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- Evaluating the Recent "2+26" Regional Strategy for Air Quality
- Improvement During Two Orange Air Pollution Alerts in Beijing: 2
- variations of PM_{2.5} concentrations, source apportionment, and the 3
- relative contribution of local emission and regional transport 4
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Abstract 26

- 27 To comprehensively evaluate the effects of the recent "2+26" regional strategy for air quality
- 28 improvement, we compared the variations in PM2.5 concentrations in Beijing during four pollution
- 29 episodes with different emission-reduction strategies. The "2+26" strategy implemented in March
- 30 2018 led to a mean PM_{2.5} concentrations of 16.43% lower than that during the pollution episode in
- March 2013, when no specific emission-reduction measures were in place. The same "2+26" 31
- 32 strategy implemented in November 2017 led to a mean PM_{2.5} concentrations of 32.70% lower than
- 33 that during the pollution episode in November 2016, when local emission-reduction measures were implemented. The results suggested that the effects of the "2+26" regional
- emission-reduction measures on PM2.5 reductions were influenced by a diversity of factors and 35
- 36 could differ significantly during specific pollution episodes. Furthermore, we found the

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- 37 proportions of sulfate ions decreased significantly and nitrate ions were the dominant PM2.5
- 38 components during the two "2+26" orange alert periods. Meanwhile, the relative contributions of
- 39 coal combustion to PM_{2.5} concentrations in Beijing during the pollution episodes in March 2013,
- 40 November 2016, November 2017 and March 2018 was 40%, 34%, 28% and 11% respectively,
- indicating that the recent "Coal to Gas" project and the contingent "2+26" strategy led to a 41
- 42 dramatic decrease in coal combustion in the Beijing-Tianjin-Hebei Region. On the other hand, the
- relative contribution of vehicle exhaust during the "2+26" orange alert periods in November 2017 43
- and March 2018 reached 40% and 54% respectively. The relative contribution of local emission to 44
- 45 PM_{2.5} concentrations in Beijing also varied significantly and ranged from 49.46% to 89.35%
- 46 during the four pollution episodes. These results suggested that the "2+26" regional
- 47 emission-reduction strategy should be implemented with red air pollution alerts during heavy
- 48 pollution episodes to intendedly reduce the dominant contribution of vehicle exhausts to PM2.5
- 49 concentrations in Beijing, while specific emission-reduction measures should be implemented
- 50 accordingly for different cities within the "2+26" framework.
- Keywords: Air pollution alert; Regional integration; Emission reduction; 51
- 52 WRF-CAMx; Beijing; "2+26".

1 Introduction 53

- 54 In January 2013, a severe haze episode with the highest concentration of hourly fine particulate
- 55 matter with a diameter of less than 2.5 micrometers (PM_{2.5}) occurred in Beijing (886 μg/ m³),
- which attracted worldwide attention. Since 2013, Beijing, located in the Beijing-Tianjin-Hebei 56
- 57 region, has been a heavily polluted area in China that suffers from continuous haze episodes
- 58 associated with high concentrations of PM2.5, especially in winter. Given the significant negative
- 59 influence of PM_{2.5} on public health (Garrett and Casimiro, 2011; Guaita et al., 2011; Pasca et al., 60 2014; Li et al., 2015), the air quality management authority in Beijing has put growing emphasis
- 61 on long-term environmental protection policies, including shutting down polluting factories and
- 62 limiting vehicle use through license plate rules. However, total emissions of airborne pollutants
- 63 remain at very high levels in Beijing, leading to frequent heavy pollution episodes (Guo et al.,
- 64 2012). To mitigate this problem, contingent emission-reduction measures, in addition to regular
- 65 environmental policies, are necessary in Beijing in order to improve local air quality during air
- 66 pollution episodes.

