Feedback effects of boundary-layer meteorological factors on cumulative explosive growth of PM$_{2.5}$ during winter heavy pollution episodes in Beijing from 2013 to 2016

Junting Zhong$^1$, Xiaoye Zhang$^{1,2}$, Yunsheng Dong$^3$, Yaqiang Wang$^1$, Cheng Liu$^{2,3,4}$, Jizhi Wang$^1$, Yangmei Zhang$^1$, Haochi Che$^5$

$^1$State Key Laboratory of Severe Weather & Key Laboratory of Atmospheric Chemistry of CMA, Chinese Academy of Meteorological Sciences, China.
$^2$Center for Excellence in Regional Atmospheric Environment, IUE, Chinese Academy of Sciences, China.
$^3$Key Laboratory of Environmental Optics and Technology, Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, China.
$^4$School of Earth and Space Sciences, University of Science and Technology of China, Hefei 230026, China
$^5$Department of Physics, University of Oxford, UK.

Correspondence to: X.Y. Zhang (xiaoye@camscma.cn)

Abstract. In January of 2013, February of 2014, December of 2015, and December of 2016 to January 10th of 2017, 12 persistent heavy aerosol pollution episodes (HPEs) occurred in Beijing, which has attracted special attention from the public. During the HPEs, the precise cause of PM$_{2.5}$ explosive growth (mass concentration at least doubled in several to ten hours) is uncertain. Here, we analyzed and estimated relative contributions of boundary-layer meteorological factors to such growth, using ground and vertical meteorological data. Beijing HPEs are generally characterized by the transport stage (TS), whose aerosol pollution formation is primarily caused by pollutants transported from the south of Beijing, and the cumulative stage (CS), in which the cumulative explosive growth of PM$_{2.5}$ mass is dominated by stable atmospheric stratification characteristic of southerly slight or calm winds, near-ground anomalous inversion, and moisture accumulation. During the CSs, observed southerly weak winds facilitate local pollutant accumulation by minimizing horizontal pollutant diffusion. Established from TSs, elevated PM$_{2.5}$ levels scatter more solar radiation back to the space to reduce near-ground temperature, which very likely causes anomalous inversion. This surface cooling by PM$_{2.5}$ decreases near-ground saturation vapor pressure and increases relative humidity significantly; the inversion subsequently reduces vertical
turbulent diffusion and boundary layer height to trap pollutants and accumulate water vapor. Appreciable near-ground moisture accumulation (RH>80%) would further enhance aerosol hygroscopic growth and accelerate liquid-phase and heterogeneous reactions, in which incompletely quantified chemical mechanisms need more investigation. Noted positive meteorological feedback on PM$_{2.5}$ mass explains over 70% of cumulative explosive growth.

**Keyword:** Southerly transport; anomalous inversion; moisture accumulation; meteorological feedback.
39 1 Introduction

Since a persistent heavy fog and haze event occurred in eastern China in January 2013, fine particulate matter smaller than 2.5 μm in diameter (PM$_{2.5}$), as a key component of pollution episodes, has drawn wide attention all over China. Elevated PM$_{2.5}$ leads a sharp decrease in visibility that affects economic activities by causing traffic disruptions and contains toxic substances that affect respiratory and circulatory system (Chen et al., 2013; Bai et al., 2007). The interaction between aerosol and radiation directly and indirectly affects weather and climate (Zhang et al., 2013; Zhang et al., 2015b; Wei et al., 2011; Boucher et al., 2013; Wang et al., 2010). China has experienced heavy aerosol pollution episodes recently, with PM$_{2.5}$ reaching unprecedentedly high levels in many cities, particularly Beijing and its vicinity (BIV), which is one of the nation’s most polluted regions (Zhang et al., 2012).

