Impacts of Atmospheric Boundary Layer Vertical Structure on Haze Pollution Observed by Tethered Balloon and Lidar

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ABSTRACT

In this paper, the characteristics of the atmospheric boundary layer (ABL) vertical structure over the North China Plain (NCP) during a comprehensive observation experiment conducted during 15–21 December 2018 were investigated. Observational data were obtained with a large tethered balloon, Doppler wind lidar, and ground-level instruments. The maximum concentration of PM$_{2.5}$ exceeded 200 μg m$^{-3}$, and the ratio of PM$_{2.5}$/PM$_{10}$ was approximately 0.4 (its maximum was approximately 0.8) during the whole observation period, indicating the explosive growth of dominant fine-mode aerosols in the winter heating season. Elevated concentrations of pollutants decreased the solar irradiance received by the ground, resulting in lower temperature at ground level. Our results illustrate three distinct types of vertical profiles: Type 1 (convective state)—the concentration of PM$_{2.5}$ decreased nearly linearly with increase of the height below approximately 600 m; Type 2 (stable state)—the PM$_{2.5}$ concentration sharply decreased from the ground to approximately 200 m; and Type 3 (multilayer structure)—some pollutants were suspended aloft in the upper air layer. Diurnal evolution of the vertical profiles of PM$_{2.5}$ and their relationship with the changes in meteorological factors were identified. From daytime to nighttime, the vertical profiles evolved from Type 1 to Type 2 or Type 3. All the 33 vertical PM$_{2.5}$ profiles that we obtained showed a strong relationship with elements of the ABL structure, such as the distributions of winds, the inversion layer, and turbulence activities. A light-wind layer and weak turbulence activity, especially within the inversion layer, contributed greatly to the accumulation of pollutants. Vertical PM$_{2.5}$ concentration patterns were also greatly affected by local ground-level emission sources and regional transport processes.

Key words: haze, pollution, PM$_{2.5}$, vertical structure, inversion, atmospheric boundary layer (ABL)

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1. Introduction

Rapid industrial development and urbanization have caused severe air pollution in China. A large number of anthropogenic pollutants have been emitted into the air, posing a significant threat to the living environment (Zhao et al., 2013; Sun et al., 2015). Environmental pollution impacts air quality, climate change, and human health. Deterioration in air quality is mainly caused by air pollution. Increased aerosols in the air could affect climate through their effects on the radiation budget. According to the World Health Organization, up to a third of lung cancer and heart disease deaths are attributable to air pollution (Mauderly and Chow, 2008; Jacob and WInner, 2009).

As the largest developing country in the world, China has experienced severe haze pollution attacks, which have frequently occurred over the North China Plain (NCP) (Huang et al., 2014; Lyu et al., 2018; Shen et al., 2018; Zhang et al., 2018). According to the China National Environmental Monitoring Centre (http://www.cnemc.cn/), measurements in the NCP region conducted

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in the first half of 2017 revealed that the daily average concentration of PM$_{2.5}$ (particulate matter with an aerodynamic diameter smaller than 2.5 μm) was 72 μg m$^{-3}$. China has strengthened its efforts on air quality monitoring and regulation since 2013 (Zhang et al., 2016; Sun, 2018), but the NCP still experiences long-lasting particulate matter pollution episodes (approximately 3–5 days in duration), especially in winter, under unfavorable dispersion conditions such as the low wind speed, high humidity, and high anthropogenic emissions (Sun et al., 2006; Quan et al., 2011; Zhao et al., 2013). Haze pollution processes in the NCP area usually persist for 3–4 days at a time in autumn and winter (Wu et al., 2017).

Haze pollution processes are closely related to the atmospheric boundary layer (ABL) structure, that is, the structure of the lowest part of the atmosphere directly influenced by the surface of the earth (Li et al., 2019). Most of the pollutants or their precursors are directly emitted into the ABL (Gautam et al., 2016; Provençal et al., 2017). For example, NO$_x$ directly emitted from the surface in the nighttime was limited to approximately 40 m from the ground (Han et al., 2009). Under stagnant weather conditions, the lower ABL height limits the environmental capacity for pollutant dispersion, and the low wind speed also contributes to the persistence and increases in pollutants (Hu et al., 2014; Miao et al., 2017). In recent years, extensive studies have been conducted to characterize the sources, formation mechanisms, and evolution processes of severe haze pollution episodes (Zhang et al., 2013; Oozeer et al., 2016; Yang et al., 2017). The relationship between the ABL structure and pollution processes has also been reported. Han et al. (2009) analyzed the impact of the nocturnal boundary layer on urban air pollutants based on measurements from a 250-m tower.