- In 2013, the Beijing Municipal Government published the "Heavy Air Pollution Contingency 67
- 68 Plan" and revised this plan in 2015 to better manage air quality during pollution episodes.
- 69 According to the predicted concentrations of different airborne pollutants and the duration of
- 70 pollution episodes, there are four levels of air pollution alerts for Beijing, which are blue, yellow,
- 71 orange, and red alerts. Specific emission-reduction measures are implemented when each type of
- air pollution alerts is in effect. The red alert is the most stringent level of air pollution alerts and 73 predicts severe air pollution episodes (Air Quality Index [AQI] >300) that will last for more than
- 74 three days. Emission-reduction measures during red alerts mainly include the implementation of
- 75 the odd-even license plate policy (only about half of all of the cars in Beijing is allowed to run

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within the fifth-ring district in each day), the suspension of all outdoor construction work and temporary shutdown of listed polluting factories. The orange alert predicts heavy air pollution episodes (AQI >200) that will last for more than three days. Emission-reduction measures during orange alerts mainly include forbidding vehicles that cannot meet the Environmental Standard Levels I and II, the suspension of specific outdoor work (e.g., painting) and temporary shutdown of listed polluting factories (the list for red alerts includes more factories than that for orange alerts). The blue and yellow alerts predict heavy air pollution episodes that will last for more than one and two days respectively. There are very few compulsory emission-reduction measures for blue and yellow alerts and most emission-reduction measures are suggestive. The characteristics and effects of these emission-reduction measures during alert periods have been massively studied (Zhong, J. et al., 2017; Zhang, Z. et al., 2017; Wang, X. et al., 2017; Zeng, W. et al., 2018; Shang, X. et al., 2018). However, previous emission-reduction measures during orange and red alerts were solely conducted in a specific city (e.g., Beijing) while regional emission-reduction measures implemented simultaneously in many adjacent cities have rarely been implemented and evaluated.

Although the peak PM_{2.5} concentrations in Beijing could be reduced by 20% through strict emission-reduction measures (Cheng et al., 2017), local PM_{2.5} concentrations remained at very high levels during red alert periods. This is mainly attributed to the regional transport of airborne pollutants from neighboring cities to Beijing (Chen et al., 2016). Therefore, regional integration has become one of the major solutions for further reducing PM_{2.5} concentrations in Beijing during heavy pollution episodes. To promote this strategy, the Ministry of Environmental Protection of the People's Republic of China released the "2017 Air Pollution Prevention and Management Plan for the Beijing-Tianjin-Hebei Region and its Surrounding Areas" (MEP, 2017). This plan suggests that Beijing, Tianjin, eight cities in Hebei Province, four cities in Shanxi Province, seven cities in Shandong Province and seven cities in Henan Province (2+26) constitute the regional network involved in the long-distance transport of airborne pollutants surrounding Beijing. Therefore, during heavy pollution episodes, unified emission-reduction measures should be carried out in these cities simultaneously to reduce extremely high PM_{2.5} concentrations in Beijing.

Since the launch of the "2+26" plan, Beijing experienced two pollution episodes in November 2017 and March 2018, when MEP released two orange alerts and implemented corresponding emission-reduction measures in all 28 cities simultaneously. The two orange alerts were the first two attempts of the "2+26" plan to reduce PM_{2.5} concentrations in Beijing. To better evaluate this "2+26" regional strategy and for a comprehensive comparison, we also included in this study two other pollution episodes in Beijing: November 2016 (with local emission-reduction measures) and March 2013 (with no emission-reduction measure). We first analyzed the variations in PM_{2.5} concentrations in Beijing during the four pollution episodes. Following this, we quantified the relative contributions of local emissions and regional transport to PM_{2.5} concentrations in Beijing during these four pollution episodes. The methodology and findings of this research not only holds practical significance for further improving the "2+26" regional strategy, but also shed some light on the regional integration of air quality management in other parts of China.

