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Spatial oscillation of the particle pollution in eastern China during winter: Implications for regional air quality and climate

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HIGHLIGHTS

- Two distinct pollution processes exist in fringe areas and the interior of eastern China during winter.
- Spatial oscillation of particle pollutants is responsible for the abrupt air pollution events.
- $PM_{2.5}$ in the central part of northern China can accumulate to >100 μg/m^3 within 1–2 days.

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ABSTRACT

We provided a large-scale observational insight into spatial variations of the particle pollution in eastern China during winter based on recently extended air quality monitoring networks. Severe particle pollution with $PM_{2.5}$ >150 μg/m^3 prevailed in most areas of eastern China during December 2015, when red alert of haze pollution was released in many places. It was found that two distinct pollution processes existed in eastern China during winter. In the fringe areas such as Beijing and Shanghai in eastern China, most of air pollution events were characterized by abrupt peak values and short duration. By comparison, particle pollution in the interior exhibited obvious accumulation and decline processes with much higher $PM_{2.5}$ concentration. Regional observations in ground networks show notable spatial oscillation of particle pollutants in eastern China, which is the main driver of the abrupt particle pollution in fringe areas. Despite common alternation of northerly and southerly airflows within planetary boundary layer (PBL), particle pollution in central part of eastern China was under the influence of regional accumulated pollutants due to lack of persistent strong winds. In addition, spatial oscillation of particle pollutants weakened with low PBL (<400–600 m). Our results demonstrate that spatial variations of particle pollutants in the central part of northern China play a significant role in regulating air quality in eastern China.

1. Introduction

Particulate matter (PM) is a complex mixture of small particles and liquid droplets, which originates from emissions of natural process and human activities such as sea salt, dust, fire smoke, acidic salts (sulfates, nitrates, etc.) and organic matter. With the rapid economic development during the last decades, large increase in anthropogenic emissions has caused serious particle pollution in several densely populated regions of the world (Van Donkelaar et al., 2010). Epidemiological studies have shown robust correlations between levels of fine particles ($PM_{2.5}$, particular matter with aerodynamic diameter equal to or less than 2.5 μm) and adverse public health effects (Pope et al., 2002). On the other hand, these fine particles can scatter and absorb sunlight,

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which is the main cause of visibility degradation and hazy days (Han et al., 2014; Zhang et al., 2012). Moreover, air pollution can modify cloud properties and precipitation (Guo et al., 2016a; Wang et al., 2015).

With the release of real-time monitoring information of primary pollutants such as $PM_{2.5}$ and PM_{10}, sulfur dioxide (SO_2), nitrogen dioxide (NO_2) and Ozone (O_3) since 2013, fine particle pollution in China has caused wide concerns due to its high concentration and extensive influence area. Satellite observations show that widespread haze plumes usually cover over eastern China (Guo et al., 2011, 2016b; Tao et al., 2012, 2013). In particular, extreme haze events with record-breaking particle pollution occurred repeatedly in many megacities in recent years (Wang et al., 2014; Yang et al., 2015). During December of 2015, Beijing issued the first and second ever red alert of haze pollution, which predicted that air pollution with $PM_{2.5}$ >150 μg/m^3 could last for more than 3 days.

There have been numerous studies concerning formation mechanism of the extreme particle pollution in China (Gao et al., 2015; Li et al., 2016; Petäjä et al., 2016; Yang et al., 2015; Zheng et al., 2015). Accumulation and transformation of secondary aerosols were considered as the main cause of the high-level $PM_{2.5}$ in megacities such as Beijing (Guo et al., 2014; Wang et al., 2014). Aerosol feedback on atmospheric boundary layer can make air pollution particularly severe in stagnant weather conditions (Gao et al., 2015; Petäjä et al., 2016). Some studies suggested that the extreme particle pollution resulted from the combined effects of secondary production, regional transport, and meteorological conditions (Yang et al., 2015; Zheng et al., 2015). However, previous studies are mostly based on observations in single or several ground sites, which cannot reflect regional variations of the particle pollution. So far, it remains unclear how the particle pollutants change over space and time in extreme pollution events, especially their influence on air quality in megacities.

The environmental agency of China has established nearly 1500 national ground-based monitoring sites by 2015, which offers an unprecedented opportunity to examine variations of the particle pollution in regional scale. In this study, we provided a large-scale observational insight into formation process of the particle pollution over eastern China using integrated observations from satellite and ground stations network. General information concerning ground monitoring networks, satellite observations, meteorological data as well as geographical conditions was presented in

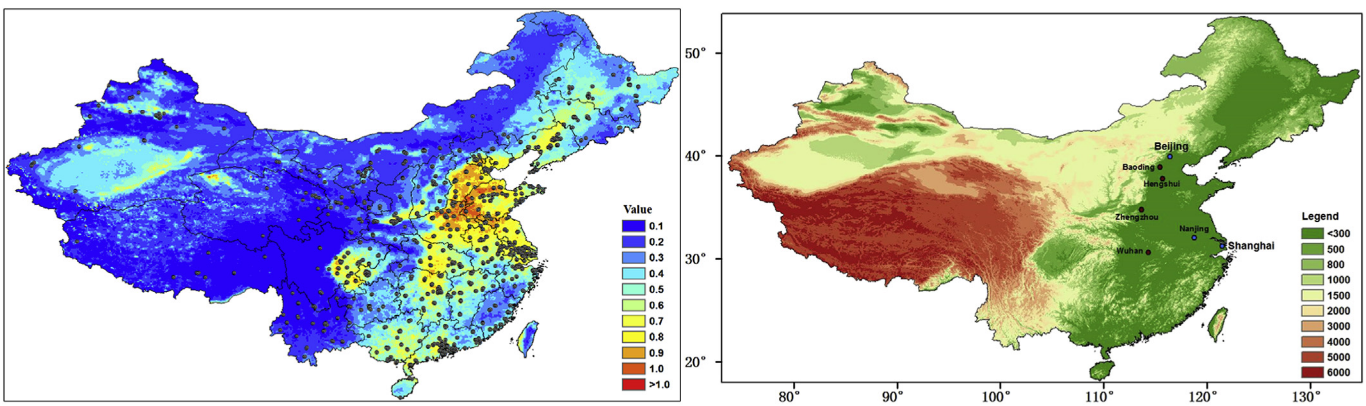

Fig. 1. a) Annual average value of MODIS Deep Blue AOD in 2015 with national $PM_{2.5}$ monitoring sites in China; b) Terrain of the main land areas of China. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

