Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-845
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 15 September 2017

Atmospheric Chemistry and Physics
Discussions

Feedback effects of boundary-layer meteorological factors on 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], Jizhi Wang[1], Yangmei Zhang[1], Haochi Che[4]

[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]Department of Physics, University of Oxford, UK.

Correspondence to: X.Y. Zhang (xiaoye@camscma.cn); Y.S. Dong (ysdong@aiofm.ac.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 explosive growth in fine particulate matter (PM) 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 (PM mass concentration doubled in ~10 hours) 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 limiting the invasion of northerly clean winds and minimizing horizontal pollutant diffusion. Established from TSs, elevated PM levels scatter more solar radiation back to the space to reduce near-ground temperature. This radiation reduction decreases near-ground saturation vapor pressure and very likely causes anomalous inversion. The decreased saturation pressure significantly increases relative humidity; 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%) further enhances aerosol hygroscopic growth and accelerates liquid-

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-845
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 15 September 2017

phase and heterogeneous reactions, in which incompletely quantified chemical mechanisms need more investigation. Noted meteorological feedback on PM explains over 70% in cumulative explosive growth of PM.

Keyword: Southerly transport; anomalous inversion; moisture accumulation; meteorological feedback.

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_2O_2 and O_3 to form sulfate, and heterogeneous reactions, in which NO_2 and N_2O_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

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-845
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 15 September 2017

(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 national monitoring stations of the Ministry of Environmental Protection. $PM_{2.5}$ mass concentrations of urban stations were averaged to represent urban pollution conditions. (2) Atmospheric vertical observations twice daily at 0800 h and 2000 h, 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. (3) A parameterized index, PLAM (Parameter Linking Aerosol Pollution and Meteorological Elements), calculated with the observations from the observatory (54511), based on the calculation method presented in detail in previous studies (Wang et al., 2013a;Zhang et al., 2015c;Zhang et al., 2009;Wang et al., 2012). (4) Hourly ground-level meteorological observations from automatic weather stations (AWSs) provided by the National Meteorological Information Center of the China Meteorological Administration. (5) Lidar observations were measured by one Mie-elastic backscatter polarization lidar emitting short pulses of 20 Hz at 532nm in the Institute of Atmospheric Physics (116.38E, 39.98N), 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).

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. During HPEs, the growth processes in which the $PM_{2.5}$ mass concentration increases from less than 30 µg m^{-3} to 150~300 µg m^{-3} in the initial several or ten hours or increases by over 150 µg m^{-3} rapidly afterward are termed explosive growth processes.

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 (Figure 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 explosive growth processes (color-mark) in HPEs were selected, which appeared in 11 of the 12 HPEs. The green-mark explosive growth processes are tentatively referred to as transport explosive growth processes, because they generally occur consistently with relatively strong southerly winds compared with subsequent explosive growth and vary sensitively and rapidly in response to wind shift from northerly to southerly in the BL. The red-mark explosive growth processes are tentatively

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-845
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 15 September 2017

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 (Figure 6) with weak wind velocity and no anomalous inversion. The early stages of HPEs during which transport explosive growth occurs 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 (Figure 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 (Figure 1-4 (b, c, d)), which indicates warm and humid southerly airflow transport more water vapor and heat into Beijing. During 15 transport explosive growth 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 transport explosive growth. Nevertheless, weak inversion and more near-ground moisture favor growth.

If we assume that the primary cause of this explosive growth is pollution accumulation due to local emissions, the growth 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 are more favorable for local accumulation. However, the majority of later TSs with calm air do not exhibit such explosive growth (Figure 1-4 (a, b)), which suggests local emissions under weak winds are likely conducive but not dominant with respect to growth.