The vertical distribution of pollutants in the ABL is very important for the study of pollutant evolution, accumulation, dispersion, and transport. A detailed description of the vertical and spatial distributions of pollutants can greatly improve air quality prediction models (Wang et al., 2014). Pollutant distribution is closely related to the ABL structure. The thermodynamic structure of the ABL affects vertical air pollutant mixing (Li et al., 2020). The occurrence of nocturnal low-level jets can also affect vertical pollutant profiles (Corsmeier et al., 1997; Salmond and McKendry, 2005; Hu et al., 2013). Largeron and Staquet (2016) reported that pollution episodes were primarily driven by persistent inversions. Simultaneous measurement of pollutants and meteorological elements is relatively difficult (Han et al., 2009). Many measurement techniques have been implemented to characterize the ABL structure and pollutants, e.g., ground-based lidar, aircrafts, unmanned aerial vehicles, and radiosondes (Sangiorgi et al., 2011; Li et al., 2017; Shi et al., 2019; Liu et al., 2020). There have been many studies based on ground observations but relatively fewer based on vertical observations, especially both meteorological and pollution vertical observations.

Tethered balloons are a powerful platform for detecting the physicochemical features of the ABL (Sangiorgi et al., 2011; Li et al., 2015; Zhang et al., 2017), especially large tethered balloons with high payload capacities (approximately 200 kg). They can carry many instruments, allowing direct measurements of the vertical distribution of pollution and meteorological elements simultaneously in the boundary layer with high degree of data reliability.

In this paper, we investigate the correlation between ABL structure and haze pollution during an intensive observation experiment conducted during 10–25 December 2018 over the NCP. Based on all the profiles observed during the study period, typical vertical PM$_{2.5}$ profiles were identified.

2. Description of the measurement site and instrumentation

During 10–25 December 2018, an intensive ABL observation experiment was conducted in Wangdu County (38.66°N, 115.25°E; the red dot in Fig. 1a), which is surrounded by city clusters (such as Beijing, Tianjing, and Shijiazhuang) of the NCP. The PM$_{2.5}$ concentration in the city clusters of the NCP had generally been high (Yang et al., 2017). During the winters of 2013 and 2014, the monthly average PM$_{2.5}$ concentration was as high as 151.2 μg m$^{-3}$ (Chen et al., 2017). In December 2016, Beijing issued a red-alert for a haze episode when the ground-level PM$_{2.5}$ concentration exceeded 450 μg m$^{-3}$ (Shi et al., 2019). On the basis of the data from sources such as a large tethered balloon, a Doppler wind lidar, and ground-level measurements, our comprehensive observations revealed the vertical distributions of both physical and chemical elements in the ABL. The instruments at the observation site in Wangdu County and the data employed in this study are as follows.

1. A tethered-balloon observation platform (shown in Fig. 1b) in Wangdu County simultaneously measured the vertical profiles of pollutants and meteorological parameters. The measurement instruments were mounted on a platform below the balloon (Fig. 1b). The concentrations of PM$_{2.5}$ and PM$_{10}$ were measured in real time with a time resolution of 1 minute by the instruments [MAS...
(Mini Air Station)-AF300, China] mounted on the tethered balloon. Additionally, a GPS [HC (Hemisphere Company)-12, China] instrument was deployed to track the balloon. Meteorological sensors (HC2-S, Rotronic, Switzerland) were also used to measure the air temperature ($T$), relative humidity (RH), and air pressure ($p$). The accuracy for particulate matter, $T$, RH, and $p$ observed with the tethered balloon was 0.1 μg m$^{-3}$, 0.1°C, 0.1%, and 0.1 hPa, respectively.