To elucidate the causes of such heavy pollution episodes, a variety of explanations have been proposed (Huang et al., 2014; Sun et al., 2014a; Sun et al., 2014b; Wang et al., 2014b; Wang et al., 2014c). Previous studies found that atmospheric conditions represented one critical parameter in regulating the cycles of pollution episodes in Beijing in autumn 2013 (Guo et al., 2014; Zhang et al., 2009) and in the North China Plain and other areas in China (Zhang et al., 2015c). During one pollution episode, an analysis of atmospheric background fields revealed dynamic and thermodynamic effects substantially affected pollution formation (Zhang et al., 2014). Specifically, aerosol pollution in Beijing was possibly contributed by southerly/southwesterly surface wind (Wang et al., 2013b). This likely attribution was further verified by source apportionment from the Beijing Environmental Protection Bureau in 2012–2013. In addition, aerosol pollution can be formed by secondary aerosol formation through atmospheric chemical reactions, including liquid-phase reactions, in which aqueous SO$_2$ is oxidized by NO$_2$, H$_2$O$_2$ and O$_3$ to form sulfate, and heterogeneous reactions, in which NO$_2$ and N$_2$O$_5$ form nitrates with water (Zheng et al., 2015a; Cheng et al., 2016).

Although these cited studies existed, the formation mechanism during different stages for heavy aerosol pollution in Beijing, especially the explosive growth stage of PM$_{2.5}$ mass concentration, is still not clear. Previous studies focused more on whether unfamiliar chemical mechanisms were not or inadequately considered in Beijing, a region with high concentrations of various aerosol components.
(Wang et al., 2014b). This view was questioned by subsequent research, suggesting that such rapid growth is mainly attributable to the regional transport of clean and polluted air mass, which derived from the comparison between surface meteorological factors and the PM$_{2.5}$ mass concentration in several cities of the North China Plain (Zheng et al., 2015b). However, the attribution of such growth’s drivers is unreasonable occasionally, because the rapid growth may occur with weak surface winds and stable stratification, which are unfavorable for transport. Then vertical meteorological variations in the boundary layer (BL) in one autumnal episode have been analyzed, which significantly affect the PM$_{2.5}$ mass concentration near the ground (Hua et al., 2016). However, in the absence of long-term observations of meteorological factors and pollutant concentrations, most research concerning pollution causes focuses on one or several consecutive pollution episodes in a certain time, and almost no research attempt to investigate, conclude and quantify the contributions of meteorological factors to the majority of heavy pollution episodes since 2013, particularly the feedback effect of meteorological factors during explosive growth processes. Such investigations will definitely provide a clearer understanding of roles that various vertical meteorological factors play in heavy pollution episodes. Therefore, this paper primarily uses vertical measurements of meteorological factors in the BL from 2013 to 2016, investigates their contributions to the explosive growth of PM$_{2.5}$ during the heavy pollution episodes in Beijing, and also attempts to quantify the effect of meteorological factors on the explosive growth of PM$_{2.5}$ levels.
2 Methods

In this study, the following data are used. (1) Hourly PM$_{2.5}$ mass concentration measured by state-controlled stations of the Ministry of Environmental Protection in Beijing and Baoding. PM$_{2.5}$ mass concentrations of urban stations were averaged to represent urban pollution conditions. (2) Atmospheric vertical observations twice daily at 0800 Beijing Time (BJT) and 2000 BJT, including winds, temperature and relative humidity (RH), measured using L-band radiosonde radar at the observatory (54511) in southern Beijing from 1 January 2013 to 31 January 2013, 1 February 2014 to 28 February 2014, 26 November 2015 to 31 December 2015 and 21 December 2016 to 10 January 2017. The observatory is located on the edge of urban area of Beijing, so it could be used to represent urban vertical meteorological conditions to some degree due to the regional change characteristics of air masses. (3) Hourly ground-level meteorological observations from automatic weather stations (AWSs) provided by the National Meteorological Information Center of the China Meteorological Administration. (4) Lidar observations were measured every fifteen minutes by one Mie-elastic backscatter polarization lidar emitting short pulses of 20 Hz at 532nm in the Institute of Atmospheric Physics, located in the northern urban area of Beijing. The optical parameters of the aerosol particles were retrieved by the backscattering signals. Then the vertical profiles of the aerosol extinction coefficient and linear depolarization ratio were obtained based on the assumptive lidar ratios as 50 for aerosols using the Fernald’s method (Fernald, 1984;Lv et al., 2017). (5) A parameterized index, PLAM (Parameter Linking Aerosol Pollution and Meteorological Elements), calculated with the observations from the observatory (54511). PLAM was built as a function of the following parameters:

$$ PLAM (F) \in f(p, t, w, rh, e, s, c', \ldots) $$

(1)