Before transport explosive growth during $HPE_{1\sim2}$, 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 transport explosive growth (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 transport explosive growth. Such processes indicate southerly pollutant transport is primarily responsible for the explosive growth, 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 limiting the invasion of northerly cold air and leading northeast movement of southerly winds. (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 (Figure 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 exceeds 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 (Figure 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 transport explosive growth. Before the cumulative explosive growth, existing aerosols are concentrated below 500 m (Figure 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 ((F1~4 (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); the BL height decreases from ~500 m in the early morning, to ~350 m at noon, even to ~250 m at night (Figure 5), which coincide with the increase of $PM_{2.5}$ from ~200 to ~450 μg m^{-3} (Figure 4 (a)). The striking layered structure in the BL occurs at the height of ~300 m on 20 December 2016 (Figure 5), which is consistent with the lower edge of anomalous inversion (Figure 4), which verifies the strong inhibition of anomalous inversion. Additionally, a short

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-845
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 15 September 2017

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 reduce the $PM_{2.5}$ mass concentration in part of northern urban area (Figure 5), which slightly reduces the urban mean mass concentration of $PM_{2.5}$ (Figure 4 (1)). However, the anomalous inversion rapidly restore 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 (Figure 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 Figure 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 Figure 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.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-845
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 15 September 2017

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 soluble organic aerosols, sulfate, nitrate, and ammonium rapidly increase with RH (Figure 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% (Y. 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 (Figure 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

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-845
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 15 September 2017

turbulent diffusion coefficient rapidly decreases from 100 to 50 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 h 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℃ at 2000 h on 19 December to -3℃ at 0800 h on 20 December after elevated ground $PM_{2.5}$ levels (Figure 4 (d)). Then it had remained at ~-1℃ 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 $HPE_{4\sim5}$ without inversion and near-ground moisture accumulation (Figure 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 (Figure 6 (a, d)) and during (Figure 6 (b, c; d, e)) the convergent explosive growth processes in $HPE_{4\text{-}5}$ 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 $HPE_{6\sim7}$ with air convergence (Figure 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 (Figure 7 (a~d)). The squared correlation coefficients between PLAM and $PM_{2.5}$ from 2013 to 2016 are 0.71, 0.76, 0.69, and 0.71 respectively, exceeding the 0.05 significance level. The mean value of four coefficients is 0.72, which suggests the noted feedback of worsening meteorological conditions on PM explains over 70% in cumulative 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 $PM_{2.5}$ during different stages, including transport, cumulative, and convergent explosive growth. Meteorological causes to such growth are elucidated, based on observations of vertical meteorological factors within the BL (Figure 8). Beijing HPEs can generally be divided into the TS, whose explosive growth 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 transport explosive growth 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 limiting the invasion of northerly clean winds and 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 72% 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

Bai, N., Khazaei, M., van Eeden, S. F., and Laher, I.: The pharmacology of particulate matter air pollution-induced cardiovascular dysfunction, Pharmacology & Therapeutics, 113, 16, 2007.

Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V. V. M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S. K., Sherwood, S., Stevens, B., and Zhang, X. Y.: Clouds and Aerosols, in: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Stocker, T. F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P. M., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 571-658, 2013.

Chen, Y., Ebenstein, A., Greenstone, M., and Li, H.: Evidence on the impact of sustained exposure to air pollution on life expectancy from China's Huai River policy, Proceedings of the National Academy of Sciences of the United States of America, 110, 12936-12941, 10.1073/pnas.1300018110, 2013.

Cheng, Y., Zheng, G., Chao, W., Mu, Q., Bo, Z., Wang, Z., Meng, G., Qiang, Z., He, K., and Carmichael, G.: Reactive nitrogen chemistry in aerosol water as a source of sulfate during haze events in China, Science Advances, 2, 2016.

Ervens, B., Turpin, B. J., and Weber, R. J.: Secondary organic aerosol formation in cloud droplets and aqueous particles (aqSOA): a review of laboratory, field and model studies, Atmospheric Chemistry and Physics, 11, 11069-11102, 10.5194/acp-11-11069-2011, 2011.

Fernald, F. G.: Analysis of atmospheric lidar observations: some comments, Applied Optics, 23, 652, 1984.

Gao, Y., Zhang, M., Liu, Z., Wang, L., Wang, P., Xia, X., Tao, M., and Zhu, L.: Modeling the feedback between aerosol and meteorological variables in the atmospheric boundary layer during a severe fog–haze event over the North China Plain, Atmospheric Chemistry and Physics, 15, 4279-4295, 10.5194/acp-15-4279-2015, 2015.