The large tethered balloon was 1900 m$^3$ in volume, and its maximum carrying weight was 200 kg, with the highest observation altitude at approximately 1000 m. A custom-designed cable connected the balloon and a ground-level computer. This cable provided power to the devices suspended below the balloon, and transferred measurement data to the ground-level computer. An electric winch controlled the ascent and descent rate of the balloon at approximately 0.5 m s$^{-1}$. As shown in Fig. 1b, there were no distinct emission sources around the observation site.

(2) A Doppler wind lidar system [FC (Fengcai)-II, Norinco Group, China] was installed at the same measurement location in Wangdu County, approximately 100 m away from the tethered balloon, to retrieve wind profiles with a spatial resolution of 50 m and a time resolution of 2–3 s. The accuracy of wind speed (WS) measurements was 0.4 m s$^{-1}$ (WS $\leq$ 10 m s$^{-1}$), 3% of WS (WS $>10$ m s$^{-1}$); and the wind direction (WD) accuracy was 3°.

**Fig. 1.** (a) Map of the local topography of the North China Plain (NCP). The red circle represents the specific measurement position in Wangdu County. The map was retrieved from Google Maps©. (b) Large tethered-balloon detection platform utilized during the observation period in Wangdu County. The Doppler wind lidar system, meteorological station, and atmospheric environment monitoring vehicle were located near the tethered balloon (approximately 100 m away).
(WS > 5 m s\(^{-1}\)).

(3) An atmospheric environment monitoring vehicle simultaneously measured the ground-level concentrations of PM\(_{2.5}\) (TEMO1405, Thermo Fisher, USA), NO (42i, Thermo Fisher, USA), NO\(_2\) (42i, Thermo Fisher, USA), SO\(_2\) (43i, Thermo Fisher, USA), CO (48i, Thermo Fisher, USA), and O\(_3\) (49i, Thermo Fisher, USA) every minute during the observation period. The atmospheric environment monitoring vehicle was also installed approximately 100 m away from the tethered balloon (Fig. 1b). The sampling accuracies for PM\(_{2.5}\), O\(_3\), and CO were 0.1 μg m\(^{-3}\), 1 ppb, and 0.01 ppm, respectively; and the accuracy for NO, NO\(_2\), and SO\(_2\) was 1 ppb.

(4) Meteorological parameters such as WS, WD, \(T\), and solar irradiance at 6 m above the ground were obtained from a meteorological station (FT-QXC7, Fengtu, China) at the observation site (Fig. 1b). The resolutions for WS, WD, \(T\), and solar irradiance were 0.1 m s\(^{-1}\), 1\(^\circ\), 0.1°C, and 1 W m\(^{-2}\), respectively.

In addition, indirectly observed parameters such as potential temperature (\(\theta\)) and water vapor mixing ratio (\(r\)) were calculated according to the formula in Stull (1988) and analyzed in this paper.

### 3. Results and discussion

#### 3.1 Description of the haze episodes based on ground-level observations

Synoptic-scale weather patterns during the entire observation period reveal that the observation site was mainly influenced by high-pressure systems and typical synoptic weather circulations in winter over the NCP (Fig. 2; Miao et al., 2017). The small high-pressure system was likely separated from the Mongolian high in the northwest (Figs. 2a–c). At 0800 BT (Beijing Time) 16, 0800 BT 19, and 0800 BT 21 December, the observation site was mainly controlled by cold advection at 850 hPa. Previous studies have proved that synoptic-scale weather patterns can influence the evolution of the atmospheric pollution process (Miao et al., 2017; Wu et al., 2017).

Figure 3 shows the concentration time series of PM\(_{2.5}\) and PM\(_{10}\) as well as the temporal variations of ground-level meteorological parameters WS, WD, \(T\), RH, and solar irradiance during the entire observation period. The shaded areas represent the three pollution episodes (S1, S2, and S3, respectively) and the one clean event.

