Two Inversion Layers and Their Impacts on PM$_{2.5}$ Concentration over the Yangtze River Delta, China

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ABSTRACT

An integrated winter field campaign was conducted to investigate the atmospheric boundary layer structure and PM$_{2.5}$ concentration at three sites over the Yangtze River delta (YRD) in China: Shouxian (a rural area), a site in a northern suburb of Nanjing, and Dongshan (a residential area). Two temperature inversion layers and air pollution events occurred simultaneously from 30 to 31 December 2016, local time, over the YRD. It was found that the two inversion layers were related to the presence of a high pressure system, resulting in divergence in the upper boundary layer and radiative cooling near the ground at night. Dominated by agricultural and residential biomass burning, the surface emission sources from the Shouxian rural area were moderately strong. After the formation of the two inversions, the vertical distribution of PM$_{2.5}$ concentration below the upper inversion layer was uniform as a result of thorough boundary layer mixing in the earlier hours. During nighttime at the Nanjing site, air pollutant plumes from nearby elevated point sources could not easily diffuse downward/upward between the two inversion layers, which led to a distinct peak in the PM$_{2.5}$ concentration. At the Dongshan site, the emission sources were weak and the nighttime PM$_{2.5}$ concentration above 100 m was high. The surface PM$_{2.5}$ concentration gradually increased from early morning to noon, which was attributed to emissions related to the local residents. The results indicated that the vertical distribution of pollutants was affected by a combination of local emissions, vertical boundary layer structure, and horizontal and vertical transports.

1. Introduction

With the rapid development of China’s economy and the accelerated urbanization process, air pollution has become a serious problem. Air pollution can increase the incidence of human respiratory and cardiovascular diseases, which adversely affect human health. Air pollution often occurs in winter because of the substantial fuel combustion emissions from winter heating under adverse meteorological conditions. Many studies have indicated that certain meteorological conditions, such as low wind speed, high relative humidity (RH), and stable atmospheric stratification as well as the presence of temperature inversions and shallow mixing layers, are not conducive to the diffusion of pollutants (e.g., Chan et al. 2005; Z. Li et al. 2017; Wang et al. 2017; Wu et al. 2013; Zhang et al. 2017).

Previous studies indicated that temperature inversions in the low and middle troposphere occur as a result of various processes, including radiation inversion, advection inversion, subsidence inversion, and frontal inversion. Y. Zhang et al. (2009) found that two notable inversion layers occurred in the low and middle troposphere over Yichang (30.42°N, 111.18°E), China, in January of 2007 based on an analysis of intensive radiosonde observations. Li et al. (2012) found that there was a seasonal dependency of the height of temperature inversion layer based on an analysis of sounding data.
from stations near 30°N in China. Inversions in the low and middle troposphere mostly occur in autumn and winter. Occasionally, multiple inversion layers can simultaneously occur in winter. Severe air pollution occurs frequently when the temperature difference between the top and bottom of an inversion layer is large, for example, greater than 6°C (Daugherty 1960). Temperature inversions could change atmospheric dynamic structure (Nodzu et al. 2006; Cao et al. 2007), prevent vertical mixing and diffusion processes (Palmen and Newton 1969; Abdul-Wahab 2003; Johnson et al. 1999), suppress convection and the formation of clouds (Ding et al. 2016), and finally lead to the accumulation of air pollution within a shallow boundary layer (e.g., Wallace and Kanaroglou 2009; Y. Zhang et al. 2009; Petäjä et al. 2016).

Based on aircraft measurements over Beijing, Q. Zhang et al. (2009) described three types of weather conditions that have different impacts on the vertical distribution of aerosol concentrations in the atmospheric boundary layer, which are under high pressure systems, low pressure systems, and between two high pressure systems. Under the influence of the high pressure system, the vertical mixing and horizontal transport were efficient, which led to the strong dilution of aerosols. As a result, the aerosol concentration was low and did not change much with height. Between two high pressure systems, vertical mixing was strong, and the horizontal transport was moderate. In this case, the aerosol concentrations gradually decreased with altitude, with moderate surface aerosol concentrations. Associated with a low pressure system, a cold front could lead to a so-called frontal inversion. As a result, aerosol concentrations were strongly depressed in the boundary layer. Observational measurements collected by a meteorological tower in Beijing in 2003 also showed that the vertical distribution of aerosols exhibited strong temporal (seasonal and daily variations) and spatial variations under different boundary layer dynamics and weather systems (Ding et al. 2005).