Guo, S., Hu, M., Zamora, M. L., Peng, J. F., Shang, D. J., Zheng, J., Du, Z. F., Wu, Z., Shao, M., Zeng, L. M., Molina, M. J., and Zhang, R. Y.: Elucidating severe urban haze formation in China, Proceedings of the National Academy of Sciences of the United States of America, 111, 17373-17378, 10.1073/pnas.1419604111, 2014.

Hua, Y., Wang, S., Wang, J., Jiang, J., Zhang, T., Song, Y., Kang, L., Zhou, W., Cai, R., Wu, D., Fan, S.,

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-845
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 15 September 2017

Wang, T., Tang, X., Wei, Q., Sun, F., and Xiao, Z.: Investigating the impact of regional transport on PM2.5 formation using vertical observation during APEC 2014 Summit in Beijing, Atmospheric Chemistry and Physics, 16, 15451-15460, 10.5194/acp-16-15451-2016, 2016.

Huang, R.-J., Zhang, Y., Bozzetti, C., Ho, K.-F., Cao, J.-J., Han, Y., Daellenbach, K. R., Slowik, J. G., Platt, S. M., and Canonaco, F.: High secondary aerosol contribution to particulate pollution during haze events in China, Nature, 514, 218-222, 2014.

Jia, Y., Rahn, K. A., He, K., Wen, T., and Wang, Y.: A novel technique for quantifying the regional component of urban aerosol solely from its sawtooth cycles, Journal of Geophysical Research Atmospheres, 113, 6089-6098, 2008.

Liu, X. G., Li, J., Qu, Y., Han, T., Hou, L., Gu, J., Chen, C., Yang, Y., Liu, X., and Yang, T.: Formation and evolution mechanism of regional haze: a case study in the megacity Beijing, China, Atmospheric Chemistry & Physics, 13, 4501-4514, 2013.

Lv, L., Liu, W., Zhang, T., Chen, Z., Dong, Y., Fan, G., Xiang, Y., Yao, Y., Yang, N., and Chu, B.: Observations of particle extinction, PM 2.5 mass concentration profile and flux in north China based on mobile lidar technique, Atmospheric Environment, 2017.

Pan, X. L., Yan, P., Tang, J., Ma, J. Z., Wang, Z. F., Gbaguidi, A., and Sun, Y. L.: Observational study of influence of aerosol hygroscopic growth on scattering coefficient over rural area near Beijing mega-city, Atmospheric Chemistry & Physics, 9, 7519-7530, 2009.

Pilinis, C., Seinfeld, J. H., and Grosjean, D.: Water content of atmospheric aerosols, Atmospheric Environment, 23, 1601-1606, 1989.

Seinfeld, J. H., and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, John Wiley & Sons, USA, 1986.

Su, F., Gao, Q., Zhang, Z., REN, Z.-h., and YANG, X.-x.: Transport pathways of pollutants from outside in atmosphere boundary layer, Research of Environmental Sciences, 1, 26-29, 2004.

Sun, Y., Jiang, Q., Wang, Z., Fu, P., Li, J., Yang, T., and Yin, Y.: Investigation of the sources and evolution processes of severe haze pollution in Beijing in January 2013, Journal of Geophysical Research: Atmospheres, 119, 4380-4398, 2014a.

Sun, Y. L., Jiang, Q., Wang, Z. F., Fu, P. Q., Li, J., Yang, T., and Yin, Y.: Investigation of the sources and evolution processes of severe haze pollution in Beijing in January 2013, J. Geophys. Res.-Atmos., 119, 4380-4398, 10.1002/2014jd021641, 2014b.

Wallace, J. M., and Hobbs, P. V.: Atmospheric science : an introductory survey, Elsevier, 2006.

Wang, G., Zhang, R., Gomez, M. E., Yang, L., Levy Zamora, M., Hu, M., Lin, Y., Peng, J., Guo, S., Meng, J., Li, J., Cheng, C., Hu, T., Ren, Y., Wang, Y., Gao, J., Cao, J., An, Z., Zhou, W., Li, G., Wang, J., Tian, P., Marrero-Ortiz, W., Secrest, J., Du, Z., Zheng, J., Shang, D., Zeng, L., Shao, M., Wang, W., Huang, Y., Wang, Y., Zhu, Y., Li, Y., Hu, J., Pan, B., Cai, L., Cheng, Y., Ji, Y., Zhang, F., Rosenfeld, D., Liss, P. S., Duce, R. A., Kolb, C. E., and Molina, M. J.: Persistent sulfate formation from London Fog to Chinese haze, Proc Natl Acad Sci U S A, 113, 13630–13635, 10.1073/pnas.1616540113, 2016.

