dockery, 2006; Chen et al., 2016; Huang, 2018), and the regional climate (Z. Li et al., 2016; Wu et al., 2016; Zhao and Garrett, 2015; Yang et al., 2018a). The aerosol radiative effects can modulate the thermodynamic stability and convective potential of lower troposphere (Z. Li et al., 2016; Zhao et al., 2018a). The aerosol can also alter cloud microphysical processes and influence precipitation by acting as cloud condensation nuclei or as ice nuclei (Twomey, 1977; Tao et al., 2012; Zhao et al.,

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2018b; Yang et al., 2018b). In addition to high emissions and the fast formation of secondary aerosols (Huang et al., 2014; Zhang et al., 2013; Zhao et al., 2018c; Guo et al., 2014), meteorological conditions play an important role in modulating the pollution levels in China (Ding et al., 2016; Fu et al., 2014; Miao et al., 2015a; Wang et al., 2018), especially the meteorological factors and processes within the planetary boundary layer (PBL) (Hu et al., 2014; Miao et al., 2017b; Ye et al., 2016; Zheng et al., 2017).

The PBL is the lowest portion of the troposphere and meditates the exchanges of heat, momentum, and pollutants between the surface and the free troposphere (Garratt, 1994; Stull, 1988). After release, the dispersion of pollutants is dynamically controlled by atmospheric motion (e.g., wind and turbulence) within the PBL (Stull, 1988). In addition, the thermal stratification in the PBL is important because it controls the intensity of the thermal turbulence (i.e., buoyancy) and the depth of the PBL (Liu and Liang, 2010; Medeiros et al., 2005; Miao et al., 2018b). Together, these PBL structures/processes not only regulate the horizontal dispersion and transport of aerosols, but also influence the upward dispersion of aerosols and the exchange of cleaner air from above (Li et al., 2017; Miao et al., 2015a, 2015b; Yang et al., 2016). A shallow PBL has been found to be partially responsible for several pollution episodes in China (Miao et al., 2018a; Quan et al., 2014; Ye et al., 2016). Meanwhile, the presence of aerosols can also modulate the PBL by aerosol radiative effects (Yang et al., 2016), which in turn exert feedback on pollution (Ding et al., 2016; Gao et al., 2015; Miao et al., 2016). Under certain synoptic conditions, the terrain can further exacerbate pollution by influencing the PBL structures/processes (De Wekker, 2008; Hu et al., 2016; Miao et al., 2015a; Wang et al., 2018).

Although the importance of the PBL in relation to aerosol pollution in China has been recognized (Li et al., 2017; Quan et al., 2013), most previous studies have been undertaken for specific cases or places. To what extent the local PBL structure influences aerosol pollution over most of China remains unclear, partly due to a lack of adequate PBL observations. A national perspective is required to explore the role of the PBL in aerosol pollution in China and may help to better understand pollution formation mechanisms. Fortunately, since 2011, a radiosonde network across China has been deployed by the China Meteorological Administration (Guo et al., 2016), which provides sounding data with a fine vertical resolution (~10 m). Based on these radiosonde measurements from 2011 to 2015, the spatial distribution and seasonal variation of the boundary layer height (BLH) in China were reported (Guo et al., 2016); however, few conclusions were drawn for aerosol pollution. In addition to the basic PBL climatology, the nationwide radiosonde network also offers us a unique opportunity to examine the relationship between the PBL structure and aerosol pollution in China.

As a necessary step to reveal the complicated causes (e.g., local PBL process/structure, chemical reactions, and regional transportation) for the severe aerosol pollution in most China, this study investigates the linkages between the PBL and aerosol pollution in China using long-term sounding measurements and $PM_{2.5}$ observations from a national perspective. The results from this study could serve as a reference for evaluating the influence of the PBL on aerosol pollution in China and for obtaining a better understanding of the formation mechanisms underlying severe pollution.

