



# Non-stop industries were the main source of air pollution during the 2020 coronavirus lockdown in the North China Plain

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## Abstract

Despite large decreases of emissions of air pollution during the coronavirus disease 2019 (COVID-19) lockdown in 2020, an unexpected regional severe haze has still occurred over the North China Plain. To clarify the origin of this pollution, we studied air concentrations of fine particulate matter (PM<sub>2.5</sub>), NO<sub>2</sub>, O<sub>3</sub>, PM<sub>10</sub>, SO<sub>2</sub>, and CO in Beijing, Hengshui and Baoding during the lockdown period from January 24 to 29, 2020. Variations of PM<sub>2.5</sub> composition in inorganic ions, elemental carbon and organic matter were also investigated. The HYSPLIT model was used to calculate backward trajectories and concentration weighted trajectories. Results of the clustertrajectory analysis and model simulations show that the severe haze was caused mainly by the emissions of northeastern non-stopping industries located in Inner Mongolia, Liaoning, Hebei, and Tianjin. In Beijing, Hengshui and Baoding, the mixing layer heights were about 30% lower and the maximum relative humidity was 83% higher than the annual averages, and the average wind speeds were lower than 1.5 m s<sup>-1</sup>. The concentrations of NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>, organics and K<sup>+</sup> were the main components of PM<sub>2.5</sub> in Beijing and Hengshui, while organics, K<sup>+</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, and NH<sub>4</sub><sup>+</sup> were the main components of PM<sub>2.5</sub> in Baoding. Contrary to previous reports suggesting a southerly transport of air pollution, we found that northeast transport caused the haze formation.

**Keywords** Spring festival · COVID-19 · North China Plain · Haze

## Introduction

Haze is a weather phenomenon in which dust, smog, and other dry particles affect the atmospheric visibility (Cai et al. 2017; Chang et al. 2009). The USA and European countries began to experience haze in the 1950s, but after decades of governance, air quality has been greatly improved. Unlike the haze in London in 1952 caused by coal burning and Los Angeles smog caused by vehicle emissions in the 1950s, haze in China is the consequence of diverse and high primary emissions, and efficient secondary generations (An et al. 2019). Although the Chinese government has made many years of efforts to control haze by transferring heavy industries from core cities to neighboring cities recently, the North China Plain has a high incidence of heavy haze in winter, and air pollutions there are more serious during the Spring Festival (Xiao et al. 2015; An et al. 2019). The Spring Festival is the most important and lively traditional festival in China, marking the beginning of the Lunar New Year by setting off fireworks and firecrackers. Previous studies have shown that residential heating, industrial production,

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and vehicle emissions caused severe haze pollution in North China Plain in winter (Li et al. 2014; Zhang et al. 2010; Kong et al. 2015).

Meanwhile, the topography and emission characteristics in the North China Plain (Zhang et al. 2019) are conducive to the regional transport and accumulation of pollutants because its north and west sides are Yanshan and Taihang Mountains, respectively. For example, in the winter of 2013, a nationwide haze broke out in China, especially in the North China Plain. Due to the fact that North China Plain is the largest industrial urban agglomeration in China, North China Plain bear the brunt of nationwide haze in 2013. The peak mass concentration of  $PM_{2.5}$  (aerodynamic equivalent diameter less than or equal to  $2.5\ \mu\text{m}$ ) in Beijing in January 2013 reached  $855\ \mu\text{g m}^{-3}$  (Jin et al. 2020; Sun et al. 2017; Yang et al. 2015; Zheng et al. 2015). In addition to the impacts from local emissions, the average contribution rates of regional transport to Beijing's  $PM_{2.5}$  concentrations were found to exceed 40% during the whole year of 2013 (Ge et al. 2018). Several recent studies have shown that areas outside the North China Plain could bring pollutants into cities in the North China Plain through regional transports (Pu et al. 2020; Zhu et al. 2021).

Meteorological factors are another key factor causing air pollution in North China Plain (NCP) in winter. Stable weather conditions are not conducive to the diffusion and sedimentation of pollutants. When haze frequently occurs in winter, the North China Plain is mostly stable weather, which is generally characterized by low wind speed, low mixed layer height (MLH), and high relative humidity (Beyrich 1997; Jin et al. 2020; Sun et al. 2016; Yu et al. 2018; Kanawade et al. 2020; Zhu et al. 2018). The mixed layer is regarded as the link between the near surface and the free atmosphere and a low mixing layer height is not conducive to the diffusion of pollutants (Seidel et al. 2010).

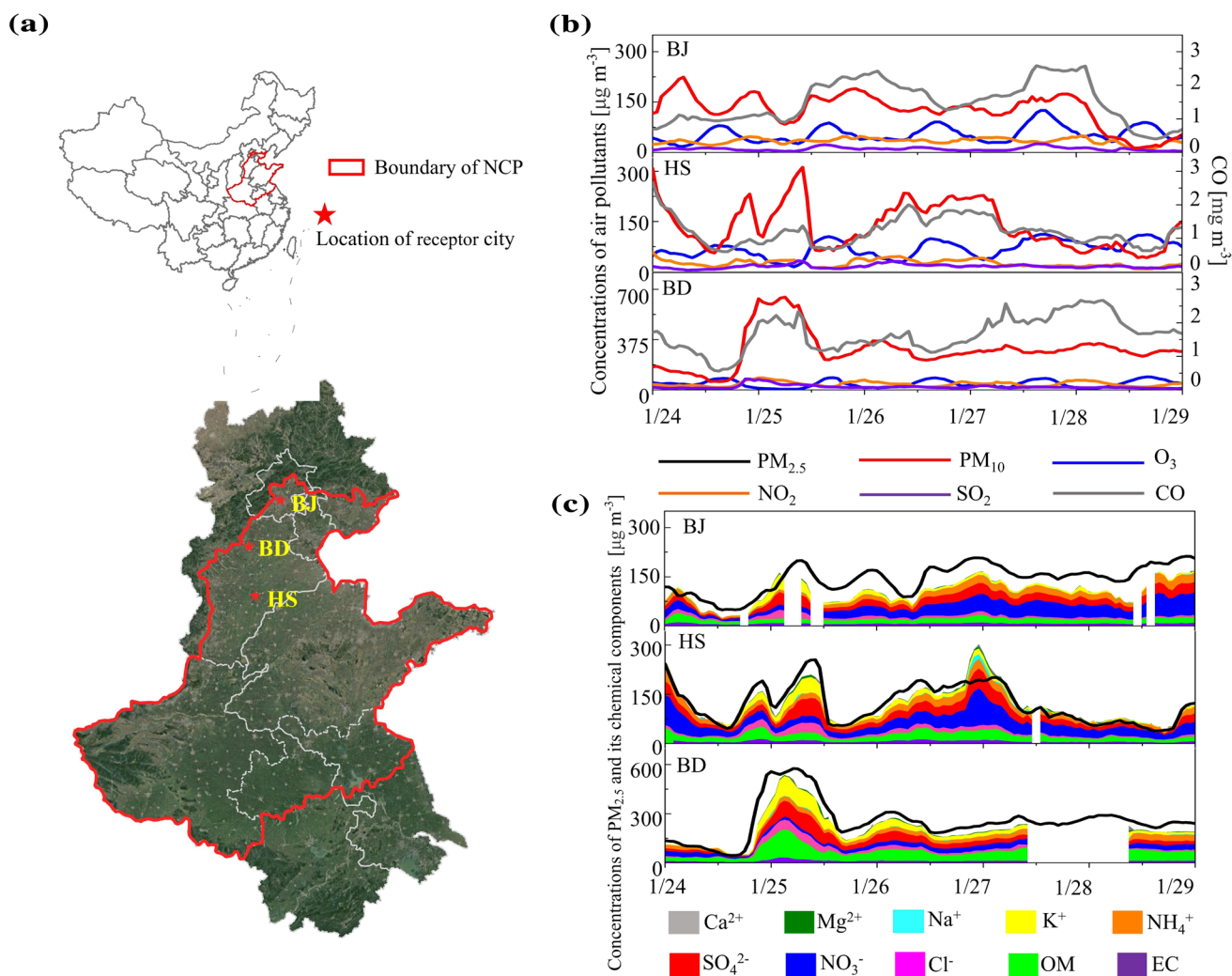
Due to the outbreak of the COVID-19 pandemic, the Spring Festival in 2020 was very special with imposing extreme measures of nationwide lockdown by the Chinese government. The national emergency responses include extension of the Spring Festival holiday, suspension of all public transportation, closes of schools and entertainment venues, prohibition of public gatherings, enhancement of social distancing, and stay-at-home orders and closes of non-essential businesses (Wang et al. 2020a, b; Wang et al. 2021). Some recent scientific studies have shown that the improvement of air quality was one of the social benefits (Dutheil et al. 2020; He et al. 2020), while other studies have shown that limiting human activities by lockdown were not the fundamental way to alleviate air pollution (Wang et al. 2020a, b; Chang et al. 2020). It is worth studying why the unexpected haze problem still existed over the North China Plain when the air pollutant emissions were significantly reduced during the COVID-19 pandemic. The reduction