Wang, H., Zhang, X., Gong, S., Chen, Y., Shi, G., and Li, W.: Radiative feedback of dust aerosols on the East Asian dust storms, Journal of Geophysical Research, 115, 6696-6705, 2010.

Wang, J., Wang, Y., Liu, H., Yang, Y., Zhang, X., Li, Y., Zhang, Y., and Deng, G.: Diagnostic identification of the impact of meteorological conditions on PM2.5 concentrations in Beijing, Atmospheric Environment, 81, 158-165, 10.1016/j.atmosenv.2013.08.033, 2013a.

Wang, J., Wang, S., Jiang, J., Ding, A., Zheng, M., Zhao, B., Wong, D. C., Zhou, W., Zheng, G., Wang, L., Pleim, J. E., and Hao, J.: Impact of aerosol–meteorology interactions on fine particle pollution during China's severe haze episode in January 2013, Environmental Research Letters, 9, 094002, 10.1088/1748-9326/9/9/094002, 2014a.

Wang, J. Z., Gong, S., Zhang, X. Y., Yang, Y. Q., Hou, Q., Zhou, C., and Wang, Y.: A parameterized method for air-quality diagnosis and its applications, Advance of Meteorology, doi:10.1155/2012/238589, 1-10, 2012.

Wang, X., Chen, J., Sun, J., Li, W., Yang, L., Wen, L., Wang, W., Wang, X., Collett, J. L., and Shi, Y.: Severe haze episodes and seriously polluted fog water in Ji'nan, China, Sci. Total Environ., 493, 133-137, 2014b.

Wang, Z., Li, J., Wang, Z., Yang, W., Tang, X., Ge, B., Yan, P., Zhu, L., Chen, X., and Chen, H.: Modeling study of regional severe hazes over mid-eastern China in January 2013 and its implications on pollution prevention and control, Science China Earth Sciences, 57, 3-13, 2014c.

Wang, Z. B., Hu, M., Wu, Z. J., Yue, D. L., He, L. Y., Huang, X. F., Liu, X. G., and Wiedensohler, A.: Long-term measurements of particle number size distributions and the relationships with air mass history and source apportionment in the summer of Beijing, Atmospheric Chemistry and Physics, 13, 10159-10170, 10.5194/acp-13-10159-2013, 2013b.

Wei, P., Cheng, S. Y., Li, J. B., and Su, F. Q.: Impact of boundary-layer anticyclonic weather system on regional air quality, Atmospheric Environment, 45, 2453-2463, 10.1016/j.atmosenv.2011.01.045, 2011.

Y. Liu, E.R. Gibson, J.P. Cain, H. Wang, V.H. Grassian, ‡ and, and A. Laskin: Kinetics of Heterogeneous Reaction of CaCO3 Particles with Gaseous HNO3 over a Wide Range of Humidity, Journal of Physical Chemistry A, 112, 1561, 2008.

Zhang, L., Sun, J. Y., Shen, X. J., Zhang, Y. M., Che, H., Ma, Q. L., Zhang, Y. W., Zhang, X. Y., and Ogren, J. A.: Observations of relative humidity effects on aerosol light scattering in the Yangtze River Delta of China, Atmospheric Chemistry & Physics, 15, 2853-2904, 2015a.

Zhang, R. H., Li, Q., and Zhang, R. N.: Meteorological conditions for the persistent severe fog and haze event over eastern China in January 2013, Science China Earth Sciences, 57, 26-35, 2014.

Zhang, R. Y., Wang, G. H., Guo, S., Zarnora, M. L., Ying, Q., Lin, Y., Wang, W. G., Hu, M., and Wang, Y.: Formation of Urban Fine Particulate Matter, Chemical Reviews, 115, 3803-3855, 10.1021/acs.chemrev.5b00067, 2015b.