Selection of the three pollution episodes (S1, S2, and S3) was based on the ground-level concentrations of PM\(_{2.5}\) greater than 100 μg m\(^{-3}\), and the clean period was identified with the PM\(_{2.5}\) concentration lower than 35 μg m\(^{-3}\). In addition, the sampling altitude of the large tethered balloon is shown in Fig. 3d. Figure 3a indicates that the observation experiment captured approximately five pollution periods based on ground-level PM\(_{2.5}\) (or PM\(_{10}\)) concentrations. The maximum PM\(_{2.5}\) concentration exceeded 200 μg m\(^{-3}\), and the PM\(_{10}\) concentration peaked at approximately 500 μg m\(^{-3}\) during the whole observation period. The time series of PM\(_{2.5}\) (or PM\(_{10}\)) concentration during the observation period was generally higher in the early morning and decreased to a certain extent at noon. Baumbach and Vogt (2003) reported that pollutant emissions were mainly confined beneath the surface inversion layer, which developed in the late afternoon. The highest PM\(_{10}\) concentration observed in current study was 504.4 μg m\(^{-3}\) at 0200 BT 16 December due to the blocking effects of the inversion layer. In addition, the PM\(_{10}\) concentration dropped to 154.8 μg m\(^{-3}\) at 1200 BT 16 December.

Previous studies have concluded that northwest winds can effectively remove pollution around our observation site (Zhao et al., 2013; Shi et al., 2019). However, when northwest winds were observed at the ground during this observation experiment, the PM\(_{2.5}\) concentration was still high, which was likely due to the low WS at this time. In addition, the ratio of PM\(_{2.5}\) to PM\(_{10}\) is often used to measure the proportion of coarse-particulate concentrations (Liu et al., 2015). The major sources of PM\(_{10}\) are urban fugitive dust, crustal soil, biomass burning, coal combustion, and vehicle emissions; while PM\(_{2.5}\) mainly originates from industrial emissions, secondary aerosols, coal combustion, traffic exhaust, and biomass burning (Liu et al., 2015). Figure 3a shows that PM\(_{2.5}\) concentrations presented a pattern of variation very similar to PM\(_{10}\), indicating that the increase in PM\(_{2.5}\) was mainly responsible for the increase in PM\(_{10}\). During the whole observation episode, the averaged PM\(_{2.5}\)/PM\(_{10}\) ratio was approximately 0.4; its minimum value was 0.19 (clean episode); and its maximum was 0.78 (S2 episode), indicating the enhanced role of fine-mode aerosols.

Due to the abatement policies regarding coal quality and usage, the coarse particles have been greatly reduced. This is also reflected by the high PM\(_{2.5}\)/PM\(_{10}\) ratios around the observation site. The variations of PM\(_{2.5}\)/PM\(_{10}\) ratios were also due to the pollutant sources around the site. As shown in Fig. 1b, the observation site mainly consisted of a wheat field. According to the Baoding Yearbook 2019 (Zhang, 2019), there were no direct industrial sources near the observation site, and most of the pollutants mainly came from agricultural combustion, traffic emissions, and residential sources, but there were no direct industrial sources at the time of this study.

There was a distinct inverse correlation between \(T\) and
RH. Both RH and $T$ had obvious diurnal variations. RH was nearly 80% in winter, which promoted increased moisture absorption in aerosol particles (Svenningsson et al., 1992). Solar irradiance also exhibited clear diurnal variations. At noon on each day, a clear peak value was recorded due to the large amounts of direct and scattered radiation from the sun. Moreover, compared with the peak value of solar irradiance during the high-pollution periods, the peak value was slightly higher on clean days, demonstrating that particles could reduce the solar irradiance received by the ground. In addition, the presence of the stratiform clouds on 19 and 20 December greatly decreased the solar irradiance received by the ground.

Combined with the PM$_{2.5}$ concentration variation and the observation records from the tethered balloon, Table 1 summarizes the mean values of the concentrations of the seven air pollutants (CO, SO$_2$, NO, NO$_2$, O$_3$, PM$_{2.5}$, and PM$_{10}$), WS, $T$, and RH during the whole observation period, the one clean period, and the three severe haze pollution episodes. Table 1 indicates that the average PM$_{2.5}$ concentration during the most severe haze episode was 190.6 $\mu$g m$^{-3}$ (S1), which was 12 times higher than that during the clean episode (14.8 $\mu$g m$^{-3}$). Concentrations of the various pollutants on the polluted days increased significantly; the lower WS and higher RH contributed to pollutant accumulation, and the contributions of regional transport cannot be ignored either.