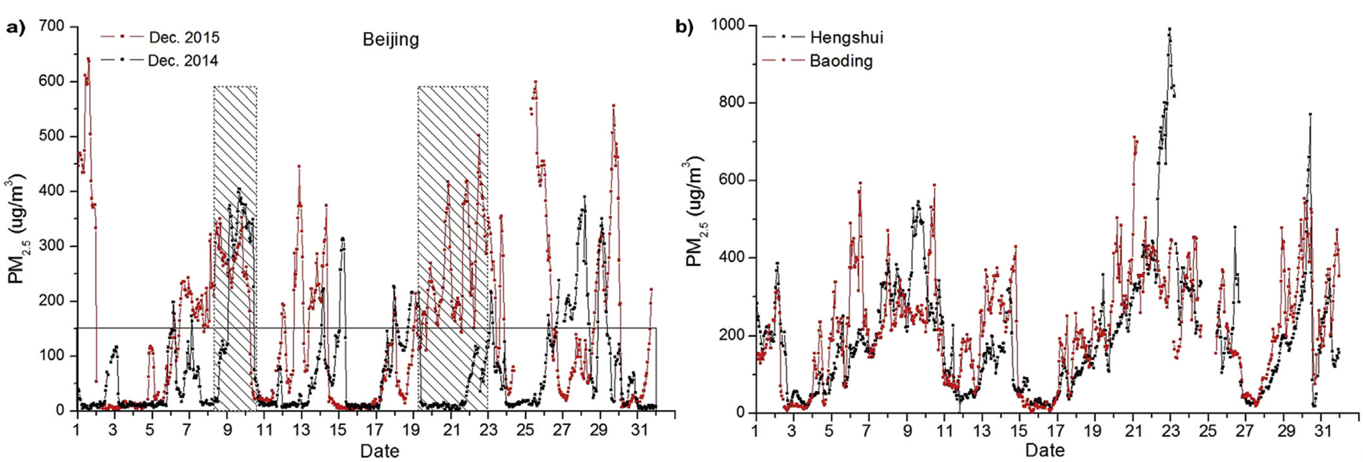

Fig. 2. a) Hourly concentration of $PM_{2.5}$ in Beijing during the December of 2014 and 2015, the filled rectangles denote the two red alert periods of haze pollution; b) Hourly concentration of $PM_{2.5}$ in Baoding and Hengshui during December 2015. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

section 2. We analyzed regional characteristics and change processes of the fine particles in several heavy air pollution episodes in eastern China in section 3.1. The role of regional transport on particle pollution in megacities was evaluated specially in section 3.2. As a focus of this paper, we discussed implications of spatial oscillation of particle pollutants on regional air quality in section 3.3. Finally, section 4 gave a summary and some remarkable conclusions.

2. Data and methods

2.1. Ground measurements

The China Environmental Monitoring Center (CEMC) publishes hourly concentration of PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and carbon monoxide (CO) in major cities since 2013 (http://106.37.208.233:20035/). As shown in Fig. 1a, the number of national air quality monitoring sites in China nearly reaches 1500 by 2015. Although

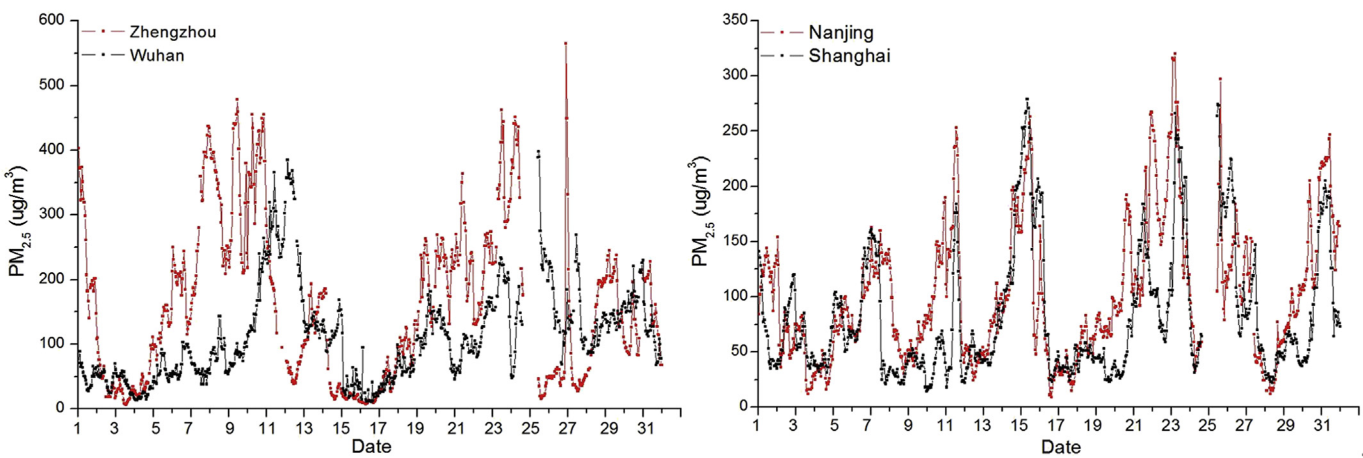

Fig. 3. Hourly concentration of $PM_{2.5}$ in Zhengzhou, Wuhan, Nanjing, and Shanghai during December 2015.

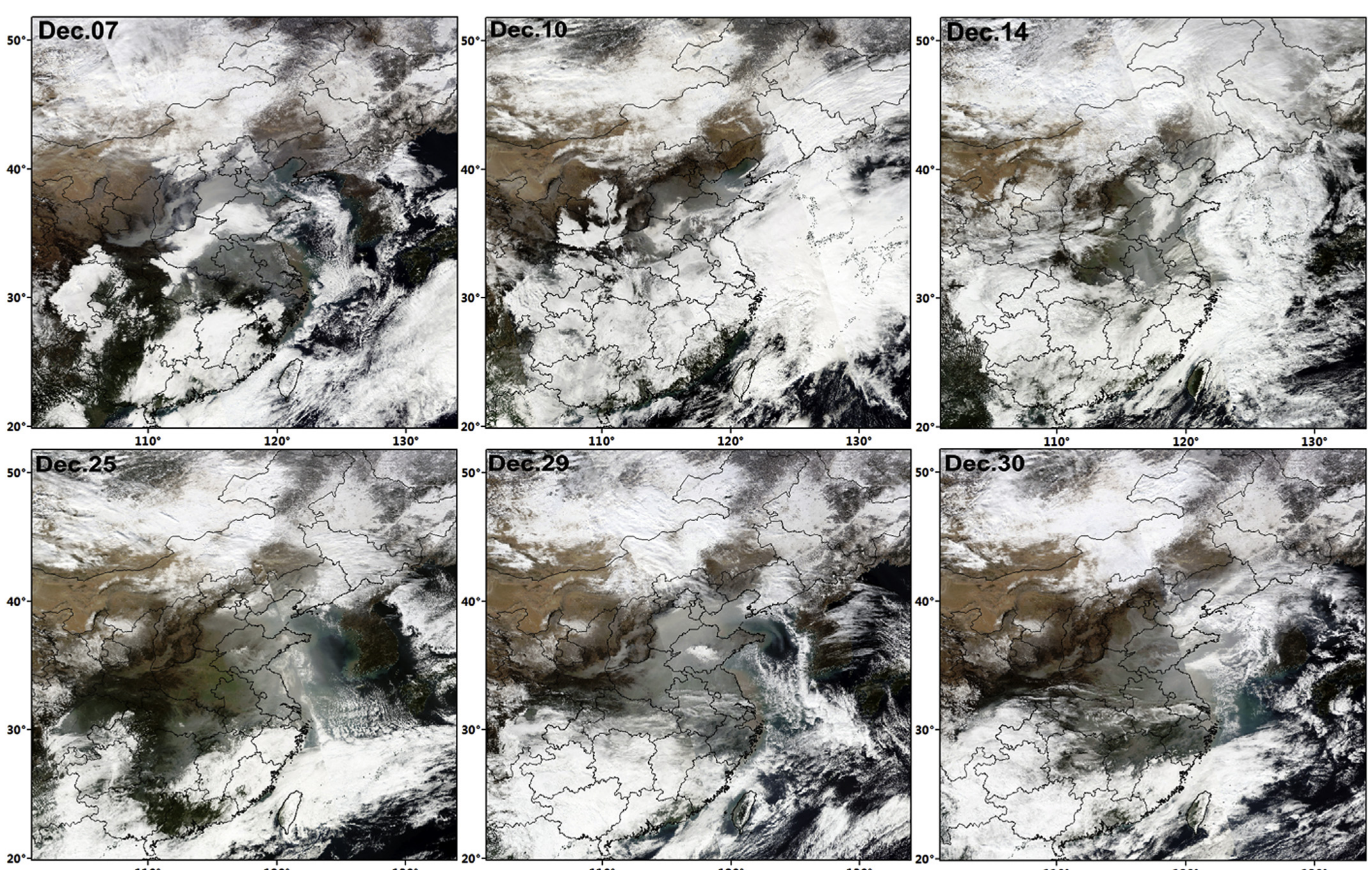

Fig. 4. NPP VIIRS true color images of regional haze pollution over eastern China during December 2015.