Zhang, X., Sun, J., Wang, Y., Li, W., Zhang, Q., Wang, W., Quan, J., Cao, G., Wang, J., Yang, Y., and Zhang, Y.: Factors contributing to haze and fog in China, Chinese Science Bulletin (Chinese Version), 58, 1178, 10.1360/972013-150, 2013.

Zhang, X. Y., Wang, Y. Q., Lin, W. L., Zhang, Y. M., Zhang, X. C., Gong, S., Zhao, P., Yang, Y. Q., Wang, J. Z., and Hou, Q.: Changes of Atmospheric Composition and Optical Properties Over BEIJING—2008 Olympic Monitoring Campaign, Bulletin of the American Meteorological Society, 90, 1633–1651, 2009.

Zhang, X. Y., Wang, Y. Q., Niu, T., Zhang, X. C., Gong, S. L., Zhang, Y. M., and Sun, J. Y.: Atmospheric aerosol compositions in China: spatial/temporal variability, chemical signature, regional haze distribution and comparisons with global aerosols, Atmos. Chem. Phys., 11, 26571-26615, 2012.

Zhang, X. Y., Wang, J. Z., Wang, Y. Q., Liu, H. L., Sun, J. Y., and Zhang, Y. M.: Changes in chemical components of aerosol particles in different haze regions in China from 2006 to 2013 and contribution of meteorological factors, Atmos. Chem. Phys., 15, 12935-12952, 10.5194/acp-15-12935-2015, 2015c.

Zheng, B., Zhang, Q., Zhang, Y., He, K. B., Wang, K., Zheng, G. J., Duan, F. K., Ma, Y. L., and Kimoto, T.: Heterogeneous chemistry: a mechanism missing in current models to explain secondary inorganic aerosol formation during the January 2013 haze episode in North China, Atmospheric Chemistry and Physics, 15, 2031-2049, 10.5194/acp-15-2031-2015, 2015a.

Zheng, G. J., Duan, F. K., Su, H., Ma, Y. L., Cheng, Y., Zheng, B., Zhang, Q., Huang, T., Kimoto, T., Chang, D., Poschl, U., Cheng, Y. F., and He, K. B.: Exploring the severe winter haze in Beijing: the impact of synoptic weather, regional transport and heterogeneous reactions, Atmospheric Chemistry and Physics, 15, 2969-2983, 10.5194/acp-15-2969-2015, 2015b.

Zhu, T., Shang, J., and Zhao, D. F.: The roles of heterogeneous chemical processes in the formation of an air pollution complex and gray haze, Science China Chemistry, 54, 145-153, 2011.

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), 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.

Figures

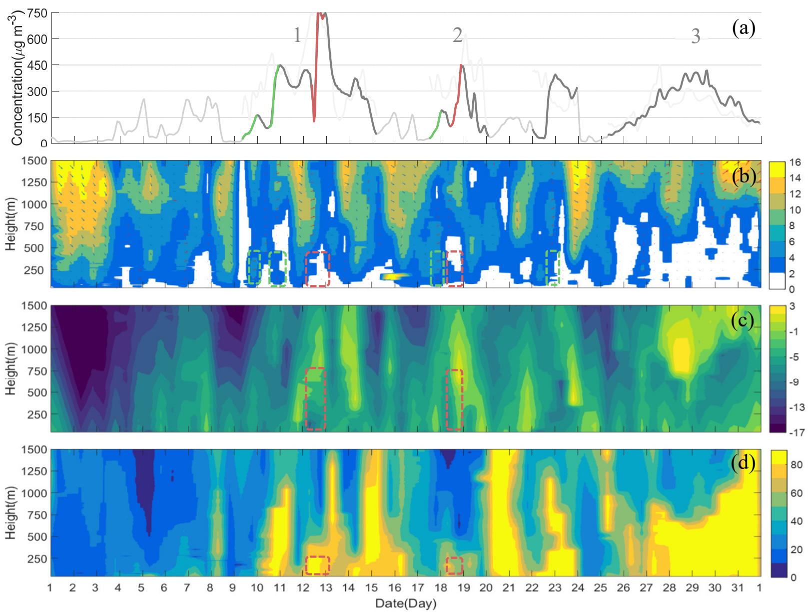

Figure 1. Temporal variations in urban mean $PM_{2.5}$ and vertical distributions of meteorological factors in January 2013. (a)$PM_{2.5}$ mass concentration (dark gray or gray: Beijing; light gray: Baoding); (b) winds (vectors; red vectors: southwesterly winds) and wind velocity (shadings; units: m/s); (c)temperature (shadings; units:℃); (d)RH (shadings; units: %); (green boxes: transport explosive stages; red boxes: cumulative explosive stages)