2. Data and method

In the present study, aerosol pollution levels were denoted by hourly ground-level $PM_{2.5}$ concentration measurements of China National Environment Monitoring Center. The real-time $PM_{2.5}$ measurement in China was first released to public in January 2013. Then, the monitoring network was extended to cover most major cities in China in the end of 2013 (Zhang and Cao, 2015; Zhao et al., 2016). This study investigated the $PM_{2.5}$ pollution from 2014 to 2017, using the measurements collected from 235 monitoring sites (marked by grey crosses in Fig. 1a) in 28 major cities across China. At each monitoring site, the hourly

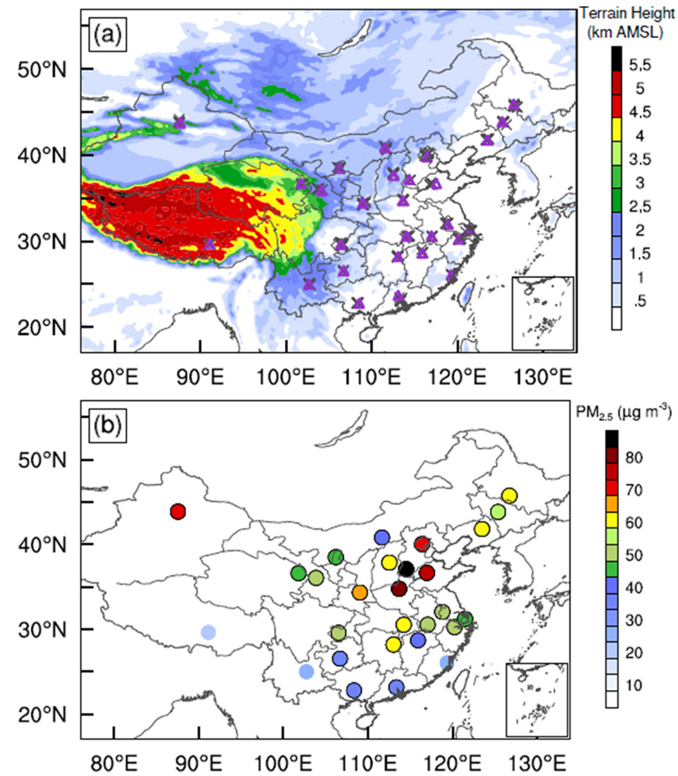

Fig. 1. (a) Map showing $PM_{2.5}$ monitoring (grey crosses) sites and sounding stations (violet triangles) in 28 major cities in China, overlaid with terrain height. (b) Spatial distribution of average $PM_{2.5}$ concentration in China from 2014 to 2017. The city exceeding the annual limit of the Chinese Ambient Air Quality Standards (35 μg m^{-3}) is shown in black outline. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

mass concentration of $PM_{2.5}$ was measured using the micro oscillating balance method (TEO from Rupprecht & Patashnick Co., Inc., USA) and the β absorption method (BAM 1020 from Met One Instrument Inc., USA or Tianhong Co., China or Xianhe Co., China), which can provide accurate measurements of aerosols (Zhang and Cao, 2015; Zhao et al., 2016). The instrument operation, maintenance, data assurance and quality control were all properly conducted based on the China Environmental Protection Standards. The uncertainty of $PM_{2.5}$ mass concentration is <5 μg m^{-3}.

To investigate the linkages between the PBL and aerosol pollution, radiosonde measurements from 2014 to 2017 were also collected from all 28 cities (Fig. 1a). All these sounding stations are equipped with L-band radiosonde systems (Guo et al., 2016), which provide high vertical resolution profiles of potential temperature (PT), humidity, and wind twice a day at 0800 and 2000 Beijing Time (BJT). As reported by previous studies (Bian et al., 2011; Ma et al., 2011; Guo et al., 2016; Zhang et al., 2018), the accuracy of L-band sounding system is similar to that of GPS RS 92 radiosonde (Vaisala Co., Finland) within the lower troposphere. Specifically, the accuracy of PBL temperature is <0.1 K. Therefore, the L-band radiosonde measurements are good enough to investigate the PBL thermal structures. In total, there were 80,671 soundings available for the present study: 40,311 soundings at 0800 BJT and 43,360 soundings at 2000 BJT. As illustrated in Fig. S1, most cities have >1400 soundings at both 0800 and 2000 BJT. In addition, hourly 2-m temperature, 10-m wind, and precipitation measurements were collected at the same sounding stations. The air quality monitoring sites and sounding stations were lumped together according to the district of each studied city (Fig. 1a).

To characterize the relationship between the PBL structure and pollution, the BLH concept has been widely applied (Guo et al., 2016; Seibert, 2000; Seidel et al., 2012), because it involves the vertical