of emissions from traffic and industrial sources during the COVID-19 lockdown in China provided a unique nature experiment for evaluating the efficiency of air pollution control measures (Le et al. 2020). The overall goal of this study is to investigate the regional severe haze over the North China Plain during the COVID-19 lockdown period from January 24 to January 29, 2020, by conducting a detailed analysis of the characteristics of haze at three representative cities in the North China Plain and digging the origins of the haze formation. The study explored the impact of regional transport on the formation of haze by identifying the pollution source areas of the target cities, and assessed the role of meteorological factors and ground emissions in the special atmospheric pollution during the lockdown.

## Experimental

### Observational data

The study period was from January 24 to January 29, 2020, of which January 24 is the New Year's Eve of 2020 and the first day when China adopted lockdown measures. Figure 1a shows the North China Plain area (Zhang et al. 2019), which spans 5 provinces (Hebei, Shandong, Henan, Anhui, and Jiangsu) and two municipality cities (Beijing, Tianjin), and is one of the three plains and the most populous plain in China. The three receptor cities selected in this study are Beijing, Hengshui and Baoding, all located in the northern part of the North China Plain. One of the reasons for this selection is that the observational data in the three cities were available. Figure S1 shows that the North China Plain (NCP) area borders Yanshan Mountain to the north, Taihang Mountain to the west, and Bohai Sea to the east. Hourly  $PM_{2.5}$ ,  $NO_2$ ,  $O_3$ ,  $PM_{10}$ ,  $SO_2$ , and CO concentrations in three representative cities (there are 12, 4 and 6 monitoring stations in Beijing, Hengshui and Baoding cities, respectively) were obtained from China National Environmental Monitoring Center (CNEMC, <http://www.cnemc.cn>). The hourly chemical component concentrations of  $PM_{2.5}$  at three representative cities were obtained from the National Air Pollution Prevention and Control Joint Center (<https://view.inews.qq.com/media/6474488>). The chemical components include water-soluble inorganic ions (WSIIs) ( $Cl^-$ ,  $NO_3^-$ ,  $SO_4^{2-}$ ,  $NH_4^+$ ,  $Na^+$ ,  $K^+$ ,  $Mg^{2+}$  and  $Ca^{2+}$ ), elemental carbon (EC) and organic matter (OM). According to the hourly concentrations of  $PM_{2.5}$ , the pollution levels were divided into light haze ( $75 \leq PM_{2.5} < 115\ \mu\text{g m}^{-3}$ ), moderate haze ( $115 \leq PM_{2.5} < 150\ \mu\text{g m}^{-3}$ ), heavy haze ( $150 \leq PM_{2.5} < 250\ \mu\text{g m}^{-3}$ ), and severe haze ( $PM_{2.5} \geq 250\ \mu\text{g m}^{-3}$ ). The observed meteorological data were derived from the National Meteorological Data Center website (<http://www.nmic.cn/site/index.html>).



**Fig. 1** **a** Map of China showing the boundary of North China Plain (NCP) and locations of Beijing (BJ), Hengshui (HS), and Baoding (BD) cities. **b** Hourly mass concentrations of five air pollutants; **c** Hourly mass concentrations of  $\text{PM}_{2.5}$  and chemical components in Beijing (BJ), Hengshui (HS), and Baoding (BD) cities during the study period (January 24 to January 29, 2020). The mass concentrations of chemical components were stacked in the figure. The highest values of  $\text{PM}_{10}$ ,  $\text{O}_3$ ,  $\text{NO}_2$ ,  $\text{SO}_2$  and  $\text{CO}$  were  $692 \mu\text{g m}^{-3}$  in Baoding,

$125 \mu\text{g m}^{-3}$  in Beijing,  $90 \mu\text{g m}^{-3}$  in Baoding,  $81 \mu\text{g m}^{-3}$  in Baoding, and  $2.7 \mu\text{g m}^{-3}$  in Baoding, respectively. The maximum  $\text{PM}_{2.5}$  concentrations in Beijing, Hengshui and Baoding were  $266 \mu\text{g m}^{-3}$  (13:00, January 28),  $286 \mu\text{g m}^{-3}$  (11:00, January 26), and  $641 \mu\text{g m}^{-3}$  (3:00, January 25), respectively. The concentrations of  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$ , organics and  $\text{K}^+$  were the main components of fine particles ( $\text{PM}_{2.5}$ ) in Beijing and Hengshui, while organics,  $\text{K}^+$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{NH}_4^+$  were the main components of  $\text{PM}_{2.5}$  in Baoding

The meteorological data with a high spatial resolution of  $0.5^\circ \times 0.5^\circ$  required for the Hybrid single-particle Lagrangian integral trajectory (HYSPLIT) model were obtained from the NOAA website (<ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas0p5>).