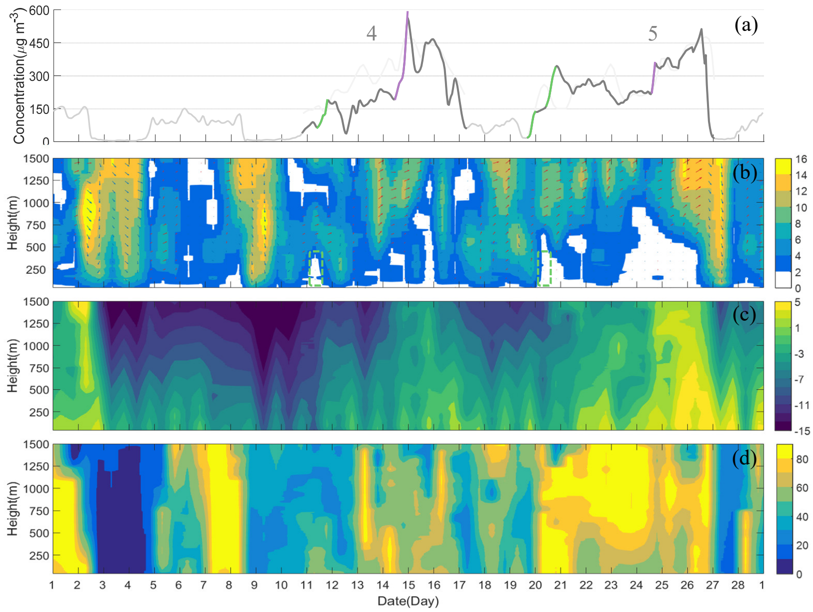

Figure 2. Temporal variations in urban mean $PM_{2.5}$ and vertical distributions of meteorological factors in February 2014. (a)$PM_{2.5}$ mass concentration (dark gray or gray: Beijing; light gray: Baoding); (b) winds (vectors; red vectors: southwesterly winds) and wind velocity (shadings; units: m/s); (c)temperature (shadings; units:℃); (d)RH (shadings; units: %); (green boxes: transport explosive stages)

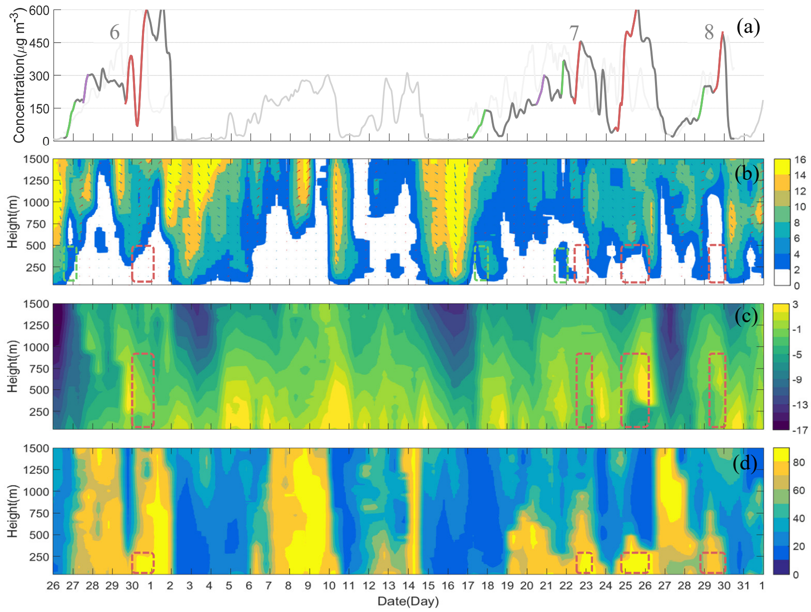

Figure 3. Temporal variations in urban mean $PM_{2.5}$ and vertical distributions of meteorological factors in December 2015. (a)$PM_{2.5}$ mass concentration (dark gray or gray: Beijing; light gray: Baoding); (b) winds (vectors; red vectors: southwesterly winds) and wind velocity (shadings; units: m/s); (c)temperature (shadings; units:℃); (d)RH (shadings; units: %); (green boxes: transport explosive stages; red boxes: cumulative explosive stages)