the PM concentrations can be measured by instruments with different methods, uncertainties of ground measurements in all the national sites are within requirements of the CEMC (http://kjs.mep.gov.cn/hjbhbz/bzwb/dqhjbh/jcgfffbz/index_2.shtml). Hourly concentration of $PM_{2.5}$ in the network was used to analyze formation process of the severe pollution in eastern China during December 2015. Considering that monitoring sites are distributed in various areas of one city, we use average value of $PM_{2.5}$ in urban region to examine its variations driven by local emissions and regional transport.

According to the evaluation standard of air quality of China in 2012 (http://kjs.mep.gov.cn/hjbhbz/bzwb/dqhjbh/index_4.shtml), real-time concentration of NO_2, SO_2, O_3, and CO as well as 24-h average concentration of $PM_{2.5}$ and PM_{10} are converted into Air Quality Index (AQI), the maximum of which is reported. The EPA of China divides the AQI into 6 grades: 0–50 (good), 51–100 (moderate), 101–150 (unhealthy for sensitive groups), 151–200 (unhealthy), 201–300 (very unhealthy), >300 (hazardous). For $PM_{2.5}$, the corresponding concentration to AQI grade is <35, 36–75, 76–115, 116–150, 151–250, and >250 $\mu g/m^3$, respectively. Here we refer to the grades of AQI when consider the levels of $PM_{2.5}$.

2.2. Geographical conditions and meteorological data

Terrain of the main land areas of China was shown to favor analysis of spatial variations of particle pollutants (Fig. 1). Annul mean values of satellite aerosol optical depth indicate that heavy particle pollution is mainly concentrated in the North China Plain (NCP). During winter, dry and clean northwestern airflows prevail in eastern China, with intermittent disturbance of moist southerly air masses within Planetary Boundary Layer (PBL) (Tao et al., 2012). Megacities such as Beijing, Nanjing and Shanghai are located at edge areas of NCP and the Yangtze River Delta (YRD) (Fig. 1b), where transport of particle pollutants is from the interior areas with few emissions upstream or downstream. By contrast, cities in the middle part of polluted regions are vulnerable to surrounding emissions. To examine regional transport of particle pollutants, we classified location of the cities into two types: fringe areas and interior areas.

Daily wind fields from the National Centers for Environmental Prediction (NCEP) reanalysis data were used to analyze meteorological conditions during the pollution periods (Kalnay et al., 1996). Height of PBL from the NCEP FNL (Final) Operational Global Analysis data in 1° by 1° grids was chosen to evaluate of the vertical diffusion of atmospheric pollutants.

2.3. Satellite observation

The Visible Infrared imaging Radiometer Suite (VIIRS) aboard NPP satellite since 2011 measures spectral radiation reflected and emitted from the Earth-atmospheric system with 22 bands in 0.412–12.05 μm (Jackson et al., 2013). The wide swath of ~3000 km enables VIIRS to provide global coverage every day.

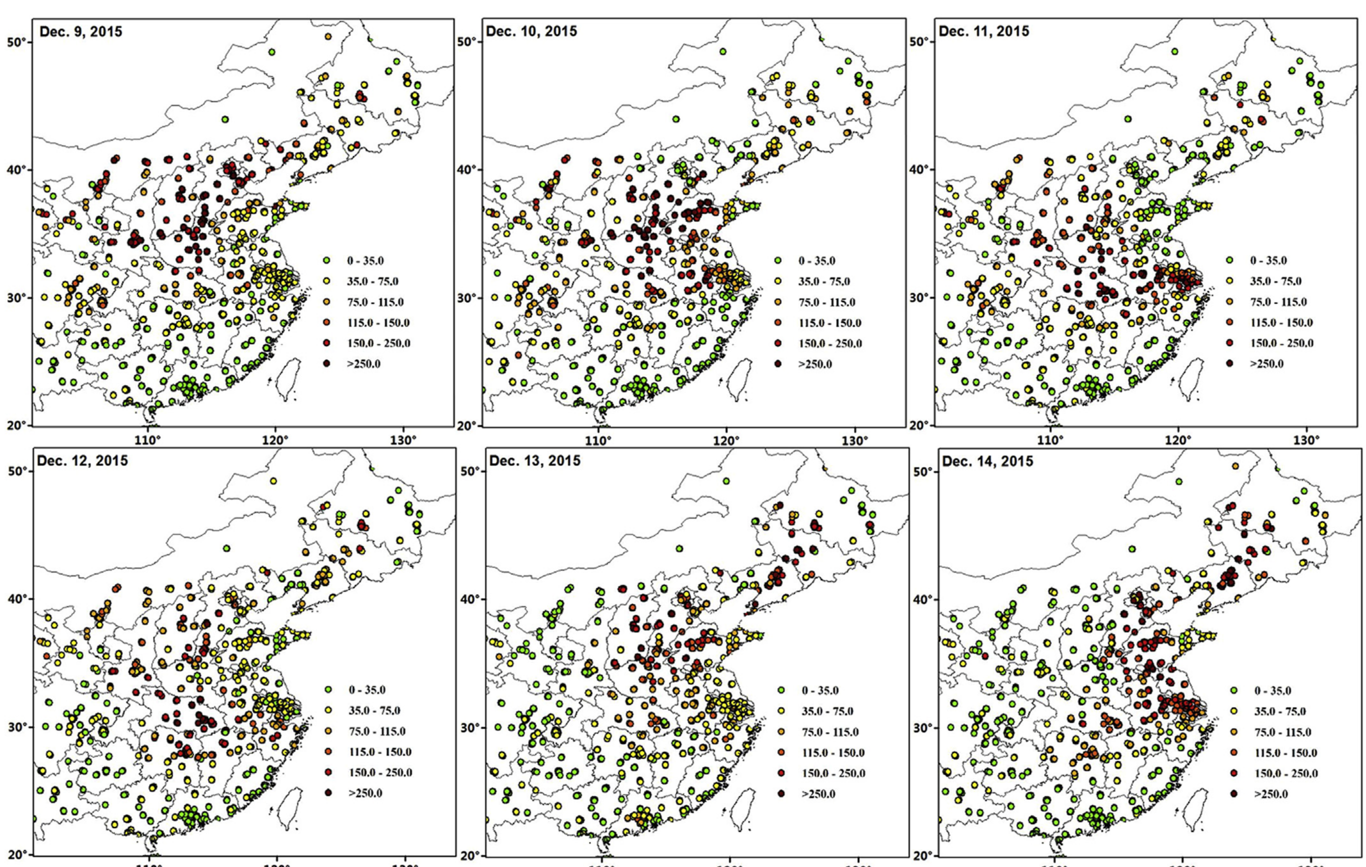

Fig. 5. a. Hourly concentration of $PM_{2.5}$ at 14:00 local time in eastern China during December 9–14, 2015. b. Height of PBL with wind fields at 925 hPa at 14:00 local time during December 9–14, 2015.