Figure 4 shows the time series of the ground-level concentrations of six pollutants (NO, NO$_2$, SO$_2$, CO, O$_3$, and PM$_{2.5}$) from 15 to 21 December 2018. The NO, NO$_2$, and CO concentrations showed similar variations to the PM$_{2.5}$ concentration, followed by the SO$_2$ concentration. From 18 to 21 December 2018, the SO$_2$ concentration varied sharply within a few hours. Kim et al. (2002) reported certain effects of the NO$_x$ concentration on particle formation in air, whereby NO$_x$ notably influenced the particle homogeneous nucleation and secondary aerosol nucleation of organic compounds. Figure 4c reveals that the O$_3$ concentration was higher in the afternoon hours (1500–1600 BT) due to the strong solar radiation, corresponding to the lower PM$_{2.5}$ and NO concentration levels.

Because of the significant pathway for the formation of particulates through heterogeneous reactions (Liggio et al., 2005; Wen et al., 2018), we made the one-to-one scatter plots (Fig. 5) showing different correlation coefficients between the ground-level CO, O$_3$, NO$_x$ (NO + NO$_2$), SO$_2$, and PM$_{2.5}$ concentrations.
The correlation coefficient between PM$_{2.5}$ and CO during the entire observation period was 0.85 (Fig 5a), indicating a strong relationship between the two pollutants. Higher PM$_{2.5}$ concentrations are often observed with higher RH. O$_3$ and PM$_{2.5}$ concentrations showed an inverse correlation. When RH exceeded approximately 70%, O$_3$ concentrations were low and became insensitive to RH variation. This implies that the relationship between O$_3$ and PM$_{2.5}$ is complex and does not obey a simple linear correlation. The correlation between NO$_x$ and PM$_{2.5}$ was somewhat weak, and NO$_x$ could be correlated with O$_3$ since NO can deplete O$_3$. The correlation between SO$_2$ and PM$_{2.5}$ was the poorest, and the SO$_2$ concentrations were not high.

3.2 Vertical distributions of PM$_{2.5}$

It is relatively difficult to obtain the vertical profile of pollutants. During the entire observation period of this experiment, a total of 33 vertical profiles of pollutant and meteorological variables were obtained. Table 2 lists the weather conditions, starting time, ending time, and measurement height for all 33 profiles. These profiles were
acquired during different periods, and the vertical profiles of physical and chemical elements could be simultaneously acquired during the planetary boundary layer evolution process. To better understand the vertical PM$_{2.5}$ distribution, we calculated the dispersion ($d$) for each vertical profile; $d$ was defined as follows:

$$d = (\text{max} + \text{min})/(2 \times \text{mean}),$$  \hspace{1cm} (1)

where “max” and “min” are the maximum and minimum PM$_{2.5}$ concentrations, respectively, in each profile, and “mean” is the average PM$_{2.5}$ concentration. Three distinctly different types of vertical profiles (Types 1, 2, and 3 in Fig. 6b) were identified.

In Type-1 profiles, with $d$ ranging within 0.9–1.2, the PM$_{2.5}$ concentration decreased nearly linearly as a function of the height below approximately 600 m, such as in profile No. 11. This kind of vertical profile of PM$_{2.5}$ indicates a convective state. Type-2 profiles show a stable state, exhibiting a sharp decreasing of PM$_{2.5}$ concentration from the ground to approximately 200 m, thus characterized by substantial vertical dispersion/differences ($d > 1.2$). A typical case is represented by profile No. 22 (Fig. 6b). Type-3 profiles show increased particulates suspended aloft in the upper air, revealing a multilayer structure, and a typical vertical profile No. 1 is shown in Fig. 6b. The vertical profiles of Type 3 were characterized by increasing PM$_{2.5}$ as a function of height, and the

| WS (m s$^{-1}$) | 1.5 | 3.9 | 1.3 | 1.1 | 0.78 |
|----------------|-----|-----|-----|-----|-----|
| $T$ (°C) | 2.2 | 4.9 | −0.8 | 0.3 | 1.7 |
| RH (%) | 57.4 | 38.2 | 66.9 | 64.2 | 64.3 |
| CO (ppm) | 1.8 | 0.18 | 3.2 | 2.1 | 1.71 |
| SO$_2$ (ppb) | 17.6 | 6.6 | 13.9 | 19.5 | 28.5 |
| NO (ppb) | 66.8 | 2.4 | 67.7 | 113.5 | 116.4 |
| NO$_2$ (ppb) | 28.8 | 14.7 | 32.4 | 35.7 | 40.6 |
| O$_3$ (ppb) | 10.7 | 17.0 | 6.7 | 7.6 | 8.29 |
| PM$_{2.5}$ (μg m$^{-3}$) | 112.8 | 14.8 | 190.6 | 153.1 | 130.1 |
| PM$_{10}$ (μg m$^{-3}$) | 244.9 | 78.6 | 369.1 | 310.6 | 273.2 |