where $p$, $t$, $w$, $rh$, $e$, $s$, and $c'$ represent air pressure, air temperature, winds, relative humidity, evaporability, stability, and effective parameter associated with the contribution of air pollution $\beta(c')$, respectively. PLAM is further attributed to two major separate factors: (1) initial meteorological conditions $\alpha(m)$ associated with the atmospheric condensation processes and (2) a dynamic effective parameter associated with the initial contribution of air pollution $\beta(c')$:

$$ PLAM = \alpha(m) \times \beta'(c) $$

(2)
It mainly indicates the regional atmospheric stability and air condensation ability. The details of its calculation are presented in previous studies (Wang et al., 2013a; Zhang et al., 2015c; Zhang et al., 2009; Wang et al., 2012).

3 Results and discussion:

3.1 Characteristics of explosive growth in HPEs

A period during which the PM$_{2.5}$ level is less than 35 µg m$^{-3}$ is defined as a clean period based on the PM$_{2.5}$ daily mean mass concentration limit in the primary standard of China’s national environmental quality standards, while a pollution episode is referred to as an episode during which the PM$_{2.5}$ exceeds 80 µg m$^{-3}$ for 3 consecutive days between two clean periods. Pollution episodes with peak PM$_{2.5}$ values less than 300 µg m$^{-3}$ or more than 400 µg m$^{-3}$ are termed light pollution episodes (LPEs) or heavy pollution episodes (HPEs), respectively.

Based on the urban PM$_{2.5}$ monthly mean mass concentration in winter Beijing from 2013 to 2016, the months with highest mass concentration each year were selected to represent the severe PM$_{2.5}$ pollution conditions in winter, which are January in 2013, February in 2014, December in 2015 and December in 2016 respectively. These months are termed the wintertime pollution period (WPP) for the convenience of further investigation.

During the WPP, 12 HPEs occur in total (Fig. 1–4 (dark gray)), whose PM$_{2.5}$ mass concentration is 244.3 µg m$^{-3}$ on average. The maximum mean value (307.4 µg m$^{-3}$) appears in HPE$_{1}$, which has been analyzed in detail in a variety of papers (Zhang et al., 2013; Zhang et al., 2014). The concentrations of HPE$_{6}$ and HPE$_{10}$ are 304.2 µg m$^{-3}$ and 294.5 µg m$^{-3}$ respectively, which are slightly lower than HPE$_{1}$. The minimum mean concentration of PM$_{2.5}$ occurs in HPE$_{8}$ (160.4 µg m$^{-3}$), which is nearly twice as much as the mean annual mass concentration of PM$_{2.5}$ in 2015, nevertheless.

Typical PM$_{2.5}$ rising processes (color-mark) in HPEs were selected, which appeared in 11 of the 12 HPEs. The green-mark processes are tentatively referred to as rising processes, since they generally keep rising consistently with relatively strong southerly winds compared with subsequent growth and vary sensitively and rapidly in response to wind shift from northerly to southerly in the BL. During HPEs, the
growth processes in which the PM$_{2.5}$ mass concentration is at least doubled in several or ten hours in the later period of HPEs are termed explosive growth processes. The red-mark explosive growth processes are tentatively termed cumulative explosive growth processes because of anomalous inversion facilitating pollutant accumulation. The purple-mark explosive growth processes are tentatively known as convergent explosive growth processes, for local wind convergence occurs (Fig. 6) with weak wind velocity and no anomalous inversion. The early stages of HPEs during which PM$_{2.5}$ keeps rising are defined as transport stages (TSs), while the later stages during which cumulative/convergent explosive growth appears are termed cumulative stages (CSs).