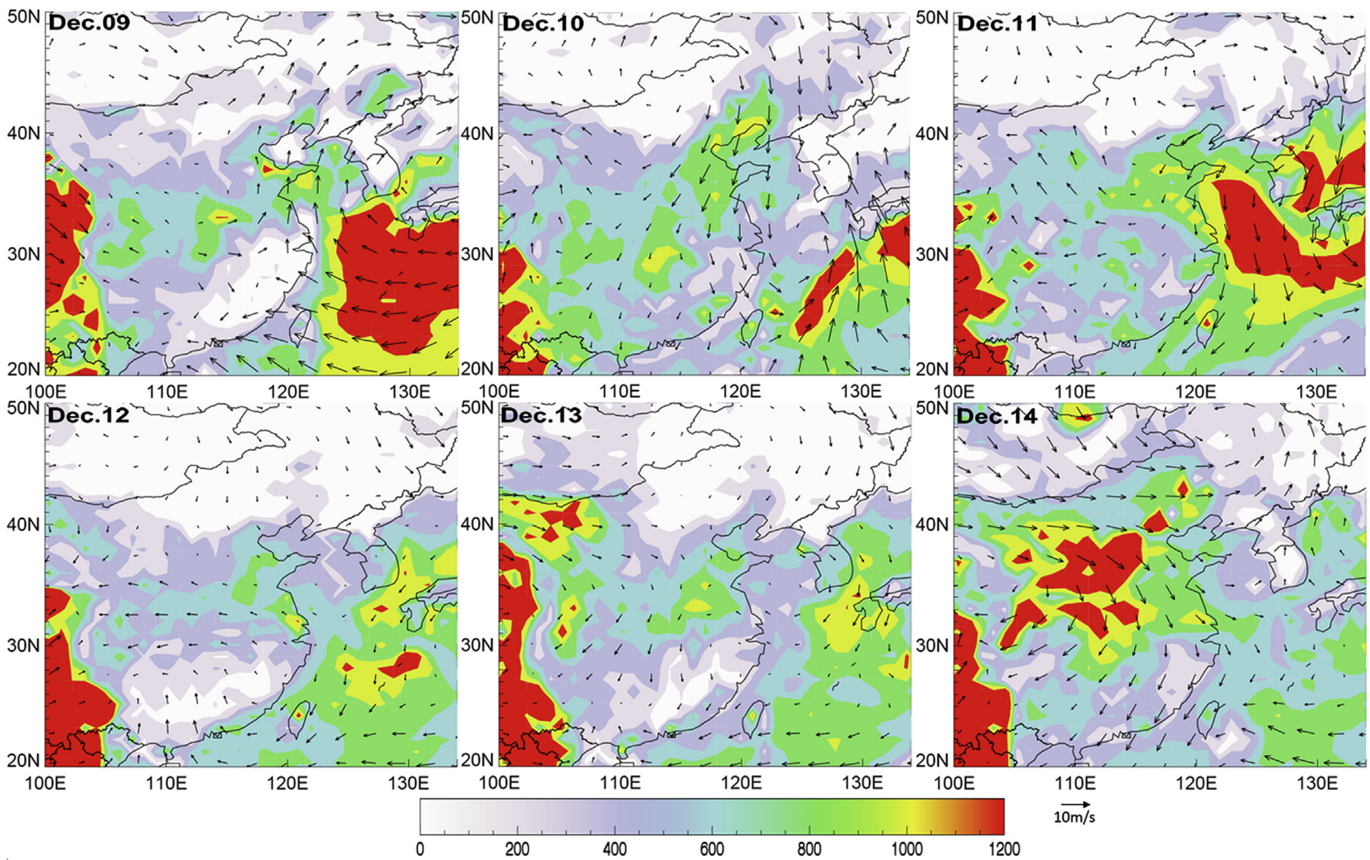

Fig. 5. (*continued*).

There are three types of bands in VIIRS: 5 imagery bands, 16 moderate resolution bands, and 1 day-night band. Here we use true color images composed of moderate resolution (750 m) bands to have a direct view of the regional air pollution over eastern China.

3. Results and discussion

3.1. Severe particle pollution in eastern China during winter

As shown in Fig. 2, durative and serious particle pollution occurred in northern China during December 2015. In the megacity of Beijing, heavy air pollution with $PM_{2.5} > 150\ \mu g/m^3$ both lasted for more than 3 days during December 6–10 and December 19–24, when red alert of haze pollution was released. Meanwhile, sudden peak values with hourly $PM_{2.5} > 500\ \mu g/m^3$ appeared on December 1, 25, and 29, but duration of these extreme pollution events was mostly within one day. It is worth noting that such abrupt pollution process is common in Beijing during winter. However, so far, early warning of serious haze pollution and temporary control in anthropogenic emissions is implemented in Beijing only when predicted $PM_{2.5}$ concentration exceeded 150 $\mu g/m^3$ for more than 3 days. Compared with the particle pollution in December 2014, $PM_{2.5}$ concentration in 2015 was much higher with more pollution episodes.

Change processes of particle pollutants in Baoding and Hengshui exhibited obvious difference from that in Beijing (Fig. 2b). Despite some abrupt variations, two durative pollution events around one week generally dominated air quality in Baoding and Hengshui during December 2015. Similar contrast in variations of $PM_{2.5}$ concentration existed between megacities in central China and the YRD (Fig. 3). Variations of $PM_{2.5}$ concentration in Zhengzhou and Wuhan were predominated by durative pollution events as that in Baoding and Hengshui. There were notable intervals between the pollution processes in Zhengzhou and Wuhan, indicating that regional transport exited. Although the duration of the air pollution events in Nanjing and Shanghai was longer than in Beijing, most of them finished within two days with abrupt peak values.

Although serious particle pollution was general among the megacities in eastern China, variations of $PM_{2.5}$ concentration exhibited distinct characteristics in difference regions. There were two typical pollution processes in eastern China during winter. Particle pollution was characterized by abrupt peak values and short duration (1–2 days) in areas such as Beijing and Shanghai, which are located near fringes of eastern China. By contrast, durative particle pollution was predominant in the interior of eastern China with both slow accumulation and decrease processes. Moreover, particle pollution in the interior was much heavier than that in fringe areas. Fig. 4 shows that widespread haze plumes existed over eastern China during December 2015 with large spatial variations, which can be associated with the abrupt peak values of $PM_{2.5}$ concentration.

3.2. Spatial variations of the extreme particle pollution

To have an observational view of spatial variations of particle pollutants in eastern China, hourly $PM_{2.5}$ concentration in ground networks was analyzed (regional distribution of $PM_{2.5}$ can be seen in supplementary material). Fig. 5a shows $PM_{2.5}$ concentration at 14:00 local time during December 9–14, 2015. Particle pollutants accumulated in the western part of NCP with $PM_{2.5}$ exceeding 250 $\mu g/m^3$ on December 9, which can be caused by slow southerly airflows under stagnant weather conditions (Fig. 5b). When northern winds prevailed within PBL on December 10, particle pollutants in Beijing were blown southward with a sudden decease in $PM_{2.5}$ concentration. Northern winds near surface was not strong enough to blow the pollutants away from eastern China, but transported the particle pollutants to central part of NCP. The continuing northeasterly winds blew particle pollutants to the YRD and central China on December 11. Then easterly air masses along the YRD transported the particle pollutants to central China on December 12. Meanwhile, $PM_{2.5}$ began to accumulate in northern China. Accumulation of particle pollutants in northern China was enhanced by weak winds on December 13 with $PM_{2.5}$ >150 $\mu g/m^3$. During December 14, strong northwesterly airflows blew the particle pollutants to the eastern part of NCP and central China. The regional particle pollution did not clear away until the northwestern winds persisted for another two days.