Ding et al. (2013) clarified the interaction between air pollution and weather conditions with five types. Li et al. (2015) quantified the effects of the aerosol–planetary boundary layer interaction and the aerosol–cloud–radiation interaction on weather conditions. A complex interaction was found between aerosols and atmospheric boundary layers; together, they determined the air pollution concentrations. The interaction between the boundary layer and aerosols and their feedback on other meteorological factors increased the complexity of air quality investigations (Z. Li et al. 2017). Light absorbing aerosols in the atmospheric boundary layer absorbed solar radiation, and thereby produced an inversion at the top of the atmospheric boundary layer, which was often associated with serious pollution events (Li et al. 2015; Ding et al. 2016).

A variety of measurements and analysis strategies have been used to study the relation between air quality and the characteristics of the boundary layer, such as using tethered balloons (Han et al. 2018), and ceilometers (Young and Whiteman 2015; Haman et al. 2012; Di Giuseppe et al. 2012). Whiteman et al. (2014) analyzed 40 years of data to investigate the critical meteorological factors affecting daily particulate concentrations during winter in the urbanized Salt Lake Valley in Utah. Kim et al. (2002) examined the effect of different sources and formation mechanisms on the size distribution and temporal trends of ultrafine particles using near-continuous data collected for 5 months at two urban areas in Southern California. Jing et al. (2019) used the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) to analyze the emission sources of high levels of black carbon pollution in different seasons in 2016 in a northern suburb of Nanjing, China.

Located in eastern China, the Yangtze River delta (YRD) is in the subtropical monsoon climate zone. During the period of cold front inactivity in autumn and winter, atmospheric stratification is stable and near-surface wind speeds are low. Thus, condition is favorable for the accumulation of air pollutants. Consequently, this region was an area where air pollution has become severe in recent years in China (Kang et al. 2013; Quan et al. 2014; Wang et al. 2014). In this study, we analyzed an air pollution episode that occurred from 30 to 31 December 2016 [local Beijing time (BJT)] and exhibited two inversion layers at three sites: Shouxian, representative of a rural area; a northern suburb of Nanjing, which was approximately 200 km southeast of the Shouxian site; and Dongshan, representative of a residential area and located approximately 200 km southeast of the Nanjing site. Our goals are to study the weather patterns and boundary layer structure during the air pollution episode and investigate the relationship between the vertical profiles of air pollution and the two inversion layers at these sites. To address these goals, we conducted an extensive field campaign at above three sites to obtain the surface and upper-air observations.

2. Data and methods

a. Experimental sites

The observation sites in Shouxian (a rural area), a northern suburb of Nanjing (a suburban area), and Dongshan in Suzhou (a residential area) are shown in Fig. 1. The Shouxian site was far from the Shouxian
urban area and was located in a rural area surrounded by farmlands and residential houses. The Nanjing site was located at the Meteorological Observation Field of Nanjing University of Information Science and Technology, east of a highway and approximately 4 km away from the eastern industrial area. The Nanjing site was located approximately 200 km southeast of the Shouxian site. The Dongshan site was located in Dongshan Town on a peninsula in Taihu Lake, surrounded by lake on three sides. There were large farm fields with relatively flat terrain to the north. The Dongshan site was located approximately 200 km southeast of the Nanjing site. The distances among these three sites are sufficient enough to detect the movement of weather systems.

Table 1 provides a specific description of the three observation sites.

b. Field measurements

Two sounding instruments, an Anhui XLS-II tethered balloon sounding system and a Shenzhen DJI Matrice 600 drone, were used to obtain the vertical structure of meteorological variables and pollutant concentration. The XLS-II tethered balloon sounding system consisted of two major components. A helium-filled tethered balloon (volume of 5.25 m³; payload of 5 kg) acted as a carrier. The meteorological instruments were attached underneath the tethered balloon to measure wind, temperature, and humidity. The balloon ascended...
at a steady speed of 0.8 m s\(^{-1}\) until it reached a maximum height of approximately 1000 m and then descended at the same speed.