3.2 Meteorological causes of the explosive growth in HPEs

3.2.1 PM$_{2.5}$ pollution formation is primarily caused by pollutants transported from the south of Beijing, which subsequently worsen weather conditions

We found that during clean periods occur mostly strong northwesterly winds whose velocity increases with height; during the HPEs, the southwesterly winds with dramatically decreased velocity were most frequent (Fig. 1-4 (a, b)). Strong northerly winds and weak southerly winds closely correspond to the clean periods and the HPEs respectively, because northwesterly winds, which are from less populated north mountainous areas, carry unpolluted air masses while southerly winds carry polluted air masses from more populated and polluted southern industrial regions (Jia et al., 2008; Liu et al., 2013; Guo et al., 2014). During the TSs with southerly winds, air temperature and moisture substantially increase compared with clean periods with northerly winds (Fig. 1-4 (b, c, d)), which indicates warm and humid southerly airflow transport more water vapor and heat into Beijing. During 15 rising processes ((green lines)), nearly no striking near-ground (<250m) moisture accumulation appears; no anomalous inversion appears except brief weak inversion, which suggest that vertical variations of temperature and RH are unlike to primarily cause such rising. Nevertheless, weak inversion and more near-ground moisture favor growth. If we assume that the primary cause of this rising is pollution accumulation due to local emissions, the rising needs to coincide with light (0.3–1.5 m s$^{-1}$) or calm (0–0.2 m s$^{-1}$) air observed during the later
TSs instead of slight (1.6~3.3 m s\(^{-1}\)) or gentle (3.4~5.4 m s\(^{-1}\)) breeze observed during the early TSs, because weaker winds result in a stagnant condition, which is more favorable for local accumulation. However, the majority of later TSs with calm air do not exhibit such rising (Fig. 1-4 (a, b)), which suggests local emissions under weak winds are likely conducive but not dominant with respect to rising. Before rising processes during HPE\(_{1-2}\), the urban PM\(_{2.5}\) mass concentration of Baoding (light gray lines), which is typically representative of pollution conditions in the south of Beijing, was much higher than Beijing; the winds in Beijing rapidly shifted from northerly to southerly. Then the rising (green lines) occurred, consistently with southerly slight or gentle breezes in the BL (green boxes). The southerly air mass move more than 288 km d\(^{-1}\) below 500 m (estimated from the measured wind speed), which are fast enough to transport pollutants to Beijing. Similar conditions appeared in 8 of other 9 HPEs with rising. Such processes indicate southerly pollutant transport is primarily responsible for the rising, given the pollution transport pathway of the southwest wind belt determined by the unique geographic features of the North China Plain, with the Tai-hang Mountains and the Yan Mountains strengthening the southwest wind belt and leading the convergence of pollutant transport in Beijing. (Su et al., 2004). Governed by this transport pathway, PM\(_{2.5}\) mass concentration increased by ~400 μg m\(^{-3}\) from less than 35 μg m\(^{-3}\) in ten hours on 22 January 2013, when winds shifted from northerly to southerly with much higher PM\(_{2.5}\) concentrations in Baoding. Pollutants transported from the south of Beijing primarily results in PM\(_{2.5}\) pollution formation in the urban Beijing area, to which possible weak inversion and the near-surface moisture accumulation is conducive. Warm and humid airflow from the south transports more water vapor and pollutants to the North China Plain, which creates the requisite moisture and pollution accumulation conditions for subsequently cumulative explosive growth.

### 3.2.2 Worsening meteorological conditions primarily cause cumulative explosive growth

*Feedback of anomalous inversion on pollutant accumulation*

Anomalous inversion occurs during 10 of 12 HPEs (Fig. 1~4 (a, c)). The factors that cause inversion in Beijing includes topography, advection and radiation. With the Tai-hang Mountains and the Yan
Mountains lying north of Beijing, a cold air mass flows down into the urban of Beijing from the mountain peaks, which occasionally causes topography inversion; advection inversion occurs when a warm and less dense air mass moves over a cold and dense air mass. However, during most cumulative stages, the anomalous inversion appears with slight or calm winds, which suggests that the movement of air masses is not striking, so the contribution of topography and advection to such inversion is limited. The ground emits long-wave radiation at night to reduce near-ground temperature to facilitate inversion occasionally. However, almost no anomalous inversion occurs without pre-existing high PM$_{2.5}$ mass concentration in the WPP (Fig. 1–4), which suggests that the ground radiation are likely conducive to weak/normal inversion, but not dominant with respect to anomalous inversion.