More serious particle pollution occurred in eastern China during December 20–25, 2015 (Fig. 6a). Regional haze pollution with $PM_{2.5}$ >250 $\mu g/m^3$ hovered in northern China, the YRD, and central China. The height of PBL during December 21–24 ranged below 400–600 m (Fig. 6b), which was much lower than in December 9–14. Severe particle pollution was concentrated in the central part of northern China during all this episode. The sudden peak value of $PM_{2.5}$ in Hengshui nearly reached 1000 $\mu g/m^3$ on the night of December 22 (Fig. 2), which can be mostly caused by superposition of surrounding accumulated pollutants. Different from large spatial variations in $PM_{2.5}$ in December 9–14, notable changes in particle concentration mainly existed in Beijing area, the YRD, and central China. Although northwesterly winds prevailed on December 24 and 25, their directions were different in eastern China. Particle pollutants in Beijing area, central China, and the YRD were blown away on December 24, but returned the next day. Strong northwestern airflows transported particle pollutants in the central part of northern China eastward on December 25.

Another regional air pollution event occurred in eastern China during December 26–31, 2015 (Fig. 7a). As in December 9–14, $PM_{2.5}$ concentration exhibited large spatial variations with attitudes of PBL at similar levels (Fig. 7b). Heavy particle pollution mainly existed in the northeastern part of NCP with westerly winds on December 26. When strong northeastern winds prevailed on December 27, $PM_{2.5}$ concentration decreased to below 35 $\mu g/m^3$ in most areas of the NCP. Fine particles began to accumulate in the NCP under the stagnant weather conditions on December 28, and was further aggravated by southwesterly airflows the next day. Northwestern winds blew the particle pollutants around Beijing area southeastward on December 30, and there was an abrupt large increase in $PM_{2.5}$ in Baoding, Hengshui, and Nanjing (Figs. 2 and 3). Southerly winds prevented the particle pollution in the NCP from diffusion on December 31, and moist northeasterly air masses could enhance particle pollution in the YRD.

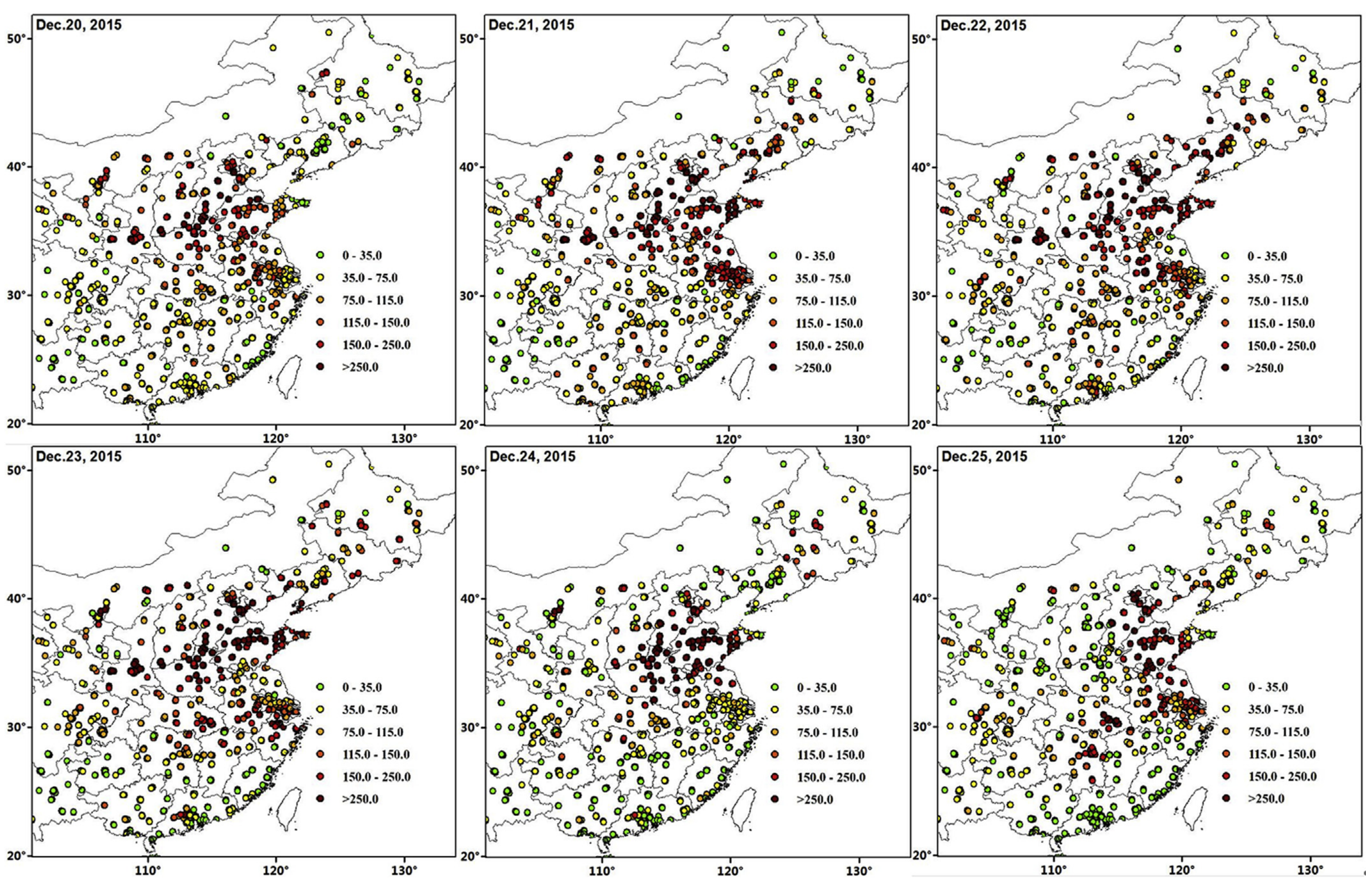

Fig. 6. a. Hourly concentration of $PM_{2.5}$ at 14:00 local time in eastern China during December 20–25, 2015. b. Height of PBL with wind fields at 925 hPa at 14:00 local time during December 20–25, 2015.