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-845
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 15 September 2017

Atmospheric Chemistry and Physics
Open Access
Discussions

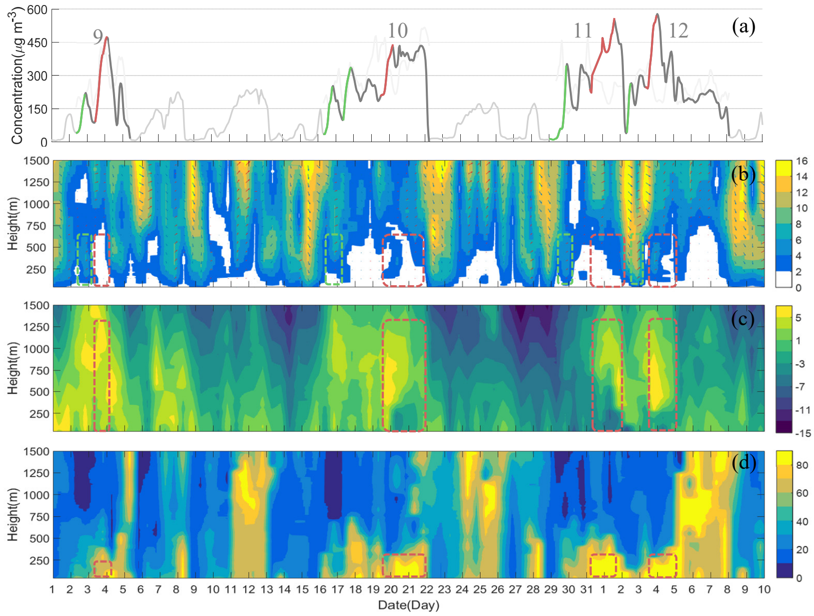

Figure 4. Temporal variations in urban mean $PM_{2.5}$ and vertical distributions of meteorological factors in December 2016. (a)$PM_{2.5}$ mass concentration (dark gray or gray: Beijing; light gray: Baoding); (b) winds (vectors; red vectors: southwesterly winds) and wind velocity (shadings; units: m/s); (c)temperature (shadings; units:℃); (d)RH (shadings; units: %); (green boxes: transport explosive stages; red boxes: cumulative explosive stages)

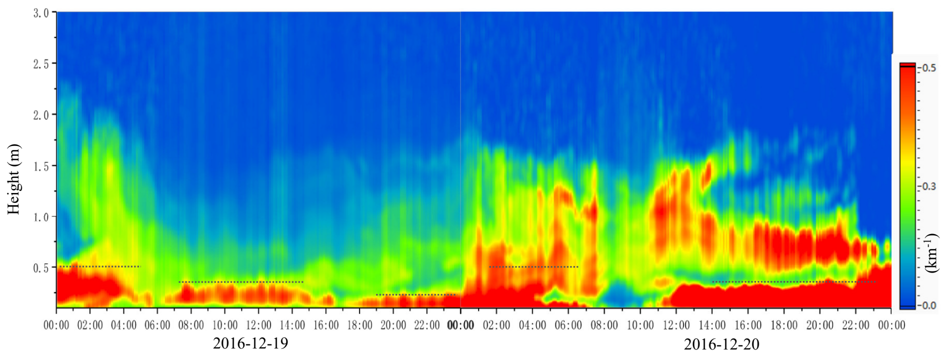

Figure 5. Time series of vertical distributions of the extinction coefficient of aerosols observed in the northern urban area of Beijing from 19 to 20 December 2016 (The dashed lines: the approximate boundary layer height)