The Matrice 600 is a six-rotor flight platform used in professional applications. The multirotor aircraft was powered by six intelligent flight batteries, which could increase its endurance. The extensible structure and maximum flight weight of 15.1 kg enabled the Matrice 600 to be equipped with several devices. The drone can ascend at a maximum speed of 5 m s\(^{-1}\) and descend at a maximum speed of 3 m s\(^{-1}\). During the experiments, the drone ascended at a steady speed of 1 m s\(^{-1}\) until it reached a maximum height of 500 m and then descended at the same speed.

A Thermo Fisher Scientific, Inc., model THERMO PDR-1500 portable aerosol particulate matter detector was used to measure the PM\(_{2.5}\) mass concentration (Han et al. 2018). The PDR-1500 is a highly sensitive photometric monitor covering a wide measurement range from 0.001 mg m\(^{-3}\) (=1 \(\mu\)g m\(^{-3}\)) to 400 mg m\(^{-3}\). The PDR-1500 includes a temperature and RH sensor to mitigate the positive bias with elevated ambient RH. Additionally, the flow control was truly volumetric. To ensure the quality and reliability of the observations, all of the deployed instruments were examined thoroughly and calibrated before the field campaign. Han et al. (2018) used the PDR-1500 to investigate the rapid formation and evolution of an extremely severe and persistent haze and fog episode over central eastern China. Pant et al. (2017) used the PDR-1500 to measure personal exposure to PM\(_{2.5}\) for healthy volunteers in Delhi, India.

The PDR-1500 was attached to the tethered balloon platform at the Shouxian and Nanjing sites and measured temperature, pressure, RH, wind speed, wind direction, and PM\(_{2.5}\) concentrations in the vertical direction in the boundary layer. The sampling interval of the meteorological variables was 1 s, and that of the PDR-1500 was 10 s. The PDR-1500 was attached to the drone (height limit of 500 m) at the Dongshan site, and the meteorological data were collected by the tethered balloon.

Launches were scheduled every 3 h between 0200 and 2300 BJT from the ground to the maximum height of 1 km or 500 m. During the observation period, scheduled flights were suspended a few times because of extreme weather, such as precipitation and strong winds, and were resumed once the condition improved. The real-time hourly surface PM\(_{2.5}\) concentration data were continuously recorded by the Chinese Ministry of Environmental Protection and can be publicly accessed from China’s National Environmental Monitoring Center (2016). The national air quality monitoring network was continuously operated and maintained by the Department of the Environment for each city in China (Zhang and Cao 2015). At each monitoring site, the real-time mass concentrations of PM\(_{2.5}\) were measured using micro-oscillating balance methods and/or the \(\beta\) absorption method from commercial instruments. The hourly concentrations of all pollutants were the averages of the hourly data from all monitoring sites in the city. Instrumental operation, maintenance, data assurance, and quality control were properly conducted according to the most recent revisions of the China Environmental Protection Standards (Zhang and Cao 2015). The data used in this study were obtained from the Internet during the period from 2000 BJT 29 to 1700 BJT 31 December 2016. Automatic weather station data were used to measure the surface meteorological parameters hourly, including wind speed and direction, temperature, and RH.

c. Reanalysis data

The mass divergence was calculated using the 0.75° × 0.75° European Centre for Medium-Range Weather Forecasts interim reanalysis (ERA-Interim). ERA-Interim is a global atmospheric reanalysis starting from 1979 and is continuously updated to present in real time. The data

<table>
<thead>
<tr>
<th>Obs point</th>
<th>Location (N, E)</th>
<th>Land cover</th>
<th>Intensity of emission source (kg day(^{-1}) km(^{-2}))</th>
<th>Surrounding environment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shouxian</td>
<td>32.5, 116.7</td>
<td>Rural</td>
<td>4.7</td>
<td>Surrounded by farmland and residential houses; the surrounding area is wide, and the terrain is relatively flat, often with straw burning of farmland and residential sources</td>
</tr>
<tr>
<td>Nanjing</td>
<td>32.0, 118.4</td>
<td>Suburban</td>
<td>7.9</td>
<td>Close to a highway to the east, a small hill to the south, residential area and farmland to the north and west, and approximately 4 km away from the eastern industrial area</td>
</tr>
<tr>
<td>Dongshan</td>
<td>31.0, 120.4</td>
<td>Residential</td>
<td>3.0</td>
<td>Located on a peninsula at Taihu Lake, surrounded by lake on three sides, with the surrounding area being wide and open; to the north there are large fields of farmland with relatively flat terrain</td>
</tr>
</tbody>
</table>
assimilation system includes a four-dimensional variational analysis with a 12-h analysis window. The spatial resolution of the dataset is approximately 80 km on 60 vertical levels from the surface up to 0.1 hPa. The interval of the assimilation analysis is 6 h and it is cycled every 12 h. For example, when the data collection starts at 0000 UTC, followed by 0600, 1200, and 1800 UTC each day, an error correction is performed at 1200 UTC.