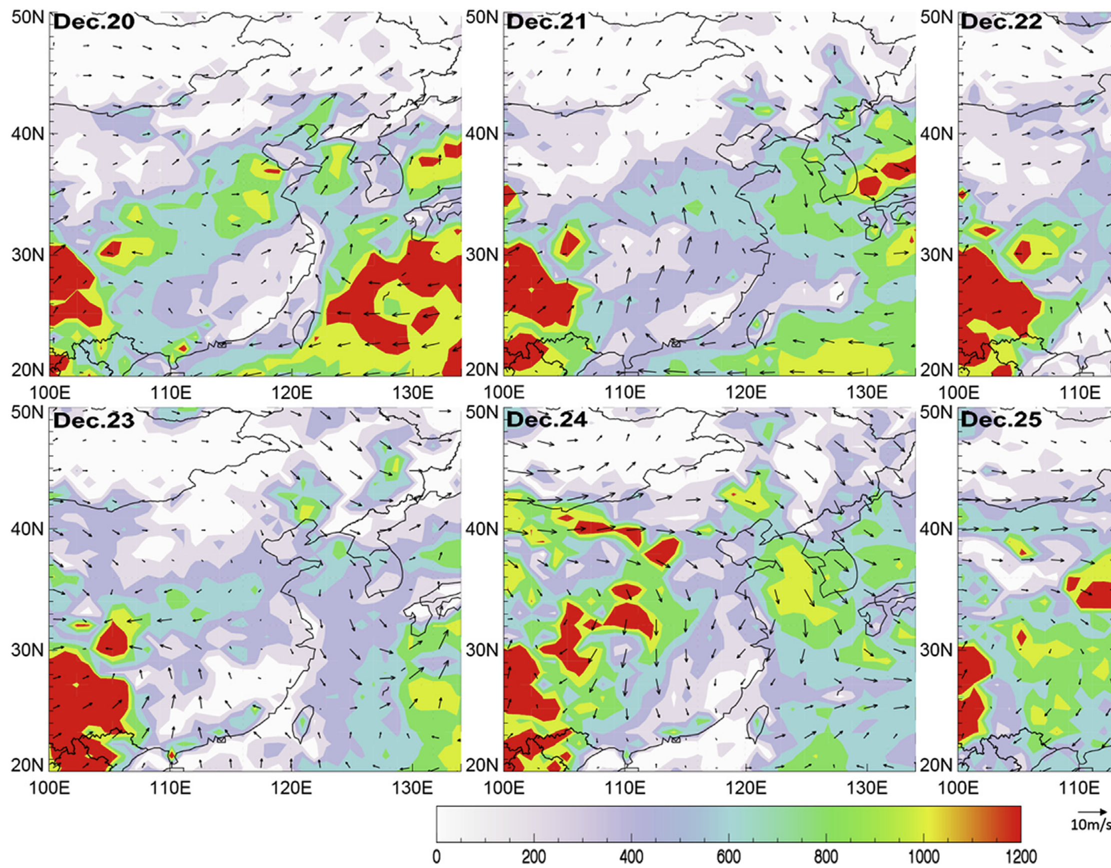

Fig. 6. (*continued*).

Spatial variations of the particle pollutants in eastern China exhibited an obvious oscillation phenomenon, which is the main driver of the abrupt peak values of $PM_{2.5}$ near the fringe areas. As one of the most polluted areas in the world, fine particles and their precursors have a relatively high background concentration in northern China due to the large amount of anthropogenic emissions (Lu et al., 2011). When atmospheric circulation weakens, southern or northern airflows transport the accumulated air pollutants in northern China to fringe areas rather than blow them away. Since there are few anthropogenic emissions upwind of the fringe regions such as Beijing area in the NCP, clean winds from northwestern arid areas and moist airflows from southern urban/industrial regions can cause large decrease and increase in particle concentration. By contrast, particle pollution in the central part is continuously under the control of local accumulation and regional transport, which has a generally slow increase and decrease process without persistent strong winds. Fig. 8 shows the basic principle of spatial oscillation of particle pollutants in eastern China.

3.3. Formation process of regional particle pollution in eastern China

Diurnal variations of $PM_{2.5}$ concentration after a thorough cleaning process on December 16 were examined to evaluate daily amount of the fine particles in eastern China (Fig. 9). Gradual increase in $PM_{2.5}$ in Baoding, Hengshui, Wuhan, Zhengzhou and Nanjing during December 17–19 indicates a regional accumulation process (Figs. 2 and 3). $PM_{2.5}$ concentration ranged from 36 to 75 $\mu g/m^3$ on the morning of December 17, and exceeded 75 $\mu g/m^3$ in some industrial cities. Diurnal variations in the height of PBL led to a large decrease in $PM_{2.5}$ concentration in the late afternoon by convection of particle pollutants (Tang et al., 2016). Similar variations occurred on December 18 with continued accumulation of fine particles. $PM_{2.5}$ concentration in major regions of northern China exceeded 150 $\mu g/m^3$ on the morning of December 19, and still maintained at high level at 17:00 local time. Although not very quantitative, the comparison indicates that accumulation of $PM_{2.5}$ in northern China can easily exceed 100 $\mu g/m^3$ within 1–2 days in stagnant weather conditions.

It is worth noting that high values of $PM_{2.5}$ concentration were mainly concentrated in the central part of northern China during the regional accumulation process (Fig. 9), indicating that anthropogenic emissions from this region play a significant role in regulating regional air quality. Since persistent strong winds are usually absent within PBL due to weakening of atmospheric circulation in northern China during winter, spatial oscillation of such high-level particle pollutants can lead to abrupt pollution events in megacities such as Beijing and Shanghai in fringe areas. Considering regional accumulation of particle pollutants in northern China, substantial reduction in anthropogenic emission in regional scales is needed to prevent the heavy particle pollution in winter. In addition, optimal emission control measures should be considered in regional scales when improving local air quality.

Different from previous studies that emphasized predominant contribution of local accumulation in severe urban haze pollution (Guo et al., 2014), regional transport can be predominant in abrupt extreme pollution (Li et al., 2013; Tao et al., 2014; Wang et al.,