Noted anomalous inversion is preceded by existing relatively high PM$_{2.5}$ levels generally established by the rising processes. Before the cumulative explosive growth, existing aerosols are concentrated below 500 m (Fig. 5). These low-layer aerosols back-scatter amounts of radiation to space (Wang et al., 2014a; Gao et al., 2015), and cause a significant reduction in radiation reaching the ground, which further reduces near-ground temperature. These findings indicate that anomalous inversion is primarily due to radiation cooling effect of pre-existing aerosols. Below the inversion, near-ground temperature reduction cools down plumes or thermals of originally warm surface air to decrease thermal turbulence; observed weakened vertical shear of horizontal winds (Fig. 1–4 (b)) produces less vorticity to reduce mechanical turbulence, which further strengthens the existing inversion.

Anomalous inversion traps pollution-laden air beneath it due to its strong static stability (Wallace and Hobbs, 2006). It facilitates pollutant accumulation by suppressing vertical air mixing and reducing BL height. During the cumulative explosive growth with anomalous inversion in the HPE$_{10}$, the turbulent diffusion coefficient rapidly decreases from 100 m$^2$ s$^{-1}$ to 50 m$^2$ s$^{-1}$ (model output of CUACE/Chem, the meso-scale China Meteorological Administration (CMA) Unified Atmospheric Chemistry modelling system, personal communication with Dr. Hong Wang), and similar conditions of turbulent diffusion have been modelled in another pollution episode in Beijing (Wang et al., 2015a; Wang et al., 2015b); the BL height decreases from ~500 m in the early morning, to ~350 m at noon, even to ~250 m at night (Fig. 5), which coincide with the increase of PM$_{2.5}$ from ~200 to ~450 μg m$^{-3}$ (Fig. 4 (a)). The striking layered structure in the BL occurs at the height of ~300 m on 20 December 2016 (Fig. 5), which is consistent
with the lower edge of anomalous inversion (Fig. 4), which verifies the strong inhibition of anomalous inversion. Additionally, a short cold air mass invades the northern urban area of Beijing in the early morning on 20 December 2016. The enhanced movement increases the BL height and reduces the PM$_{2.5}$ mass concentration in part of northern urban area (Fig. 5), which slightly reduces the urban mean mass concentration of PM$_{2.5}$ (Fig. 4 (1)). However, the anomalous inversion rapidly restores its original structure to facilitate pollutant accumulation.

The occurrence of anomalous inversion in 9 HPEs coincides with cumulative explosive growth of PM$_{2.5}$ levels (F1–4 (a, c)), which verifies the suppression of anomalous inversion to pollutants. Note that no cumulative explosive growth of PM$_{2.5}$ levels appears in HPE$_3$ despite anomalous inversion, partly because the height of the lowest inversion layer in HPE$_3$ (~750 m) is much higher than that in the 9 HPEs (~250 meters), which suggests near-ground inversion is more favorable for pollutant accumulation.

**Anomalous inversion results in near-surface moisture accumulation**

During clean periods, the moisture is evenly distributed in the BL with RH less than 40%, while during the HPEs, RH is over 60% (even 80%) in the lower or upper BL (Fig. 1–4 (c, d)). During the HPEs, in the absence of temperature inversion, moisture vertically distributes in the BL, and the RH in the upper BL is occasionally higher than that of the near-ground surface; in the presence of weak inversion, the lower edge of the inversion layer is in approximate agreement with the RH contour of 60%.

In the presence of anomalous inversion (red boxes in Fig. 1–4 (c)) in the BL, the lower edge of the strong inversion layer frequently coincides with an RH contour of 80% (red boxes in Fig. 1–4 (d)), which is observed in most cumulative explosive growth processes.

The previously noted relation of vertical temperature and RH indicates that anomalous inversion results in appreciable near-surface moisture accumulation by suppressing the vertical mixing of the water vapor (Wallace and Hobbs, 2006). The vertical diffusion of the near-surface water vapor as the anomalous inversion disappeared on 1 December 2016, 26 December 2016, and 5 January 2017 verifies the cited research outcome. Noted that mentioned near-ground temperature reduction caused by cooling effects of aerosols is also conducive to moisture accumulation by decreasing near-ground saturation vapor pressure to increase RH.
Moisture accumulation facilitates aerosol hygroscopic growth and additional secondary aerosol formation