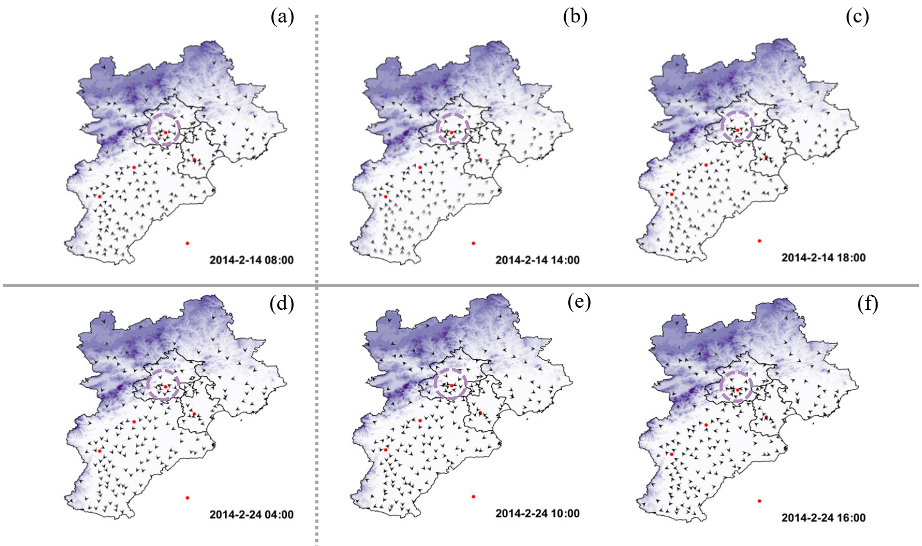

Figure 6. Surface wind distributions before (a, d) and during (b, c; d, e) two convergent explosive growth processes in February 2014 on the North China Plain.

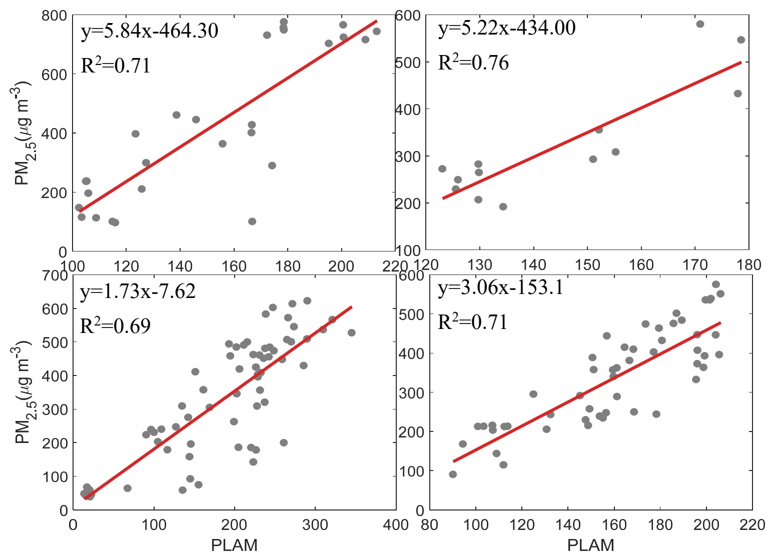

Figure 7. Correlation between PLAM and PM2.5 during the cumulative explosive growth processes in January 2013 (a), February 2014 (b), December 2015 (c), and December 2016 (d) respectively.

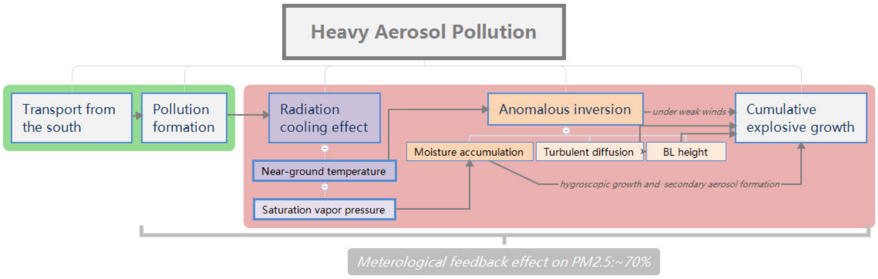

Figure 8. A schematic figure of the formation mechanism for winter heavy pollution episodes in Beijing, which consist of the transport stage (green background) and the cumulative stage (red background).