d. Experimental methods

The following equation is used to calculate the inversion intensity:

\[
\text{Intensity} [\text{°C} (100 \text{ m})^{-1}] = \frac{(T_2 - T_1) \times 100}{H},
\]

where \( T_1 (\text{°C}) \) is the temperature at the bottom of the inversion layer, \( T_2 (\text{°C}) \) is the temperature at the top of the inversion layer, and \( H (\text{m}) \) is the thickness of the inversion layer.

To investigate the sources of pollution and their trajectories, we used the HYSPLIT model from the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory. The model boundary conditions were obtained from the National Centers for Environmental Prediction Global Data Assimilation System (GDAS) with horizontal resolution of 1°. In this study, we calculated 36-h backward trajectories for each time at several different heights at each observation station. The objective of using a backward trajectory model was to determine the origins of the air masses and to establish source–receptor relationships (Jing et al. 2019).

3. Results and discussion

a. Weather and air pollution overview

According to the average and anomaly of geopotential height at 500 hPa in December 2016 (Liu and Ma 2017), the East Asian trough and the Baikal ridge were weaker than normal. Furthermore, the Eurasian mid–high latitudes were characterized by a straight zonal circulation in late December, which was not favorable for the southward flow of cold air and led to higher temperatures than the average in most parts of China. The wintertime southern branch trough (the semipermanent trough generated by the subtropical southern branch westerlies over the Bay of Bengal, south of the Tibetan Plateau) was weak, which was not in favor of warm and humid airflow from the Bay of Bengal and the Indian Ocean ahead of the trough. The subtropical high gradually extended westward and was slightly stronger than normal. Therefore, cold air advection was weak. The YRD was affected by high pressure

(Figs. 2a, b) from 30 to 31 December. During this period, the high pressure center moved from west to east (from Shouxian through Nanjing to Dongshan and finally over the ocean), with weak surface winds and low cloud fractions dominating the entire area. Therefore, radiation cooling at night was strong, leading to the formation of an inversion and the accumulation of pollutants (Baumbach and Vogt 2003).

The accumulation of PM\(_{2.5}\) first occurred in Shouxian from 1400 to 1800 BJT 30 December 2016, followed by Nanjing, where it began to increase approximately 2 h later, and it finally increased in Dongshan as well (Fig. 3). The PM\(_{2.5}\) in Shouxian and Nanjing rapidly accumulated and stabilized at a high concentration, while the PM\(_{2.5}\) in Dongshan showed a longer period of accumulation (the shaded area in Fig. 3). The emission sources of Dongshan were weak in comparison with those of Shouxian and Nanjing. The intensity of the primary PM\(_{2.5}\) emission source was 3.0 kg day\(^{-1}\) km\(^{-2}\) in Dongshan, 4.7 kg day\(^{-1}\) km\(^{-2}\) in Shouxian, and 7.9 kg day\(^{-1}\) km\(^{-2}\) in Nanjing (Table 1). Shouxian was dominated by biomass burning associated with agricultural production and residential sources. Smoke from kitchen chimneys in the morning and evening led to relatively high surface pollutant concentrations, and the PM\(_{2.5}\) concentration in Dongshan showed a longer period of accumulation (approximately 150 μg m\(^{-3}\)). Emissions at the northern suburb of Nanjing were dominated by elevated industrial and ground sources. Industrial emission and transportation produced surface pollution. The concentration of PM\(_{2.5}\) in Nanjing, stabilizing at approximately 100 μg m\(^{-3}\), was not as high as that in Shouxian, because the site in Nanjing was located farther away from the pollution sources. Dongshan is close to a lake and has fewer anthropogenic sources; therefore, the PM\(_{2.5}\) in Dongshan was considerably lower than that in Shouxian and Nanjing.