Table 1. The mean values of the seven air pollutant concentrations (CO, SO$_2$, NO, NO$_2$, O$_3$, PM$_{2.5}$, and PM$_{10}$), WS, $T$, and RH during the entire study period, the one clean episode, and the three severe haze pollution episodes (S1, S2, and S3)

**Fig. 4.** Temporal variations of ground-level concentrations of (a) NO and NO$_2$, (b) SO$_2$ and CO, and (c) O$_3$ and PM$_{2.5}$ from 15 to 21 December 2018. The shadings represent the one clean episode (yellow; Clean) and three high-pollution episodes (gray; S1, S2, and S3). (d) The detection altitude of the tethered balloon.
slopes for this type approached a positive slope, although the $d$ values were small.

### 3.2.1 Vertical profiles of Type 1: Convective state

As shown in Fig. 7a, the PM$_{2.5}$ concentration in Type-1 profiles decreased almost linearly as a function of the height below approximately 600 m (Nos. 11, 17, and 24), and all differences were smaller than 1.2, where noticeable reductions in aerosol particles were observed above approximately 600 m. These vertical profiles accounted for about half of all the profiles that we obtained in this study, and most of the Type-1 vertical profiles were typically observed in the daytime with a relatively high $\theta$ and strong turbulence activities. A well-mixed distribution of PM$_{2.5}$ below 600 m in the daytime indicates that the estimated mixing layer height (MLH) at the observation site was approximately 600 m in winter. Figure 7a shows that $\theta$ of the three vertical profiles mainly increased with height. The water vapor mixing ratio $r$ of No. 11 decreased with height uniformly and showed a high correlation (0.92) with the PM$_{2.5}$ vertical profile.

Moreover, the WS values of Nos. 11 and 17 were very similar and decreased similarly between 500 and 800 m, exhibiting opposite vertical distributions compared with vertical profile No. 24. However, a common characteristic of all WS profiles is that a pronounced WS change was observed at approximately 600 m, reflecting notable wind shear. PM$_{2.5}$ was also concentrated below this height due to high wind shear in the daytime.

Furthermore, there was little difference in $\theta$ between Nos. 11 and 17, but $r$ of No. 17 was basically higher than that of No. 11. Since the observation period of these two profiles varied little, the higher PM$_{2.5}$ concentration of No. 17 may be the result of the increase in water vapor, which could promote the growth of particulates.

**Fig. 5.** Scatter plots of PM$_{2.5}$ and (a) CO, (b) O$_3$, (c) NO$_x$, and (d) SO$_2$ observed at ground level during the whole observation period. Colors of the dots represent different RH values.
3.2.2 Vertical profiles of Type 2: Stable state

Type-2 vertical profiles were also frequently observed during the entire observation period. Figure 7b shows three typical Type-2 profiles with sharp decreases in
PM$_{2.5}$ concentration above approximately 250 m in the lower layer. Type 2 often occurred during 0900–1100 BT and 1600–1800 BT. For the period of 1600–1800 BT, with the weakening of daytime mixing, the role of local emission sources became more prominent. In this period, the surface inversion layer was not as obvious in No. 20; perhaps the radiative cooling effects from the ground were not strong. The WS of both vertical profiles did not exceed 4 m s$^{-1}$ below 500 m, and light winds also contributed to pollutant accumulation. In the winter morning, the surface inversion layer was very significant. For example, the surface inversion layer of No. 21 was quite distinct, and the inversion intensity was estimated to be approximately 0.02ºC m$^{-1}$. The dispersion conditions in the nighttime stable boundary layer were unfavorable, so pollutants became more easily concentrated within the inversion layer during this period (Baumbach and Vogt, 2003; Largeron and Staquet, 2016).