### The HYSPLIT model

We used the HYSPLIT model to calculate backward trajectories and concentration weighted trajectories (CWT). The backward trajectory provides a transport path for air pollutants, which is used to determine whether the high-concentration pollutants in one location originate from the

pollution transport of another location (Draxler and Hess 1998). This study calculated the 48 h backward trajectory of  $\text{PM}_{2.5}$  at a height of 100 m with the starting calculation time points for each day during the study period on 00:00, 03:00, 06:00, 09:00, 12:00, 15:00, 18:00 and 21:00 (UTC time). One back trajectory for one monitoring station were calculated each time. Trajectory cluster analysis based on the trajectory space similarity is used to group a large number of trajectories. In this work, we used the angular distance to do cluster analysis which is based on the trajectory space similarity to group a large number of trajectories. This is mainly because the trajectories can be applied to locate the directions and sources from which the air masses reaching

the receptor site had transported (Yan et al. 2015). The CWT method assigns a weighted concentration to each grid cell, which is obtained by averaging the sample concentrations associated with the trajectory passing through the grid cell (Reizer and Orza 2018). This method calculates the  $CWT_{ij}$  value in each grid cell ( $i, j$ ) as follows (Hsu et al. 2003; Yan et al. 2015):

$$CWT_{ij} = \frac{1}{\sum_{l=1}^M \tau_{ijl}} \sum_{l=1}^M C_l \tau_{ijl}$$

where  $l$  and  $M$  represent the index and the total number of trajectories in the grid cell, respectively.  $C_l$  is the concentration detected at the receptor position on the arrival of trajectory  $l$ .  $\tau_{ijl}$  is the time that the trajectory  $l$  spent in the grid cell ( $i, j$ ). A high  $CWT_{ij}$  value indicates that the air mass with attributions of high potential pollution has a significant contribution to the receptor site (Yan et al. 2015). In order to find the specific location of the pollution sources, the common pollution source area calculation method of Chen et al (2020) was adopted to determine the common source area of  $PM_{2.5}$  in the three cities during the study period.

### Calculation of meteorological parameters

This study used mixed layer height (MLH), 10 m wind speed (WS), relative humidity (RH) and static weather index to represent the meteorological conditions in the three recipient cities. The MLH values can be calculated using an empirical method as follows (Nozaki 1973; Beyrich 1997):

$$MLH = \frac{121}{6} (6 - P)(T - T_d) + \frac{0.169P(U_z + 0.257)}{12f \ln(Z/z_0)}$$

$$f = 2\Omega \sin \varphi$$

where  $P$  is Pasquill's stability coefficient,  $T$  is the surface temperature,  $T_d$  is the dew point temperature,  $U_z$  is the average wind speed (m/s) at the height of  $Z$  ( $Z = 10$  m),  $f$  is the Coriolis parameter ( $s^{-1}$ ),  $z_0$  is the surface roughness (0.03–0.2 m for rural area, 0.8–2 m for urban area, and 1.4 m is used in this study),  $\Omega$  is the Earth's angular velocity ( $7.29 \times 10^{-5}$  rad  $s^{-1}$ ), and  $\varphi$  is the latitude value. Pasquill divides atmospheric stability into six levels from A to F (Pasquill 1961), and the six levels from A to F are replaced by 1–6 numbers in this study.

By correlating the air pollutant levels with a combination of meteorological factors in a specific region or city, Zhang et al (2017) developed the stable weather index (SWI) which included the advection, vertical diffusion and humidity and other meteorological factors (Gong et al. 2021; Liu et al. 2021). A higher SWI value means a weaker diffusion of air pollutants. The detailed information about the calculation

method of SWI can be found in Zhang et al (2017). In this study, the SWI values were calculated and used to assess the atmospheric stable states of the meteorological conditions.

### Modelling meteorological and chemical parameters

The offline weather research and forecasting (WRFv3.9.1)-Community Multiscale Air Quality Modeling (CMAQv5.3.2) model was used to simulate spatiotemporal variations in meteorological and chemical parameters. Model configurations are listed in Table S1. In this study, the Carbon Bond 6 (CB6r3) scheme and AERO7 module were responsible for gas and aerosol chemistry simulations, respectively. The CMAQ version 5.3.2 (released in 2020) is the latest version and its major science advances in detail can be found in Murphy et al. (2021), Appel et al. (2021) and US EPA (2020). A brief summary relevant to the present study is presented here. Major science advances in the new aerosol module (AERO7) includes significant updates to secondary organic aerosol (SOA) chemistry, updates for chlorine, bromine, iodine chemistry, and halogen chemistries, and the addition of dimethyl sulfide (DMS) chemistry in the CB6r3 chemical mechanism (Appel et al. 2021). The CB6r3 is the most recent version of CB implemented in CMAQ53 with 4 heterogeneous reactions, 38 gas-phase reactions, and 44 gas-phase reactions and 10 heterogeneous reactions for iodine species, and 5 aqueous-phase reactions for bromine species (Appel et al. 2021).

The model domain covering China and a portion of East Asia with a horizontal resolution of  $12 \text{ km} \times 12 \text{ km}$  and  $345 \times 395$  grid cells is shown in Fig. S5. The WRFv3.9.1 (Skamarock et al. 2008) model was used to provide meteorological fields for chemical simulations. The configurations and components for the WRF-CMAQ model used in this study are the same as those in Yu et al. (2014) and are summarized in Table S1. The anthropogenic emissions for China were generated by the Emission Inventory of Air Benefit and Cost and Attainment Assessment System (EI-ABaCAS) developed by Tsinghua University (Zheng et al. 2018). The EI-ABaCAS includes 16 anthropogenic sectors: fertilizer application, livestock, domestic bio-fuel, combustion, fossil fuel, solvent and other use, industry combustion, open burning, power plant, cement steel, other industry process, industry solvent use, road and non-road transport. The natural sources for biogenic emissions were calculated inline using the Biogenic Emission Inventory System version 3.14 (BEISv3.14).