b. Formation and evolution of the two inversion layers at the three sites

At 1100 BJT 30 December 2016, an upper-level inversion first appeared at the Shouxian site at 800 m height (Fig. 4a). At 2000 BJT 30 December, a second inversion appeared near the surface around ~90 m. The two inversion layers that formed at the Shouxian site within the boundary layer lasted until 2300 BJT 30 December. The intensity of the surface inversion increased and the intensity of the upper inversion decreased between 2000 and 2300 BJT 30 December (as shown in Table 2). The upper inversion was not clear at 0200 BJT 31 December, and it merged with the surface inversion, which had an intensity of approximately 0.84°C (100 m\(^{-1}\)). The inversion occurred only below approximately 520 m (Table 2). The wind direction

from the surface to the upper layers rotated clockwise from southerly to westerly with height in this process. The change in wind direction with height was consistent with an Ekman spiral solution (Holton 2004). The clockwise wind rotation also indicated that the large-scale weather system was relatively stable so that the vertical distribution of meteorological fields was mainly determined by the local characteristics of the boundary layer. The radiation inversion near the surface gradually disappeared after sunrise, and the wind direction throughout the whole layer became southerly.

Although the trends in the boundary layer structure at the Nanjing and Shouxian sites were similar, the upper inversion at the Nanjing site occurred 3 h later than that at the Shouxian site. At 1400 BJT 30 December 2016, the inversion occurred at a height above 800 m over the
Nanjing site (Fig. 4b), and then decreased steadily in height afterward. The surface inversion at the Nanjing site also occurred later than that at the Shouxian site, and the intensity of the surface inversion at 2000 BJT 30 December at the Nanjing site was the weakest (Table 2). The two inversion layers lasted until 0800 BJT 31 December, and the intensity of the upper inversion increased during this period. The inversion layers at the Dongshan site occurred last, and the surface inversion layer occurred below 230 m while the upper inversion occurred from 600 to 700 m at 0200 BJT 31 December (Table 2). Before sunrise at 0500 BJT, the base of the upper inversion gradually decreased to 340 m, followed by a further decrease to 210–490 m at 0800 BJT (Table 2). After 1100 BJT, the inversion layers gradually disappeared. The analysis of the vertical observational data presented above indicates that the two inversion layers at the three sites showed a clear temporal evolution and exhibited spatial similarities from northwest to southeast.

To investigate the cause of the spatial and temporal variability of the two inversion layers, the time series of divergence over the layers from 700 to 1000 hPa were analyzed using ERA-Interim data obtained every 6 h from 0800 BJT 30 December to 2000 BJT 31 December 2016 for the Shouxian, Nanjing, and Dongshan sites (Fig. 5). From the upper layer (700 hPa) to the surface, the divergence field exhibited a general trend from convergence to divergence. In general, divergence was observed below 500 m and subsidence was observed below 975 hPa, and the divergence center moved with time as the high pressure system moved across the YRD region. Weak winds were observed near the surface.
As shown in the synoptic chart (Fig. 2), the three sites were under the influence of a high pressure system with low wind speed at 0800 BJT 30 December. By 2000 BJT 31 December, the high pressure region had moved eastward and the wind direction at all sites was southeasterly. The three sites were all in an area of divergence and subsidence. In addition, the center of the divergence and subsidence region moved from northwest to southeast from 2000 BJT 30 to 0200 BJT 31 December (Fig. 5). The 1000-hPa divergence field...
was stronger than the 975-hPa divergence field (Fig. 6). The Nanjing site was in the divergence (subsidence) center from 2000 BJT 30 December to 1100 BJT 31 December (Fig. 5b), corresponding to a strong upper inversion layer (Table 2), and the divergence was smaller in Dongshan than in Shouxian and Nanjing.