Strong absorbent aerosol particles absorb and grow, when additional water vapor appears in the air (Zhang et al., 2015a). The mass concentrations of organic aerosols, sulfate, nitrate, and ammonium rapidly increase with RH (Fig. S1). After moisture absorption in North China, aerosol particle size increases 20%~60% (Pan et al., 2009) and aerosol direct radiative forcing increases ~50% (Zhang et al., 2015a). As a key component of atmospheric aerosols, aerosol water serves as a medium that enables aqueous-phase reactions (Pilinis et al., 1989; Seinfeld and Pandis, 1986; Ervens et al., 2011). For example, aerosol water serves as a reactor in which alkaline aerosol components trap SO$_2$, which is then oxidized by NO$_2$ to form sulfate in northern China (Cheng et al., 2016). The ratio of SO$_2$ to SO$_4^{2-}$ ranges from less than 0.1 at relative humidity (RH) <20% to 1.1 at RH >90%, exhibiting an exponential increase with RH (Wang et al., 2016). In addition, high RH facilitates heterogeneous chemical processes to aggravate air pollution (Zhu et al., 2011). For example, the net reaction probability of HNO$_3$ uptake on CaCO$_3$ particles was found to increase with relative humidity from ~0.003 at 10% to 0.21 at 80% (Liu et al., 2008).

Stable atmospheric stratification characteristic of southerly light or calm winds, anomalous inversion, and near-ground (<250 m) moisture accumulation (RH >80%) dominates the cumulative explosive growth of PM$_{2.5}$.

During the HPEs, nearly all 10 cumulative explosive growth processes (Fig. 1-4) occur concurrently with stable atmospheric stratification primarily characterized by southerly light or calm winds, near-ground anomalous inversion, and cumulative moisture (RH>80%). The weak southerly winds increased with height are conducive to the growth, because relatively strong southerly winds in the upper BL (~1000m) transport pollutants from the south of Beijing, while low-level (~250m) southerly light or calm winds limiting the invasion of northerly cold winds facilitates local pollution accumulation by minimizing horizontal pollutant diffusion. The anomalous inversion facilitates vertical pollutant accumulation by suppressing convection activities. During the cumulative growth process in HPE$_{10}$, the turbulent diffusion coefficient rapidly decreases from 100 m$^2$ s$^{-1}$ to 50 m$^2$ s$^{-1}$ and the BL height decreases
from 500 m to ~250 m, which is extremely favorable for pollutants accumulation. Additional suppression of vertical mixing of water by inversion and previously noted decreased saturation vapor pressure cause near-surface moisture accumulation (RH>80%). This accumulated moisture facilitates secondary aerosol formation in liquid-phase and heterogeneous reactions to increase PM$_{2.5}$ levels.

It’s likely that merely weak southerly winds or near-ground anomalous inversion primarily can cause cumulative explosive growth. However, the growth does not occur in the polluted process from 4 to 15 December 2015 with weak southerly winds, which indicates that weak southerly winds do not suffice to cause cumulative explosive growth in the absence of anomalous inversion; even with anomalous inversion, no explosive growth appeared on 14 and 24 January 2013, which suggests that anomalous inversion cannot cause explosive growth without weak southerly winds. Therefore, cumulative explosive growth in CSs is primarily resulted from the joint effects of southerly light or calm winds, near-ground anomalous inversion and moisture accumulation.

Note that the cumulative explosive growth at 2000 BJT on 3 January was accompanied by a southerly gentle breeze (3.4~5.4 m s$^{-1}$), which suggests that low-level southerly pollutant transport occasionally exerts an important impact on the growth, with anomalous inversion and near-ground moisture accumulation.

*Feedback of cumulative pollutants on worsening meteorological conditions*

Established from cumulative explosive growth, exceedingly high PM$_{2.5}$ levels further decrease the near-ground temperature by reflecting and scattering more solar radiation, which strengthens the existing anomalous inversion and subsequently results in additional pollutant accumulation until the next synoptic process occurs. The near-surface temperature decreased from 3°C at 2000 BJT on 19 December to -3°C at 0800 BJT on 20 December after elevated ground PM$_{2.5}$ levels (Fig. 4 (d)). Then it had remained at ~1°C with PM$_{2.5}$ of more than 400 μg m$^{-3}$ over the next 2 d until northerly strong and clean winds blew the pollution away on 22 December. Similar processes also occurred in the CSs of other HPEs, which verifies the outcome.
3.2.3 Local air convergence is favorable for convergent explosive growth.