## Results and discussion

### Air pollutants and meteorological conditions during the severe haze period

Figure 1 shows the time series of the concentrations of  $\text{PM}_{10}$ ,  $\text{O}_3$ ,  $\text{NO}_2$ ,  $\text{SO}_2$ ,  $\text{CO}$ ,  $\text{PM}_{2.5}$  and chemical components of  $\text{PM}_{2.5}$  in the three cities during the study period. The results show that the changing trends of air pollutants in the three cities were obviously different, indicating different  $\text{PM}_{2.5}$  formation mechanisms. The highest levels were  $692 \mu\text{g m}^{-3}$  for  $\text{PM}_{10}$  in Baoding,  $125 \mu\text{g m}^{-3}$  for  $\text{O}_3$  in Beijing,  $90 \mu\text{g m}^{-3}$  for  $\text{NO}_2$  in Baoding,  $81 \mu\text{g m}^{-3}$  for  $\text{SO}_2$  in Baoding, and  $2.7 \text{ mg m}^{-3}$  for  $\text{CO}$  in Baoding. The maximum  $\text{PM}_{2.5}$  concentrations in Beijing, Hengshui and Baoding were  $266 \mu\text{g m}^{-3}$  at 13:00 January 28 in Beijing,  $286 \mu\text{g m}^{-3}$  at 11:00, January 26 in Hengshui, and  $641 \mu\text{g m}^{-3}$  at 3:00, January 25 in Baoding. Baoding experienced a one-day severe pollution process from January 24 to 25, during which  $\text{PM}_{10}$ ,  $\text{NO}_2$  and  $\text{SO}_2$  all reached their peak values. The  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Cl}^-$  and  $\text{Ca}^{2+}$  ions from fireworks as indicated in Fig. 1 increased sharply after January 24, and reached their peaks on January 25, being consistent with the fact that the air pollution emissions from fireworks and firecrackers flooded into the air on the New Year's Eve. This indicates that the fireworks and firecrackers still affected the formation of haze in the North China Plain during the lockdown period. In addition to being derived from fireworks and firecrackers,  $\text{K}^+$  was also a tracer of biomass combustion (Cheng et al. 2013). The increases of  $\text{K}^+$  also show that the suburban biomass burning contributed significantly to the  $\text{K}^+$  in the three cities.

In Beijing, after experiencing two peaks and valleys from January 25 to January 26,  $\text{PM}_{2.5}$  rebounded after 12 noon on January 26 and then remained at high concentrations above  $147 \mu\text{g m}^{-3}$ . This shows that after January 26, when local emissions were almost unchanged, regional transport made a certain contribution to Beijing's  $\text{PM}_{2.5}$ . The main component of  $\text{PM}_{2.5}$  in Beijing was water soluble inorganic ions (WSIIs) with an average proportion of 53.6% for  $\text{PM}_{2.5}$ . The average concentration of  $\text{NO}_3^-$  in Beijing was  $32.0 \mu\text{g m}^{-3}$ , accounting for 12.1–29.7% of  $\text{PM}_{2.5}$  with an average proportion of 20.9%, while  $\text{SO}_4^{2-}$  in Beijing ranged from  $10 \mu\text{g m}^{-3}$  to  $27.8 \mu\text{g m}^{-3}$ , accounting for 12.2% of  $\text{PM}_{2.5}$  on average. The average relative humidity in Beijing was  $60.7 \pm 16.4\%$  (Fig. S2) during the study period, which promoted the formation of a large amount of  $\text{SO}_4^{2-}$  from the heterogeneous chemical reactions and liquid phase oxidations of  $\text{SO}_2$ . The concentrations of  $\text{NH}_4^+$  in Beijing ranged from  $8.9$  to  $21.9 \mu\text{g m}^{-3}$ , with an average proportion of 10.4% for  $\text{PM}_{2.5}$ . Since emissions from motor vehicles and industries have declined to a certain

extent under the COVID-19 lockdown (Le et al. 2020), residential heating and non-stop industrial productions in Beijing emitted a certain amount of  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  precursors.

In Hengshui, there were four peak values of  $\text{PM}_{2.5}$  concentrations in the early stage with decreasing trends in the later stage. The average percentage of WSIs in  $\text{PM}_{2.5}$  in Hengshui was 71.6%. Similar to Beijing, the main components of  $\text{PM}_{2.5}$  in Hengshui were  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$ , with average mass concentrations of  $36.1 \pm 20.4 \mu\text{g m}^{-3}$ ,  $22.3 \pm 13 \mu\text{g m}^{-3}$ , and  $16.9 \pm 7.6 \mu\text{g m}^{-3}$ , respectively. The proportion of WSIs in  $\text{PM}_{2.5}$  changed over time. For example, on January 25, the main component of  $\text{PM}_{2.5}$  was  $\text{SO}_4^{2-}$  and on January 26, the main component of  $\text{PM}_{2.5}$  turned to be  $\text{NO}_3^-$ , which peaked at night on January 26. Considering that the local emissions were relatively unchanged, this indicates that the contributions of regional transports to  $\text{PM}_{2.5}$  in Hengshui were considerable, and the formation mechanism of  $\text{PM}_{2.5}$  in Hengshui has changed. Figure S2 shows that the changes of relative humidity in Hengshui on January 25 were consistent with those of  $\text{PM}_{2.5}$ . It should be noted that there were diurnal differences in the concentrations of  $\text{NO}_3^-$  in Hengshui with the average concentrations of  $27.2 \mu\text{g m}^{-3}$  (daytime) and  $40.7 \mu\text{g m}^{-3}$  (night). The higher night  $\text{NO}_3^-$  concentrations in Hengshui might be affected by external sources and stable weather conditions.

In Baoding, unlike Beijing and Hengshui, organic matter dominated  $\text{PM}_{2.5}$ , accounting for  $25 \pm 6.4\%$  on average. The average concentrations of chemical components of  $\text{PM}_{2.5}$  in Baoding in the descending order were  $60.3 \pm 19 \mu\text{g m}^{-3}$  for organic matter (OM),  $35.2 \pm 20.1 \mu\text{g m}^{-3}$  for  $\text{K}^+$ ,  $33.9 \pm 20.4 \mu\text{g m}^{-3}$  for  $\text{SO}_4^{2-}$ ,  $26.6 \pm 7.0 \mu\text{g m}^{-3}$  for  $\text{NO}_3^-$ ,  $25.5 \pm 6.8 \mu\text{g m}^{-3}$  for  $\text{NH}_4^+$ ,  $16.6 \pm 11.7 \mu\text{g m}^{-3}$  for  $\text{Cl}^-$ ,  $10.7 \pm 3.3 \mu\text{g m}^{-3}$  for elemental carbon (EC),  $8.3 \pm 3.4 \mu\text{g m}^{-3}$  for  $\text{Ca}^{2+}$ ,  $3.9 \pm 1.4 \mu\text{g m}^{-3}$  for  $\text{Mg}^{2+}$ , and  $2.9 \pm 0.5 \mu\text{g m}^{-3}$  for  $\text{Na}^+$ .  $\text{PM}_{2.5}$  in Baoding peaked on January 25 with sharp drops in the next 24 h by  $380 \mu\text{g m}^{-3}$  because of the diffusion caused by the increases of the mixed layer height (MLH, Fig. S2). The concentration of organic matter (OM) in Baoding was extremely high, indicating that organic gas precursors played a key role in the formation of  $\text{PM}_{2.5}$  in Baoding. Note that the OM concentrations were still high after Jan 25 in Baoding because of the effects of the regional transports. The very high concentrations of  $\text{K}^+$  and  $\text{Cl}^-$  in Baoding's  $\text{PM}_{2.5}$  indicate substantial contributions from the emissions of fireworks and firecrackers. This is consistent with the findings of Tang et al (2016), who found that the  $\text{K}^+$ ,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$ , and  $\text{NO}_3^-$  concentrations in Tianjin's  $\text{PM}_{2.5}$  during the firework were 65.8, 42.9, 38.9, 8.7, and  $1.9 \mu\text{g m}^{-3}$ , respectively, and that the  $\text{K}^+$  and  $\text{Cl}^-$  concentrations during the peak period were 23.5 and 15.5 times higher than those during Chinese Lunar New