The vertical atmospheric structure in Fig. 4 indicates that spatially the two inversion layers occurred from the northwest to the southeast (from Shouxian to Nanjing and then to Dongshan) and that temporally the two inversion layers occurred from 2000 BJT 30 to 0800 BJT 31 December. A surface inversion began to form as a result of the effect of longwave radiation loss at the surface at night. The divergence center in the lower boundary layer first appeared at Shouxian and also moved from northwest to southeast, which consequently enhanced the surface inversion at the three sites (Fig. 5). Figure 5b shows that the Nanjing site was under the influence of divergence (subsidence) for the longest time, and the vertical observations showed that a two-inversion-layer structure lasted for 9 h at Nanjing (Fig. 4b). The intensity of divergence was weak at Dongshan (Fig. 5c). As mentioned above, Dongshan is located on a peninsula in Taihu Lake where the presence of a relatively warm water surface resulted in a weak surface-based inversion with delayed onset relative to the other sites. The specific humidity below 975 hPa at the Dongshan site was higher than at the Shouxian and Nanjing sites and exceeded 4 g kg$^{-1}$. As the high pressure center moved eastward, the divergence center also moved eastward.

**TABLE 2.** The intensity [$^\circ$C (100 m)$^{-1}$] and height (m) of temperature inversion from 30 to 31 Dec 2016 at three sites: Shouxian, Nanjing, and Dongshan. An em dash indicates no temperature inversion, and a slash denotes no data.

<table>
<thead>
<tr>
<th>Time</th>
<th>Shouxian</th>
<th>Nanjing</th>
<th>Dongshan</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Surface inversion</td>
<td>Upper inversion</td>
<td>Surface inversion</td>
</tr>
<tr>
<td>2000 BJT</td>
<td>1.13</td>
<td>0.29</td>
<td>0.44</td>
</tr>
<tr>
<td>Intensity</td>
<td>90</td>
<td>530–670</td>
<td>160</td>
</tr>
<tr>
<td>Height</td>
<td>2007</td>
<td>0.12</td>
<td>2.07</td>
</tr>
<tr>
<td>2300 BJT</td>
<td>150</td>
<td>430–670</td>
<td>110</td>
</tr>
<tr>
<td>0200 BJT</td>
<td>0.84</td>
<td>—</td>
<td>0.85</td>
</tr>
<tr>
<td>Intensity</td>
<td>520</td>
<td>—</td>
<td>200</td>
</tr>
<tr>
<td>Height</td>
<td>1.28</td>
<td>—</td>
<td>2.08</td>
</tr>
<tr>
<td>0500 BJT</td>
<td>440</td>
<td>—</td>
<td>40–220</td>
</tr>
<tr>
<td>0800 BJT</td>
<td>1.63</td>
<td>—</td>
<td>1.36</td>
</tr>
<tr>
<td>Intensity</td>
<td>370</td>
<td>—</td>
<td>60–200</td>
</tr>
<tr>
<td>Height</td>
<td>/</td>
<td>/</td>
<td>310–430</td>
</tr>
</tbody>
</table>

**FIG. 5.** Time–height cross sections of divergence (shaded; $10^{-3}$ s$^{-1}$) and specific humidity (contours; g kg$^{-1}$) at (a) Shouxian, (b) Nanjing, and (c) Dongshan from 0800 BJT 30 to 2000 BJT 31 Dec 2016. Negative values of divergence represent convergence.
and the two-inversion-layer structure moved from northwest to southeast.

c. Characteristics and causes of the vertical distribution of pollutant concentrations