The explosive growth of PM$_{2.5}$ appears in HPE$_{4.5}$ without inversion and near-ground moisture accumulation (Fig. 2), which suggests previously noted stable atmospheric stratification does not primarily cause the growth. Weak winds in convergent explosive growth processes, particularly the process in HPE$_{5}$, eliminate the likely contributions of southerly transport pollution. A comparison of surface wind distributions in the North China Plain before (Fig. 6 (a, d)) and during (Fig. 6 (b, c; d, e)) the convergent explosive growth processes in HPE$_{4.5}$ shows that the urban area of Beijing is dominated by northerly winds before the growth, while is characterized by local air convergence during the processes, which suggests that the persistent local convergence is conducive to the explosive growth by causing pollutants to further locally accumulate. The convergent explosive growth in HPE$_{6.7}$ with air convergence (Fig. S2) also verifies the outcome.

3.3 Quantification of meteorological contributions to PM$_{2.5}$ cumulative explosive growth

Cooling effects of elevated PM$_{2.5}$ levels established from TSs worsen meteorological conditions, which primarily causes cumulative explosive growth. To approximately quantify this atmospheric feedback on the growth, PLAM (Parameter Linking Aerosol Pollution and Meteorological Elements) was used, which was derived from the relationship of PM$_{2.5}$ with key meteorological parameters. The PLAM index, whose details of calculation have been described in Wang et al. (Wang et al., 2013a; Wang et al., 2012), primarily reflects the stability of the air mass and the condensation rate of water vapor on aerosol particles. It has been employed to identify the contribution of specific meteorological factors to a 10 d haze–fog event in 2013 (Zhang et al., 2013) and to evaluate the contribution of meteorological factors to changes in atmospheric composition and optical properties over Beijing during the 2008 Olympic Games (Zhang et al., 2009). During cumulative explosive growth processes, the hourly variation of urban mean PM$_{2.5}$ mass concentration is in closely linear agreement with that of PLAM for Beijing (Fig. 7 (a–d)). The squared correlation coefficients between hourly PLAM and PM$_{2.5}$ in 2013, 2015, and 2016 are 0.71, 0.69, and 0.71 respectively, exceeding the 0.05 significance level. The mean value of four coefficients is over 0.70, which suggests the noted feedback of worsening meteorological
conditions on PM explains over 70% in cumulative explosive growth of PM$_{2.5}$. In addition, the squared correlation coefficients between PLAM and PM$_{2.5}$ in 2014 is 0.76, which indicates enhanced regional atmospheric stability facilitate convergent explosive growth of PM$_{2.5}$.

4 Conclusion:

We have characterized different stages of 12 HPEs during the WPP (wintertime pollution periods) in Beijing and typical explosive growth of PM$_{2.5}$, including cumulative and convergent explosive growth. Meteorological causes to such growth are elucidated, based on observations of vertical meteorological factors within the BL (Fig. 8). Beijing HPEs can generally be divided into the TS, whose rising processes is primarily caused by pollutants transported from south of Beijing, and the CS, in which stable atmospheric stratification dominates the cumulative explosive growth of PM$_{2.5}$.

Polluted and humid airflow from the south of Beijing transports water vapor and pollutants to Beijing, which primarily causes rising processes and creates the requisite moisture and pollution accumulation conditions for CSs. Elevated PM$_{2.5}$ levels established from the TS reduce near-ground temperature by back scattering short-wave solar radiation. This temperature reduction very likely results in anomalous inversion, which is enhanced by the reduced mechanical turbulence that results from less vorticity caused by observed weakened vertical shear of horizontal winds in the lower BL during the later TSs and decreased thermal turbulence with cooling plumes or thermals of originally warm surface air that result from the decreased near-ground temperature. Anomalous inversion reduces turbulent diffusion and decreases the BL height to trap pollutants. The similar suppression of anomalous inversion to vertical mixing of water vapor and decreased saturation water vapor pressure caused by noted temperature reduction result in appreciable near-surface moisture accumulation (RH>80%). The accumulated moisture facilitates pollutant accumulation by enhancing hygroscopic growth and accelerating liquid-phase and heterogeneous reactions. However, specific reaction mechanisms have not been fully quantified and require additional investigation, particularly their contributions to the explosive growth and the maintenance of PM$_{2.5}$ during CSs. Note that observed southerly weak winds facilitate local pollutant accumulation by minimizing horizontal pollutant diffusion. The joint effects of southerly weak winds, near-ground anomalous inversion, and moisture accumulation dominate cumulative explosive
growth of PM$_{2.5}$. Nearly 70% of the growth is attributable to noted meteorological feedback, based on correlation analysis between PM$_{2.5}$ and PLAM index during cumulative explosive growth processes. Note that sporadic local air convergence also causes pollutants to further accumulate.