Year, respectively. Since fireworks contained charcoal and organic materials used as adhesives, they were responsible for elevated OC and EC concentrations during the firework and firecracker events (Tsai et al. 2012; Kong et al. 2015). Since potassium compounds, e.g.,  $\text{KNO}_3$ ,  $\text{KClO}_3$ ,  $\text{KClO}_4$ ,  $\text{K}_2\text{CrO}_4$ , and  $\text{K}_2\text{Cr}_2\text{O}_7$ , in black powder are used as the main oxidizers, the corresponding chemical reactions, e.g.,  $2\text{KClO}_3 = 2\text{KCl} + 3\text{O}_2$  and  $\text{KClO}_4 = \text{KCl} + 2\text{O}_2$ , can lead to high concentrations of  $\text{K}^+$  and  $\text{Cl}^-$  during the firework and firecracker events (Azhangurajan et al. 2011; Tang et al. 2016).

Figure S2 shows the time series of hourly meteorological elements in the three cities. The meteorological conditions in the three cities during the study period can be summarized as low MLH and wind speed (WS) but high stable weather index (SWI) and relative humidity. For example, the MLH mean values in Beijing, Hengshui, and Baoding were  $448.2 \pm 294.1$ ,  $497.3 \pm 209.1$ , and  $330.0 \pm 232.2$  m, respectively. Comparing to the annual average MLH values in Beijing and Shijiazhuang being  $594 \pm 183$  m and  $464 \pm 183$  m, respectively (Zhu et al. 2018), the MLH values in the three cities during the study period can be considered as relatively low. The average WS in Beijing, Hengshui, and Baoding were  $1.4 \pm 0.8$ ,  $0.8 \pm 0.5$ , and  $0.8 \pm 0.8$   $\text{m s}^{-1}$ , respectively. The low wind speeds in these three cities limit the diffusion of local sources. On the other hand, the maximum relative humidity in Beijing, Hengshui and Baoding were 83%, 92% and 97%, respectively. The SWI ranges in Beijing, Hengshui and Baoding were 8–19, 11–20 and 9–21, respectively. The high SWIs in the three cities indicate that the meteorological conditions were stable during the study period. In conclusion, the stable meteorological conditions in the three cities exacerbated the formation of the haze.

### Regional transport impacts based on the HYSPLIT analysis

Table 1 summarizes the average concentrations of  $\text{PM}_{2.5}$  and its chemical components, and the percentages for each cluster and three cluster trajectories for each receptor city are shown in Fig. S3. The results of all backward trajectories in Fig. S3 indicate that most of the air masses came from Inner Mongolia, Liaoning, Hebei and Tianjin.

For Beijing, Cluster 1, which accounted for 54.1% and carried 51.0% of the pollution trajectories, transported from the neighboring areas to Beijing through a small area in northern Hebei province. Cluster 1 had the highest concentration of  $\text{PM}_{2.5}$  ( $177.3 \pm 61.2$   $\mu\text{g m}^{-3}$ ), which was the most important transport path for  $\text{PM}_{2.5}$  in Beijing. Cluster 2 (3) accounted for 37.7% (8.2%) and carried 39.5% (9.5%) of the pollution trajectories. According to the concentrations of chemical components, Cluster 2 carries the most  $\text{K}^+$  and

$\text{Cl}^-$  probably generated by firework because Cluster 2 was not a cross-sea transport as shown in Fig. S3.

For Hengshui, all cluster trajectories were cross-sea transports in the northeast direction with obvious long-distance transport characteristics as indicated in Fig. S3. Cluster 1 with the longest transport path came from western Inner Mongolia and Liaoning Province, accounting for 43.8% and carrying 44.3% of the pollution trajectories (see Table 1). Clusters 2 and 3 passed through western Liaoning, southern Tianjin, and northern Shandong. The  $\text{PM}_{2.5}$  concentrations ( $157.6$   $\mu\text{g m}^{-3}$ ) carried by the air mass in Cluster 2 was the highest. The  $\text{Na}^+$  concentrations in Cluster 2 were the highest, further indicating that cross-sea transports had a certain influence on the formation of  $\text{PM}_{2.5}$  in Hengshui.

For Baoding, except for Cluster 3, the other two Clusters all passed through the western Liaoning and Inner Mongolia. Cluster 3, which accounted for 21.1% and carried  $\text{PM}_{2.5}$  with an average concentration of  $216.7$   $\mu\text{g m}^{-3}$ , moved around Baoding, indicating a great impact of local pollution sources on  $\text{PM}_{2.5}$  in Baoding. The percentages of Clusters 1 and 2 were 45.4% and 33.5%, respectively, and Cluster 2 carried the highest concentration of  $\text{PM}_{2.5}$  ( $295.9$   $\mu\text{g m}^{-3}$ ).

To further study the impact of regional transports on the formation of haze, Fig. 2 shows the 48-h backward trajectories in three cities, superimposed by the observed  $\text{PM}_{2.5}$  concentrations over the North China Plain every 12 h within 48 h. As can be seen, the spatial variations of  $\text{PM}_{2.5}$  in the three cities were consistent with those of the monitoring stations on the transport paths. The pollution incidents in Beijing were a typical full-time severe pollution event.  $\text{PM}_{2.5}$  concentrations in Beijing were higher at 13:00 on January 28 than those in other four time points. From January 26 to January 28, there was a rising process of  $\text{PM}_{2.5}$  in the western cities of Liaoning, which were on the pollution transport path. The concentrations of  $\text{PM}_{2.5}$  in Beijing remained at a high level above  $147$   $\mu\text{g m}^{-3}$ , indicating that  $\text{PM}_{2.5}$  in western Liaoning was imported into Beijing along the pollution transport path. Figure 2 indicates that nearby cities located in the pollution transport path had a greater impact on the increases of Beijing's  $\text{PM}_{2.5}$ . Figure 2 also shows that the spatial variations of  $\text{PM}_{2.5}$  concentrations in Hengshui and Baoding were consistent with those at the monitoring stations on the transport path. For the pollution event from January 24 to January 26, the cities in Liaoning province located on the northeast transport path for Hengshui experienced a substantial increase of  $\text{PM}_{2.5}$  within 24 h from 11:00 on January 24 to 11:00 on January 25. The  $\text{PM}_{2.5}$  concentrations in Hengshui began to rise from 23:00 on January 25, and reached a peak on January 26. Baoding's  $\text{PM}_{2.5}$  experienced a sharp rise from 15:00 on January 24. At the same time, the concentrations of  $\text{PM}_{2.5}$  in cities of western Liaoning and Hebei Provinces on the transport path for Baoding also experienced the same rising processes.