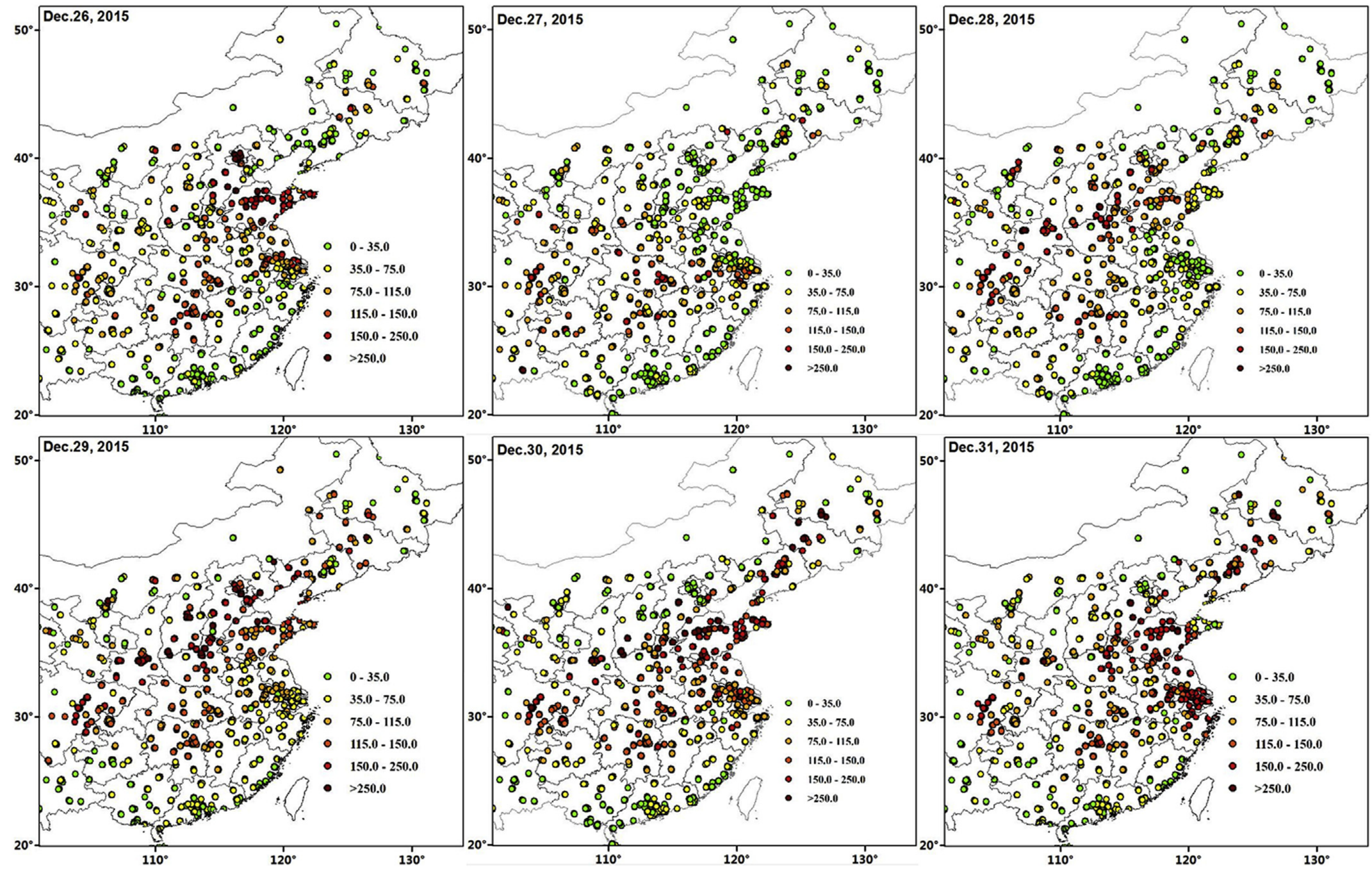

Fig. 7. a. Hourly concentration of $PM_{2.5}$ at 14:00 local time in eastern China during December 26–31, 2015. b. Height of PBL with wind fields at 925 hPa at 14:00 local time during December 26–31, 2015.

2014). Variations in atmospheric circulation play a driving effect in the general distribution and concentration of $PM_{2.5}$ besides anthropogenic emissions (Qu et al., 2015; Tao et al., 2016; Zhu et al., 2012). Unlike the prevailing strong northwestern winds above PBL (Tao et al., 2012), alternation of southerly and northerly airflows within PBL is common over northern China in winter. Chemical source apportionment was usually used to evaluate contribution of surrounding regions in megacities such as Beijing (Sun et al., 2015; Zheng et al., 2015). Spatial oscillation of particle pollutants results in repeated overlapping and mixing of different emissions, which can mislead source analysis based on ground measurements.

Recently studies suggested that aerosol radiative feedback on PBL can enhance particle pollution near surface in durative stagnant weather conditions (Gao et al., 2015; Petäjä et al., 2016). Comparisons of the particle pollution events during December 2015 show that severe accumulation of regional particle pollutants is associated with low PBL (<400–600 m) in most areas of northern China (Fig. 6). Spatial oscillation of particle pollutants can be constrained by altitudes of PBL. Considering that evolution of PBL can be affected by several factors such as temperature, cloud amount, wind speed and direction, more studies are needed to quantify the interactions between PBL and particle pollutants.

4. Conclusions

We investigated characteristics and formation process of the extreme particle pollution in eastern China by examining spatial variations of $PM_{2.5}$ concentration in ground air quality monitoring networks during December 2015. $PM_{2.5}$ concentration in eastern China exhibited distinct change processes, with abrupt and short particle pollution in fringe areas, and accumulated and durative pollution events in the central part. The intensive observations in ground networks show obvious spatial oscillation of the particle pollutants in eastern China, which is driven by repeated alternation of airflows from different directions. Since persist strong winds were usually absent within PBL in eastern China during winter, particle pollutants tend to hover round rather than be blown away. While spatial oscillation can lead to abrupt variations of $PM_{2.5}$ concentration in fringe areas such as Beijing and Shanghai, particle pollutants in the interior were frequently under influence of regional transport and accumulation with much heavier pollution.

Diurnal variations of particle pollution after a thorough cleaning process show that $PM_{2.5}$ in the central part of northern China can accumulate to >100 $\mu g/m^3$ within 1–2 days under stagnant weather conditions. The intensive anthropogenic emissions in this region can play a significant contribution in regulating air quality of surrounding areas. Spatial fluctuation of particle pollutants can be constrained when PBL is low (<400–600 m), leading to durative severe air pollution. Spatial pattern of the $PM_{2.5}$ in eastern China indicates that it is necessary to quantify the contribution in regional scale to improve air quality of certain region in an effective way.

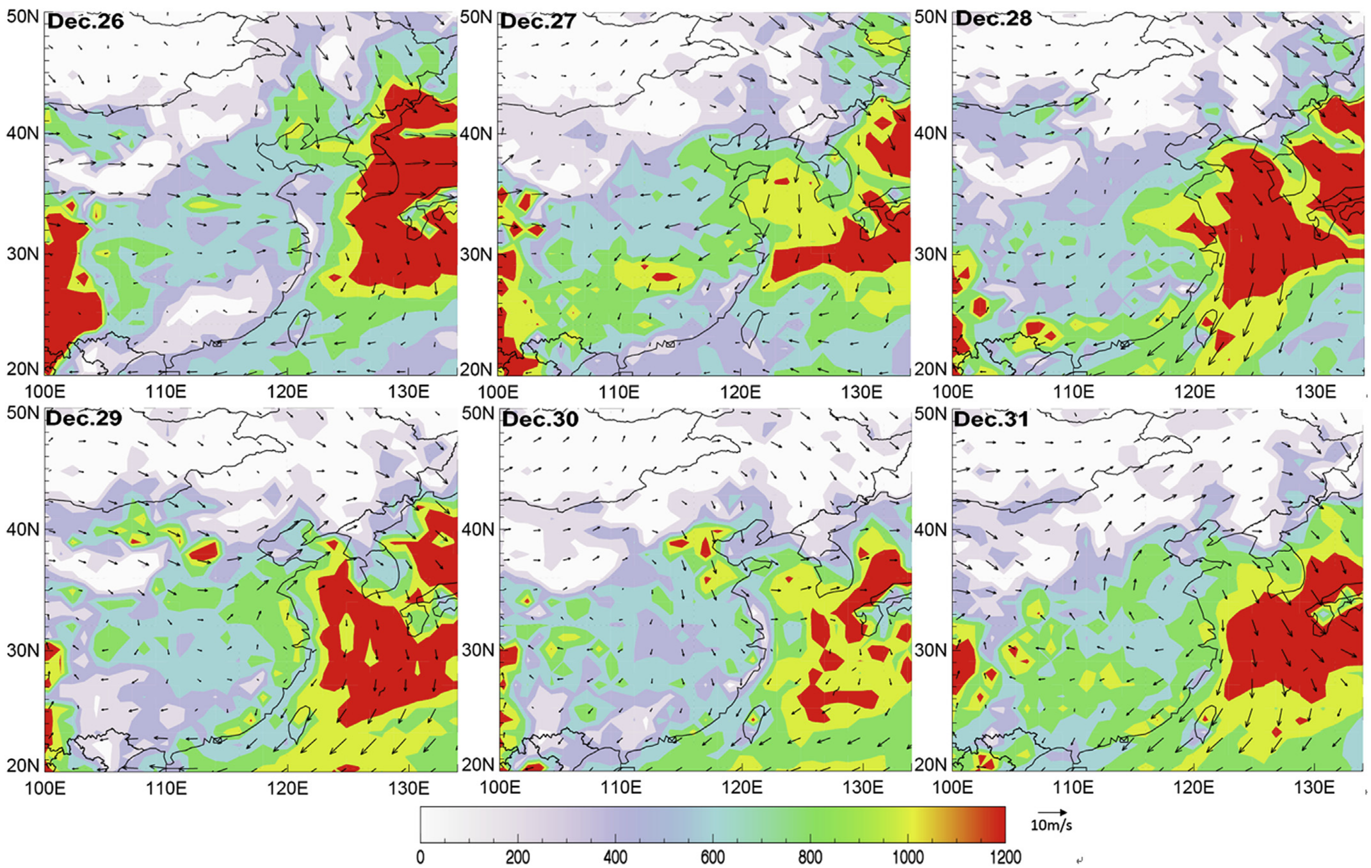

Fig. 7. (*continued*).