Established from cumulative explosive growth, exceedingly high PM$_{2.5}$ levels further decrease the near-ground temperature to strengthen the existing anomalous inversion, which results in additional pollutant accumulation until the next synoptic process occurs.

References


Data Availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Acknowledgements:

This research is supported by the National Key Project of MOST (2016YFC0203306), the Atmospheric Pollution Control - the Prime Minister Fund, and the Basic Scientific Research Progress of the Chinese Academy of Meteorological Sciences (2016Z001).

Author Contributions:

X.Y.Z. and Y.Q.W. designed the research; X.Y.Z, J.T.Z and H.C.C carried out the analysis of observations. Y.S.D provided and analyzed laser radar data. Y.M.Z provided aerosol species data. J.Z.W provided PLAM data. J.T.Z. wrote the first manuscript and X.Y.Z. revised the manuscript. All authors read and approved the final version.

Additional Information:

Competing financial interests: The authors declare no competing financial interests.
Figure 1. Temporal variations in urban mean PM$_{2.5}$ and vertical distributions of meteorological factors in January 2013. (a) PM$_{2.5}$ mass concentration (dark gray or gray: Beijing; light gray: Baoding); (b) winds (vectors; red vectors: southwesterly winds) and wind velocity (shadings; units: m/s); (c) temperature (shadings; units: °C); (d) RH (shadings; units: %); (green boxes: rising processes; red boxes: cumulative explosive processes)
Figure 2. Temporal variations in urban mean PM$_{2.5}$ and vertical distributions of meteorological factors in February 2014. (a) PM$_{2.5}$ mass concentration (dark gray or gray: Beijing; light gray: Baoding); (b) winds (vectors; red vectors: southwesterly winds) and wind velocity (shadings; units: m/s); (c) temperature (shadings; units: °C); (d) RH (shadings; units: %); (green boxes: rising processes)
Figure 3. Temporal variations in urban mean PM$_{2.5}$ and vertical distributions of meteorological factors in December 2015. (a) PM$_{2.5}$ mass concentration (dark gray or gray: Beijing; light gray: Baoding); (b) winds (vectors; red vectors: southwesterly winds) and wind velocity (shadings; units: m/s); (c) temperature (shadings; units: °C); (d) RH (shadings; units: %); (green boxes: rising processes; red boxes: cumulative explosive processes)
Figure 4. Temporal variations in urban mean PM$_{2.5}$ and vertical distributions of meteorological factors in December 2016. (a)PM$_{2.5}$ mass concentration (dark gray or gray: Beijing; light gray: Baoding); (b) winds (vectors; red vectors: southwesterly winds) and wind velocity (shadings; units: m/s); (c) temperature (shadings; units: °C); (d) RH (shadings; units: %); (green boxes: rising processes; red boxes: cumulative explosive processes)
Figure 5. Time series of vertical distributions of the extinction coefficient of aerosols observed in the northern urban area of Beijing from 19 to 20 December 2016 (The dashed lines: the approximate boundary layer height)
Figure 6. Surface wind distributions before (a, d) and during (b, c; d, e) two convergent explosive growth processes in February 2014 on the North China Plain.
Figure 7. Correlation between PLAM and PM2.5 during the cumulative explosive growth processes in January 2013 (a), December 2015 (c), December 2016 (d) respectively, and the convergent explosive growth processes in February 2014 (b).
Figure 8. A schematic figure of the formation mechanism for winter heavy pollution episodes in Beijing, which consist of the transport stage (green background) and the cumulative stage (red background).