**Table 1** Number of backward trajectories and average concentrations of PM<sub>2.5</sub> and chemical components for different clusters in Beijing, Hengshui, and Baoding during the study period

	Beijing			Hengshui			Baoding		
	Cluster 1	Cluster 2	Cluster 3	Cluster 1	Cluster 2	Cluster 3	Cluster 1	Cluster 2	Cluster 3
Number of trajectories (percent)	309 (54.1)	215 (37.7)	47 (8.2)	84 (43.8)	72 (37.5)	36 (18.8)	129 (45.4)	95 (33.5)	60 (21.1)
PM <sub>2.5</sub> [ $\mu\text{g m}^{-3}$ ] for all trajectories	177.3 $\pm$ 61.2	133.9 $\pm$ 64.0	136.2 $\pm$ 50.7	115.6 $\pm$ 56.1	140.0 $\pm$ 49.9	92.5 $\pm$ 56.4	168.0 $\pm$ 94.1	275.9 $\pm$ 142.8	206.1 $\pm$ 57.5
Number of polluted trajectories*	214 (51)	166 (39.5)	40 (9.5)	62 (44.3)	60 (42.9)	18 (12.9%)	104 (42.1%)	87 (35.2%)	56 (22.7%)
PM <sub>2.5</sub> [ $\mu\text{g m}^{-3}$ ] for polluted trajectories	150.4 $\pm$ 41.1	157.3 $\pm$ 53.3	150.7 $\pm$ 39.6	138.1 $\pm$ 47.5	157.6 $\pm$ 33.0	132.2 $\pm$ 56.0	196.3 $\pm$ 82.6	295.9 $\pm$ 132.2	216.7 $\pm$ 42.7
**Ca <sup>2+</sup>	1.4 (0.1)	0.8 (0.04)	0.8 (0.04)	2.6 (0.9)	2.8 (0.7)	2.5 (1.3)	12.5 (0.3)	10.3 (0.3)	12.5 (0.3)
Mg <sup>2+</sup>	1.3 (0.3)	1.3 (0.3)	1.2 (0.3)	1.8 (0.8)	2.4 (0.5)	1.8 (0.7)	3.2 (1.0)	3.4 (0.9)	3.2 (1.3)
K <sup>+</sup>	9.5 (4.4)	9.7 (5.8)	9.4 (5.4)	9.3 (4.7)	12.6 (4.7)	7.0 (3.3)	22.5 (7.6)	28.0 (7.0)	19.8 (6.4)
Na <sup>+</sup>	1.2 (0.4)	1.0 (0.5)	1.1 (0.4)	2.3 (0.5)	3.3 (0.5)	2.1 (0.4)	2.8 (0.5)	2.8 (0.4)	2.8 (0.6)
NH <sub>4</sub> <sup>+</sup>	18.3 (10.1)	14.7 (9.9)	15.0 (10.2)	15.0 (13.8)	19.6 (12.0)	12.6 (15.2)	27.3 (13.7)	27.0 (8.6)	28.8 (12.7)
SO <sub>4</sub> <sup>2-</sup>	22.9 (12.6)	18.7 (12.5)	18.6 (12.6)	18.0 (15.9)	25.0 (14.7)	14.7 (16.9)	23.7 (11.7)	31.2 (9.7)	25.8 (11.5)
NO <sub>3</sub> <sup>-</sup>	40.0 (22.0)	30.4 (20.2)	31.3 (21.1)	30.7 (28.6)	39.9 (23.7)	26.7 (32.5)	30.1 (15.6)	28.7(9.3)	30.7 (13.5)
Cl <sup>-</sup>	6.6 (3.7)	7.1 (5.1)	6.7 (4.8)	6.9 (5.9)	9.5 (5.9)	5.1 (5.7)	9.0 (4.2)	13.6 (4.2)	10.2 (4.4)
OM	15.5 (8.7)	14.8 (10.6)	14.7 (10.4)	17.5 (16.0)	22.2 (13.7)	14.4 (17.4)	51.0 (24.4)	57.9 (18.2)	56.7 (24.8)
EC	6.5 (4.0)	6.3 (4.5)	6.2 (4.4)	5.0 (4.5)	6.7 (4.7)	3.8 (4.4)	9.6 (4.7)	10.1 (4.2)	10.1 (4.3)

For Beijing, Cluster 1 had the highest concentration of PM<sub>2.5</sub> (177.3 $\pm$ 61.2  $\mu\text{g m}^{-3}$ ), which was the most important transport path for PM<sub>2.5</sub> in Beijing

According to the concentrations of chemical components, Cluster 2 carries the most K<sup>+</sup> and Cl<sup>-</sup> probably generated by firework because Cluster 2 was not a cross-sea transport as shown in Fig. S3. For Hengshui, the PM<sub>2.5</sub> concentrations (157.6  $\mu\text{g m}^{-3}$ ) carried by the air mass in Cluster 2 was the highest. The Na<sup>+</sup> concentrations in Cluster 2 were the highest, further indicating that cross-sea transports had a certain influence on the formation of PM<sub>2.5</sub> in Hengshui. For Baoding, Cluster 3 carried PM<sub>2.5</sub> with an average concentration of 216.7  $\mu\text{g m}^{-3}$ , moved around Baoding, indicating a great impact of local pollution sources on PM<sub>2.5</sub> in Baoding

\*Polluted trajectories: trajectories with hourly PM<sub>2.5</sub> concentrations higher than 75  $\mu\text{g m}^{-3}$ . SD: standard deviation

\*\*Values of chemical components are: average mass concentrations ( $\mu\text{g m}^{-3}$ ) and the fraction of chemical component (%) in parentheses for PM<sub>2.5</sub>

In summary, the PM<sub>2.5</sub> concentrations of the three receptor cities mainly depend on the cities located in the pollution transport paths for them. The northeast transport path, especially through Liaoning province, had the substantial impact on PM<sub>2.5</sub> in the three recipient cities. The previous studies found that the regional transports were mainly a southerly transport path for NCP (Zhu et al. 2016; Du et al. 2020; Lv et al. 2020). However, this study shows that the northeast transport path dominates the regional transport for the haze formation in the North China Plain.

### Identification of common source areas for pollution

The concentrations of PM<sub>2.5</sub> in the receptor cities were combination effects of local emissions and regional transports. Figure 3 shows the concentration weighted trajectories (CWT) for the entire and severe pollution periods and the

CWT for the light, medium and heavy pollution periods are shown in Fig. S4. Figure 3 indicates that different cities had different degrees of contributions to PM<sub>2.5</sub> in the three cities. The regions with the CWT values higher than 250  $\mu\text{g m}^{-3}$  (see Fig. 3) included Chaoyang and Huludao in Liaoning province, Qinhuangdao, Tangshan, Langfang and Chengde in Hebei province, and Tianjin, all of which contributed high concentrations of PM<sub>2.5</sub> to Beijing. For Hengshui, the potential sources of pollution during the severe pollution period were Panjin, Anshan and Yingkou in Liaoning province. It was worth noting that Liaoning province had a large amount of PM<sub>2.5</sub> input for Hengshui for all the pollution periods. For Baoding, Cangzhou, Langfang, Chaoyang, Jinzhou and Tianjin were potential source areas during the heavy pollution period. Note that some of the air masses from Liaoning were transported across the Bohai Sea, which is located in the east of the North China Plain.