It is well known that the distribution of PM$_{2.5}$ concentration is determined not only by local emission sources and the inversion characteristics but also by vertical and horizontal transports in the boundary layer. When the two-inversions structure appeared at 2000 BJT 30 December 2016 at the Shouxian site, the vertical distribution of PM$_{2.5}$ from the surface to the bottom of the upper inversion layer (~400 m) was uniform (~180 $\mu$g m$^{-2}$) (Fig. 4a) and in agreement with the surface concentration of PM$_{2.5}$ obtained by ground-based observations (Fig. 3a). The high PM$_{2.5}$ concentration in the residual layer at night originated from the remaining PM$_{2.5}$ in the mixing layer in the earlier hours (Z. Li et al. 2017). When the height of the upper inversion layer dropped from 530 to 430 m from 2000 to 2300 BJT, the maximum PM$_{2.5}$ concentrations occurred at approximately 400 m. At 0200 BJT 31 December, the high pressure center moved eastward and the divergence center at the Shouxian site also disappeared (Fig. 5a). The upper inversion layer disappeared, and the concentration of PM$_{2.5}$ in the upper boundary layer decreased significantly. However, PM$_{2.5}$ continuously accumulated in the surface inversion layer until the inversion disappeared, and the PM$_{2.5}$ was distributed over the mixing layer in the daytime. Figures 7a and 7b show the 36-h backward trajectories ending at Shouxian at 2000 and 2300 BJT 30 December (the times of formation and disappearance of the two inversion layers), respectively. All of the air masses ending at the Shouxian site originated within 200–300 km from the south and moved slowly, with swinging paths around the site during this period.
FIG. 7. The simulations of 36-h backward trajectories arriving at the three sites at the times of formation and disappearance of their two layers inversion: Shouxian at (a) 2000 and (b) 2300 BJT 30 Dec 2016, Nanjing at (c) 2300 BJT 30 and (d) 0800 BJT 31 Dec 2016, and Dongshan at (e) 0200 and (f) 0500 BJT 31 Dec 2016. The lines connected by circles, squares, and triangles represent trajectories with end points at 400, 300, and 200 m, respectively. The markers are drawn every 12 h. The lower part of each panel shows the time series of a vertical cross section of the backward trajectories.
During the last 12 h to the destination Shouxian the vertical movements of the trajectories were near the surface, indicating the high PM$_{2.5}$ concentration in the residual layer at night may originate from the boundary layer of the surrounding areas.

During the period in which the two inversion layers appeared at the Nanjing site (Fig. 4b), high PM$_{2.5}$ concentrations were mainly confined below 600 m. The PM$_{2.5}$ concentration increased significantly at night after 2000 BJT 30 December (Fig. 4b), and an obvious peak concentration of PM$_{2.5}$ appeared at approximately 400 m. The height of this PM$_{2.5}$ concentration peak decreased with time. The PM$_{2.5}$ concentration increased continuously, resulting in a maximum of 828 μg m$^{-3}$ at 0200 BJT 31 December, which was about 4 times the concentration of the surface PM$_{2.5}$ at 1400 BJT 30 December. At midnight on 31 December, the PM$_{2.5}$ concentration peak occurred in the inversion layer below 200 m. After sunrise, as a result of solar radiation heating at the surface, the surface inversion layer gradually disappeared, and the PM$_{2.5}$ was mixed upward to a height of ~300 m by noon, when the inversion disappeared completely.

As compared with the vertical distribution of the PM$_{2.5}$ concentration at the Nanjing site, the PM$_{2.5}$ concentration at the Shouxian site was considerably higher in the lower boundary layer but was lower in the upper boundary layer (Fig. 4). This difference in the vertical distribution of PM$_{2.5}$ concentration could be related to the difference between the emission sources of the two sites. The emission source of the Shouxian rural area was mainly from the burning of agricultural residuals in open fields and biomass burning for domestic-living needs (Fig. 1b), and there is no major industrial source nearby. The surface emission sources of the Nanjing site came from both transportation and residential sources (Fig. 1c). The industrial zone was approximately 4 km to the east from the site, and there were many major industrial sources. As shown in Fig. 4b, the peak concentration of PM$_{2.5}$ increased from night to early morning, and this period corresponded to the easterly wind. The high PM$_{2.5}$ concentration over the Nanjing site was likely related to pollution from elevated point sources, that is, emissions emitted through chimneys from nearby factories. The heights of these chimneys are between 100 and 200 m. The rising hot plume was able to directly enter the residual layer above the stable boundary layer at night, and pollutant emissions could not easily diffuse downward or upward between the two inversion layers, which resulted in the PM$_{2.5}$ concentration peak occurring at a height between 200 and 400 m. The 36-h backward trajectories analysis of the Nanjing site revealed that the air masses originated from 200 to 300 km southeast of the site during this period (Figs. 7c,d), showing a very slow and local movement. Figure 8 presents a schematic of the pollution derived from a chimney and the diffusion between two inversions in a residual layer of the boundary layer at night. When the wind direction near the surface became westerly at 0500 BJT 31 December, the peak concentration of PM$_{2.5}$ began to decrease greatly.