### 3.2.3 Vertical profiles of Type 3: Multilayer structure

Figure 7c shows three typical Type-3 vertical PM$_{2.5}$ profiles characterized by pollutants suspended in the upper air of the profiles. The vertical profile of PM$_{2.5}$ concentration (No. 1) below 200 m was more closely related to the winter mixing conditions in the lower ABL. The PM$_{2.5}$ concentration decreased substantially above 150 m for vertical profile No. 1. There was an obvious inversion from 600 to 900 m, and the vertical profile of PM$_{2.5}$ shows a multilayer structure. Based on examination of the solar irradiance received on the ground (Fig. 3c), there might also have been a few clouds affecting this layer at the observation site. The WS values of the three vertical profiles were similar, and the WD measurements...
above 500 m were also similar. Although there were no PM$_{2.5}$ data obtained by the tethered balloon at this time, PM$_{2.5}$ below 600 m also had a clear multilayer distribution.

### 3.3 Case study of the evolution of vertical PM$_{2.5}$ profiles

Figures 8 and 9 show the detailed diurnal evolution of PM$_{2.5}$ vertical profiles and meteorological parameters from 1200 BT 18 to 1600 BT 20 December. Here, to facilitate discussion, we defined Phase 1 from 1636 BT 18 to 1602 BT 19 December and Phase 2 from 1706 BT 19 to 1641 BT 20 December.

The vertical profiles mainly evolved from Type 1 in the daytime to Type 2 or Type 3 at nighttime. The stage of Phase 1 during 1636–2047 BT 18 December was characterized by a decreased WS from 5 to 2 m s$^{-1}$. The PM$_{2.5}$ vertical profiles evolved from Type 1 to Type 2, e.g., No. 14 (2019–2047 BT 18 December 2018). From 1046 to 1602 BT 19 December, this stage of Phase 1 was characterized by moderately higher $r$ and increased $T$. Compared with the inversion layer of No. 15 over 300–600 m, $T$ in No. 17 profile decreased nearly uniformly with height. The PM$_{2.5}$ vertical profile of No. 15 showed a clear change at approximately 200 m, indicating the presence of a mixed layer below (Stull, 1988), consistent with the bottom of the inversion.

Phase 2 was characterized by consistent northerly winds below 300 m and southerly winds above approximately 300 m (Fig. 8). From 1600 to 2000 BT 19 December, the vertical difference varied significantly, and the ground-level PM$_{2.5}$ concentration changed from 99.7 to 169.5 μg m$^{-3}$. No. 20 was a Type-2 profile measured during the nighttime.

Figure 9c displays that $r$ gradually decreased in the profile between 150 to 600 m from 1000 to 1500 BT 20 December, and the values were relatively higher mainly due to advected water vapor. In addition, $\theta$ and $r$ decreased below 200 m in their vertical profiles during 1030–1054 BT (No. 22), indicating unstable stratification. The Type-2 (No. 22) vertical profile during the morning changed to Type 1 (convective state, No. 25) in the daytime. Profile No. 27 exhibited Type-3 characteristics with a multilayer aerosol structure, and an obvious feature was that WS increased and southerly winds prevailed in the upper portion of the profile.

### 3.4 Effects of the light-wind layer and inversion layer on the vertical PM$_{2.5}$ profile

The day of 15 December 2018 was chosen as a high-haze day since the ground-level PM$_{2.5}$ concentration exceeded 250 μg m$^{-3}$. Figure 10 shows the PM$_{2.5}$, $T$, RH, and WS vertical profiles observed on that day. Here, we defined a light-wind layer as WS lower than 3 m s$^{-1}$. As shown in Fig. 10, the three vertical profiles were all char-

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**Fig. 8.** Vertical–temporal evolution of (a) WS and (b) WD retrieved by the Doppler wind lidar. Data from 2100 BT 18 to 0900 BT 19 December were unavailable due to the recalibration of the machine.
characterized by a light-wind layer below approximately 400 m. Clearly, particulates were mainly concentrated within the light-wind layer.