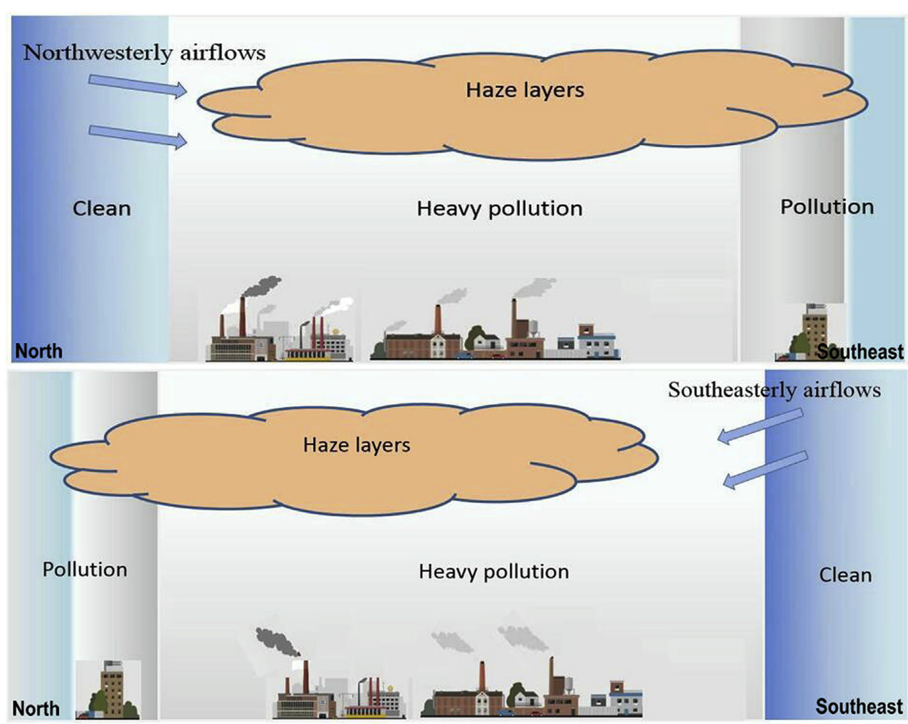

Fig. 8. Schematic figure of the spatial oscillation phenomenon of particle pollutants in eastern China. The two regions at the ends denote megacities in fringe areas of eastern China, the middle region denotes the interior of northern China, where dense urban/industrial emission sources are located; the blue color region means clean days, the gray color region represents heavy air pollution with high $PM_{2.5}$ concentration. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

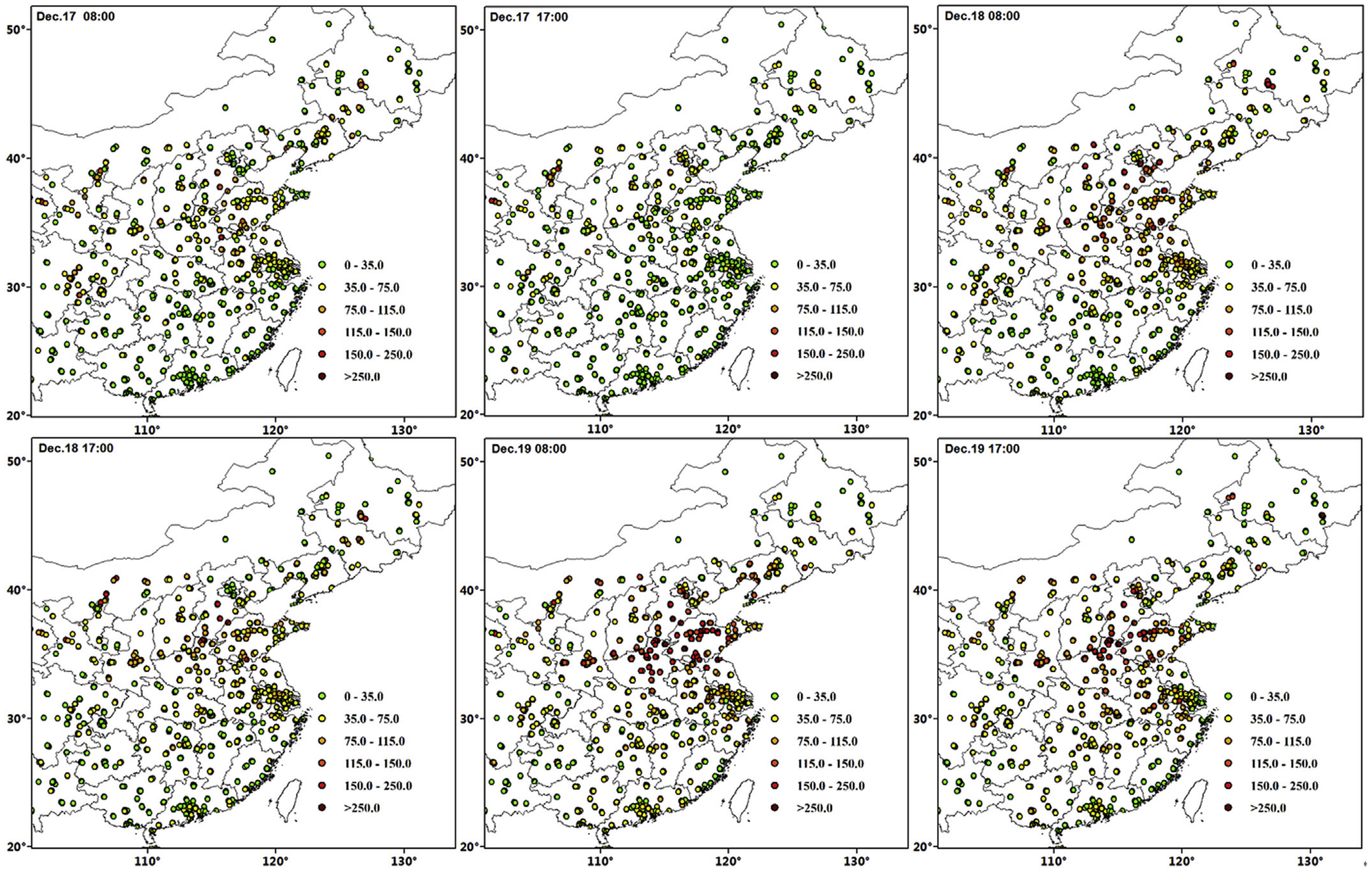

Fig. 9. Hourly concentration of $PM_{2.5}$ at local time of 08:00 and 17:00 in eastern China during December 17–19, 2015.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2016.08.049.

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