The PM$_{2.5}$ concentration at the Dongshan site was much lower than that at the Nanjing and Shouxian sites (Fig. 4c). The Dongshan site was located on relatively flat terrain on a peninsula of Taihu surrounded by lakes.
on three sides (Fig. 1d). The local emission sources of the Dongshan site were weak and included only domestic sources from local residents. When the two inversion layers occurred at 0200 BJT 31 December, the PM$_{2.5}$ concentration gradually increased above 100 m and reached a maximum at a height of approximately 250 m. The PM$_{2.5}$ concentration was homogeneously distributed above 250 m, with values of about 80 $\mu$g m$^{-3}$. At 0500 BJT, a high concentration was maintained above 250 m, whereas the PM$_{2.5}$ concentration decreased near the surface. In comparison with the backward trajectories at Shouxian and Nanjing, the source of the upper (above 200 m) high-pollution air masses (Figs. 7e,f) at the Dongshan site might come from the west of the YRD via medium- to long-range transport, and the air mass descended during the period of the two inversion layers between 0200 and 0500 BJT 31 December 2016. Trajectories initialized at other times in the night during this period also showed the same descending motion. In the morning and before noon on 31 December, the PM$_{2.5}$ accumulated below 100 m, with concentrations close to 200 $\mu$g m$^{-3}$ (shown in Fig. 4c). This accumulation may be attributed to pollutants discharged by local residents. Above 100 m, the vertical distribution of PM$_{2.5}$ concentrations was relatively uniform and maintained its concentrations at night because of a weak inversion located at approximately 100–200 m.

4. Summary and conclusions

An integrated field campaign was conducted to investigate the boundary layer structure at three sites over the YRD in China, with a focus on the spatial and temporal variability of two inversion layers and their impact on the vertical distribution of PM$_{2.5}$. During the period from 30 to 31 December 2016, Shouxian, Nanjing, and Dongshan were in the same synoptic background condition, under the influence of a high pressure system. Obvious divergence and subsidence occurred in the low and middle troposphere within this high pressure region, but the location and intensity of the divergence and subsidence were different at the three sites. During this period, two inversion layers appeared in the lowest 1000 m above ground in Shouxian, Nanjing, and Dongshan, with the upper inversion layer related to the subsidence and movement of the high pressure system and the surface-based inversion layer related to radiative cooling at night.

Three different characteristics in the vertical variability of PM$_{2.5}$ concentrations were observed in the boundary layer below 1000 m at the Shouxian, Nanjing, and Dongshan sites. At the Shouxian site, a relatively uniform PM$_{2.5}$ concentration was observed from the surface to the upper inversion layer, with a decrease of PM$_{2.5}$ concentration aloft. The emission sources in the Shouxian rural area were relatively strong and were dominated by agricultural and residential biomass burning. The concentration of PM$_{2.5}$ in the northern suburb of the Nanjing site had a distinct peak above the surface layer (100–400 m), which was related to major industrial sources on the eastern side of the site. The concentration of PM$_{2.5}$ at the Dongshan site was significantly lower than at the other two sites. The nighttime PM$_{2.5}$ concentration peak was high above 100 m, and the concentration of surface pollution gradually increased from the early morning to local noon attributed to the local residents. The emission sources of the Dongshan site were minimal and only from local residents. Backward trajectories showed that PM$_{2.5}$ in the upper parts of the boundary layer originated from medium- to long-range transport from west of the YRD.

This study highlighted that the vertical distribution of air pollutants was determined by a combination of local emissions, vertical boundary layer structure, and external transport. This finding suggests that simultaneous observations of the vertical variability of air pollutants and meteorological conditions are critical when attempting to control air pollution emissions.

Although the factors affecting pollution distribution were complex, detailed knowledge of the characteristics of the boundary layer, such as the height and intensity of the inversion layer, wind, and divergence, could be used to improve air quality forecasts. Model output, such as the ERA-Interim reanalysis, could help to detect better the unique boundary layer structure in a given region to support analysis of the observed episodes and the impact of the boundary layer structure on the vertical distribution of air pollutants.

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REFERENCES


Chan, C., X. Xu, Y. Li, K. Wong, G. Ding, L. Chan, and X. Cheng, 2005: Characteristics of vertical profiles and sources of PM$_{2.5}$,