**Fig. 2** Maps of 48 h polluted backward trajectories overlaid with the observations of  $PM_{2.5}$  concentrations ( $\mu g \cdot m^{-3}$ ) at different times (in the format of YYYYMMDD\_HH). The spatial variations of  $PM_{2.5}$  in the three cities were consistent with those of the monitoring stations on the transport paths. The pollution incidents in Beijing were a typical full-time severe pollution event. As can be seen, the  $PM_{2.5}$  concentrations of the three recipient cities partly depend on the cities located on pollution transport paths for them. The northeast transport path through Liaoning province had the greatest impact on  $PM_{2.5}$  in the three recipient cities. The results show that the northeast transport path dominates the regional transport for the haze formation in NCP

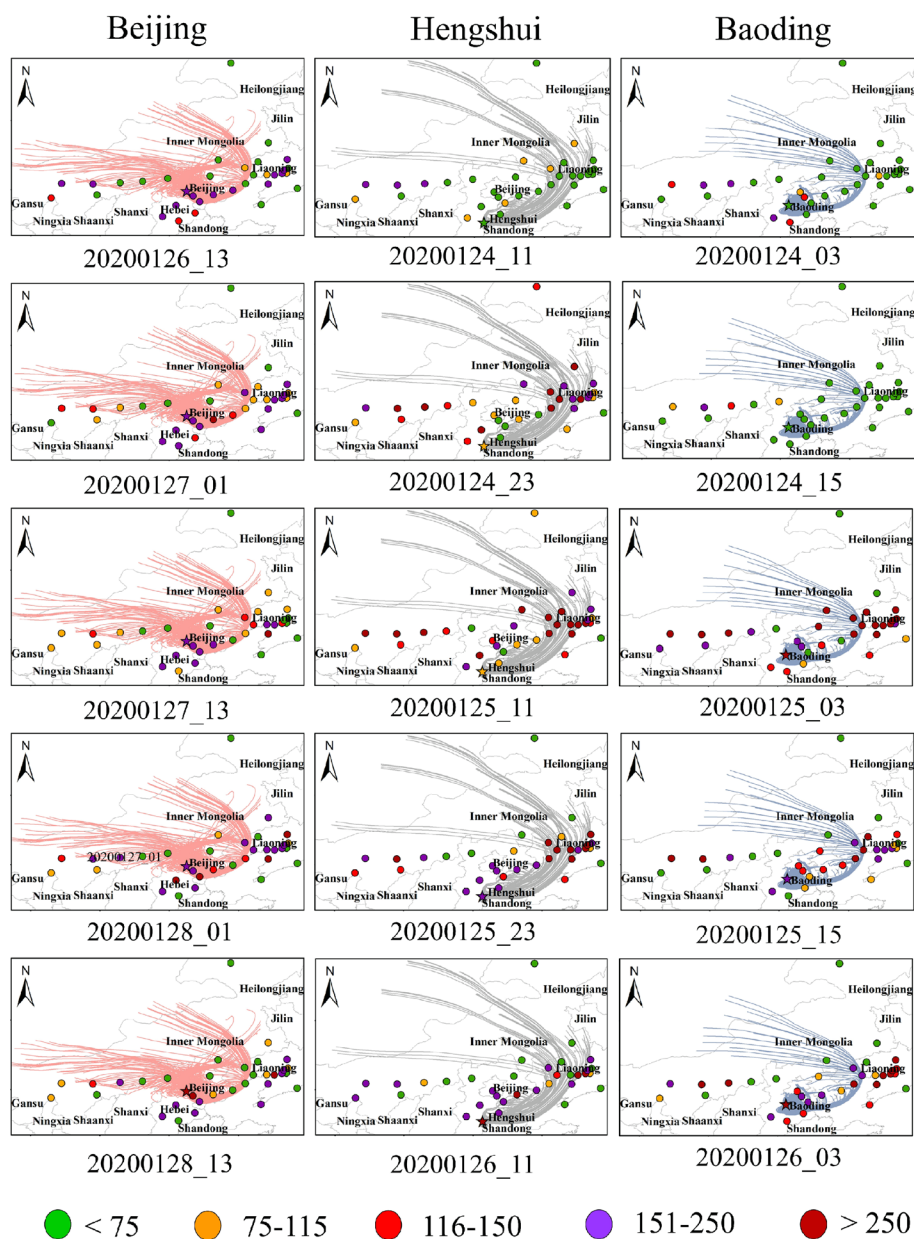
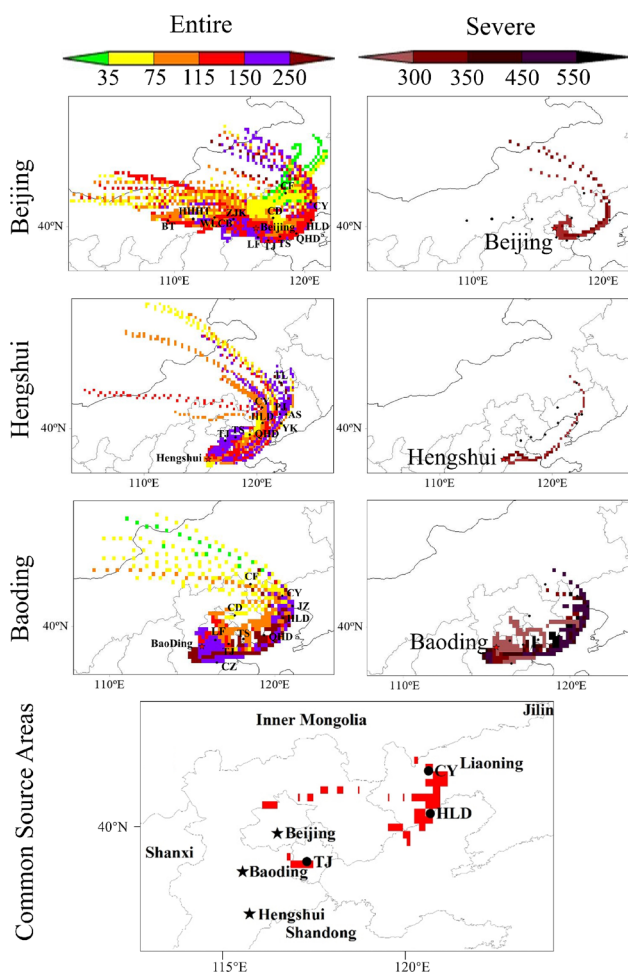


Figure 3 shows the common source areas of  $PM_{2.5}$  in the three cities during the study period. Controlling the common source area is an effective choice for air pollution control (Yu et al. 2018). As can be seen, western Liaoning, northern Hebei, and southern Tianjin were the common source areas for  $PM_{2.5}$  pollution events in the three recipient cities. This is consistent with the fact that Hebei province is the largest industrial cluster in China (Wang et al. 2014) and Liaoning province is a region with industry as its main industry (Shi et al. 2020). Although a temporary industrial shutdown was implemented to curb the spread of COVID-19, industrial emissions have not dropped significantly due to the fact that the main industrial sectors have not yet been completely closed (Wang et al. 2020a, b).