The profile during 1156–1243 BT 15 December 2018 (No. 1) was recognized as typical of regional transport aloft, and it also had light-wind layer characteristics (Fig. 10a). Another significant feature is the effect of the inversion layer over 600–900 m on the accumulation of pollutants. The profile during 1351–1419 BT 15 December 2018 also exhibited typical light-wind layer features, i.e., WS below 400 m did not exceed 2 m s\(^{-1}\), which was very favorable for pollutant accumulation (Fig. 10b). In addition, PM\(_{2.5}\) in the lower layer was relatively well mixed, partly because of the strong turbulence activity at

Fig. 9. Evolution of the vertical profiles of (a) PM\(_{2.5}\), (b) \(T\) and RH, and (c) \(\theta\) and \(r\) from 1200 BT 18 to 1600 BT 20 December 2018. The ground-level PM\(_{2.5}\) for this period is also shown in Fig. 9a.
noon. RH throughout the low-wind layer changed dramatically; namely, it decreased rapidly and had a high gradient. A dramatic decline in the PM$_{2.5}$ concentration between 350 and 500 m was observed, where WS increased accordingly. The pollutants during this period were also trapped by the capping inversion layer, which appeared at approximately 300 m, and the capping inversion layer depth was approximately 400 m. There were also many pollutants trapped in the inversion layer. The PM$_{2.5}$ concentration near 600 m was greater than 60 μg m$^{-3}$, and RH and temperature at this height both increased (RH and temperature are generally inversely correlated). The high PM$_{2.5}$ concentration near 600 m may have been caused by the transport of moist plumes with high pollutant concentrations.

Another PM$_{2.5}$ vertical profile was measured during the afternoon from 1544–1558 BT, as depicted in Fig. 10c. Although the maximum sampling altitude was nearly 600 m above the ground at this time, the lower portion of the sampled layer still exhibited low-winds with a high particle concentration. It is worth noting that during this winter, the NCP area temperature vertical profile from 1544–1558 BT shows that the inversion layer had developed during this period, and the depth of the surface inversion layer exceeded 200 m. The pollutants in the lower layer were clearly trapped by the surface inversion layer, but there were still some pollutants suspended above the inversion layer when the inversion layer was not developing deeply enough.

4. Conclusions

In this paper, the pollution characteristics in the North China Plain (NCP) area were analyzed, and the key factors influencing the vertical distribution of PM$_{2.5}$ were clarified through observations from the tethered balloon, Doppler wind lidar, and ground-level instruments. Due to local control of the coal combustion industry, the PM$_{2.5}$/PM$_{10}$ ratio was generally approximately 0.4 (its maximum was approximately 0.8) during the whole observation period, revealing that fine-mode aerosols dominated in the winter heating season. Ground-level observations indicate that the correlation coefficient between the PM$_{2.5}$ and CO concentrations was the highest (0.85) among the gas pollutants, and there was an inverse relationship between O$_3$ and PM$_{2.5}$ in general but this was not a simple linear relationship.

Based on the 33 vertical profiles acquired with the tethered balloon, three types of vertical profiles were classified to illuminate the vertical evolution characteristics of PM$_{2.5}$ during the three severe haze episodes. Type 1 was a convective state characterized by PM$_{2.5}$ concentrations decreasing nearly linearly as a function of the height below approximately 600 m. This type of vertical profiles were mainly observed in the daytime. Type 2 was a stable state, exhibiting a sharp decreasing trend from the ground to approximately 200 m, and this kind of vertical profiles of PM$_{2.5}$ were closely associated with the surface inversion layer inhibiting the upward dispersion
of pollutants. Type 3 demonstrated a multilayer structure with some pollutants remaining suspended aloft in the upper portions of the profiles. The diurnal variation of boundary layer structure had great influences on the vertical profiles of PM$_{2.5}$. Because of differences in ABL structures from daytime to nighttime, the vertical profiles evolved from Type 1 to Type 2 or Type 3. The vertical distributions of winds, inversion layers, and turbulence activities notably influenced the PM$_{2.5}$ vertical profiles. Both the light-wind layer and inversion layer, and weak turbulence activities contributed greatly to the accumulation of pollutants. Moreover, the PM$_{2.5}$ vertical distribution patterns were greatly affected by local surface emission sources and regional transport processes.

The above results will help to deepen the understanding of the relationships between the boundary layer structure and haze pollution. Further work will need to focus on the feedback between various pollutants and the ABL structure due to the effects of radiant energy balances.

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