### Regional transport impacts based on the WRF-CMAQ model simulations

To determine if the northeast transport path had the substantial impact on  $PM_{2.5}$  in the three receptor cities during the study period, the WRF-CMAQ model was applied to simulate this regional severe haze episode in the retrospective mode. The concentration weighted trajectories (CWT) values for  $PM_{2.5}$  were calculated on the basis of the air mass back trajectories and their associated  $PM_{2.5}$  concentrations observed at the three receptor sites to locate the source areas with the largest potential contributions to high concentrations. Figure S6 shows the spatial distributions of the CWT values created by combining all results at the three





**Fig. 3** Concentration weighted trajectory (CWT) maps of  $\text{PM}_{2.5}$  for Beijing, Hengshui, Baoding during the entire and severe haze periods ( $\text{PM}_{2.5} \geq 250 \mu\text{g m}^{-3}$ ), and common source areas of  $\text{PM}_{2.5}$  ( $\text{PM}_{2.5} \geq 75 \mu\text{g m}^{-3}$ ) during the entire study period (24th January–29th January, 2020). The full names of the abbreviation of cities in the figure are as follows: AS-Anshan, BT-Baotou, CD-Chengde, CF-Chifeng, CY-Chaoyang, CZ-Cangzhou, HHTH-Hohhot, HLD-Huludao, JZ-Jinzhou, LF-Langfang, PJ-Panjing, QHD-Qinhuangdao, TJ-Tianjin, TL-Tongliao, TS-Tangshan, WLCB-Wulanchabu, YK-Yingkou, ZJK-Zhangjiakou. For Hengshui, the potential sources of pollution during the severe pollution period were Panjin, Anshan and Yingkou in Liaoning province. For Baoding, Cangzhou, Langfang, Chaoyang, Jinzhou and Tianjin were potential source areas during the heavy pollution period

cities during the study period. Followed Yu et al. (2016), an emission reduction scenario was created on the basis of the CWT values in Fig. S6 by removing all anthropogenic emissions for grid cells with  $\text{CWT} \geq 75 \mu\text{g m}^{-3}$ , while those will remain uncontrolled for grid cells with  $\text{CWT} < 75 \mu\text{g m}^{-3}$ . Figures S7 and S8 show the comparisons of the spatial distributions of anthropogenic emissions for primary  $\text{PM}_{2.5}$ ,  $\text{NH}_3$ ,  $\text{NO}_x$ ,  $\text{SO}_2$ ,  $\text{VOC}_s$ , and  $\text{POC}$  between the base and reduction cases. Figure S9 shows the daily mean comparisons of the simulation results for the spatial distributions of  $\text{PM}_{2.5}$

concentrations for the base and reduction cases and their differences for each day from January 24 to 29, 2020. The maps of the  $\text{PM}_{2.5}$  reduction amounts between the base and reduction cases overlaid with the wind fields for different times in Fig. S10 clearly show that the prevailing northeasterly winds during study period brought air pollution continuously from the northeast areas to the North China Plain regions, causing the severe haze formation there. Comparisons of the time series of  $\text{PM}_{2.5}$  and its chemical components concentrations in the three receptor cities for the base and reduction cases in Fig. S11 also show that the peak values for all these pollutants in the reduction cases in the three receptor cities were effectively decreased by more than 60%. As summarized in Tables S2, S3 and S4, the  $\text{PM}_{2.5}$ ,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , OM, and  $\text{SO}_4^{2-}$  concentrations in the reduction case decreased at most by  $-106.73 \mu\text{g m}^{-3}$ ,  $-8.93 \mu\text{g m}^{-3}$ ,  $-25.96 \mu\text{g m}^{-3}$ ,  $-13.75 \mu\text{g m}^{-3}$  and  $-5.51 \mu\text{g m}^{-3}$  in Beijing, respectively, while they were  $-144.38 \mu\text{g m}^{-3}$ ,  $-12.01 \mu\text{g m}^{-3}$ ,  $-34.15 \mu\text{g m}^{-3}$ ,  $-21.77 \mu\text{g m}^{-3}$  and  $-7.79 \mu\text{g m}^{-3}$  in Hengshui, respectively, and were  $-148.32 \mu\text{g m}^{-3}$ ,  $-12.21 \mu\text{g m}^{-3}$ ,  $-35.86 \mu\text{g m}^{-3}$ ,  $-23.68 \mu\text{g m}^{-3}$  and  $-8.56 \mu\text{g m}^{-3}$  in Baoding, respectively. The mean  $\text{PM}_{2.5}$ ,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , OM, and  $\text{SO}_4^{2-}$  concentrations in the reduction case could decrease by more than 60% in the three receptor cities relative to the base case. The model sensitive simulations results highlight the fact that regional severe haze in North China Plain during the coronavirus lockdown was caused mainly by the anthropogenic emissions through the northeast transport paths.

## Conclusion

This study investigated the causes of the regional severe haze over the North China Plain during the COVID-19 lockdown period on the basis of chemical observations in the three cities (Beijing, Baoding and Hengshui) and model simulations. The concentrations of  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$ , organic matter and  $\text{K}^+$  were found to be the main components of  $\text{PM}_{2.5}$  in Beijing and Hengshui, while they were OM and  $\text{K}^+$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{NH}_4^+$  in Baoding. The results indicate that the emissions from firework, firecrackers, residential heating, and non-stop industries under the stable meteorological conditions with low wind speed, low mixing layer height, and high relative humidity caused this severe haze pollution in North China Plain. The results of backward trajectories in the three cities indicate that most of the air masses influencing the cities came from Inner Mongolia, Liaoning, Hebei and Tianjin. For Hengshui, all cluster trajectories are cross-sea transports in the northeast direction with obvious long-distance transport characteristics by passing the western Inner Mongolia and Liaoning Province. For Baoding, the two cluster trajectories also passed through the western

Liaoning and Inner Mongolia. It was found that the common source areas for the haze formation in the three cities were located in western Liaoning, northern Hebei, and southern Tianjin as the common source cities. The stagnant airflow and uninterrupted emissions from non-stop industries (e.g., power plants and petrochemical facilities) can still cause regional severe haze formation with high humidity to promote aerosol heterogeneous chemistry. The results of the cluster trajectories and model simulations show that different from the previous studies that mainly found a southerly transport path of air pollution for North China Plain, the northeast transport path dominates the regional transport for this haze formation. This indicates that the transport paths can be variable for a city for different haze events in regional scale. This implies that the variabilities of transport paths should be taken into account in the regulation of air pollution in the North China Plain, especially in the case of short-term severe pollutions.

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