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2 Materials and methods

2.1 Study sites

Beijing is located at the northwestern edge of the North China Plain. It is surrounded by mountains on three sides, resulting in a geographical condition unfavorable for the dispersion of airborne pollutants. Therefore, air pollution episodes have been frequently witnessed in Beijing since 2013, especially in winter. Based on large-scale field-experiments and model simulation, MEP (2017) pointed out that 28 cities formed a regional transport network of airborne pollutants, which influenced local PM_{2.5} concentrations in Beijing significantly. These 28 cities include two municipalities directly under the central government, Beijing and Tianjin and another 26 neighboring cities surrounding Beijing, which are Shijiazhuang, Tangshan, Lang fang, Baoding, Cangzhou, Hengshui, Xingtai and Handan in Hebei Province, Taiyuan, Yangquan, Changzhi and Jincheng in Shanxi Province, Jinan, Zibo, Jining, Dezhou, Liaocheng, Binzhou and Heze in Shandong Province, Zhengzhou, Kaifeng, Anyang, Hebi, Xinxiang, Jiaozuo and Puyang in Henan Province. The locations of these cities are shown in Fig 1. These 26 cities, especially those cities located in the Hebei provinces, are mainly industrial cities that consume a large amount of coals and produce massive amounts of airborne pollutants. To comprehensively understand the effects of the "2+26" regional strategy for air quality improvement in Beijing, all these 28 cities were selected as study sites for this research.

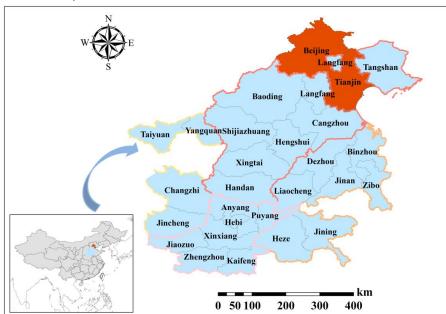


Fig 1. Geographical locations of the 28 cities within the "2+26" regional integration framework

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137 2.2 Data Sources

2.2.1 Ground PM_{2.5} and meteorological observation data

- The data of major airborne pollutants for this research were collected from the website PM25.in.
- 140 This website assembles official data of major airborne pollutants provided by the China National
- 141 Environmental Monitoring Center (CNEMC) and publishes hourly air quality information for 367
- monitored cities in China, which include all of the 28 cities in the "2+26" framework. By using a
- 143 specific API (Application Programming Interface) provided by PM25.in, we collected hourly
- pollutant data (e.g., PM_{2.5}, CO, NO₂, O₃) for these 28 cities. The hourly average concentration of
- 145 each pollutant for one city is calculated by averaging the hourly value measured at all available
- observation stations within the city. For the following analysis, we employed time series air
- quality data covering all four pollution periods: from 0 AM, November 24th to 12 PM, November
- 148 27th, 2016; from 0 AM, November 4th, 2017 to 12 PM, November 7th, 2017; from 0 AM, March
- 149 14th to 12 PM, March 17th, 2013; from 0 AM March 11th to 12 PM, March 14th, 2018.
- 150 In addition to large-scale meteorological data for the following simulation, we also employed
- 151 ground observation data to compare meteorological conditions during these four pollution
- 152 episodes. Meteorological data for this research were collected at the Guanxiangtai Station in
- 153 Beijing and were downloaded from the Department of Atmospheric Science of the University of
- 154 Wyoming (http://weather.uwyo.edu/upperair/sounding.html). Based on the comparison of the
- 155 meteorological data, we could ascertain whether large variations in meteorological conditions
- 156 existed between the four pollution episodes, as a potential influencing factor of the variations in
- 157 PM_{2.5} concentrations.

158 2.2.2 PM_{2.5} component Data

- To comprehensively understand the component of PM_{2.5} during the four pollution episodes, we
- collected PM_{2.5} sample data at the DongSi Station for further analysis. These PM_{2.5} sample data
- were collected during the pollution episodes in March, 2013, November 2016, November 2017
- and March 2018 respectively. We employed an URG-9000B Ambient Ion monitor (Thermo Fisher
- 163 Scientific), which includes two Dionex ICS-90 ion chromatography systems (DIONEX, US), to
- detect water soluble ion Na⁺, Mg²⁺, Ca²⁺, K⁺, NH₄⁺, Cl⁻, SO₄²⁻, NO³⁻, PO₄³⁻. The original temporal
- resolution for ion detection was 15 minutes and for the comparison with other components, the
- 166 resolution for water-soluble ion detection was averaged to an hour. The organic carbon
- concentration of $PM_{2.5}$ was analyzed using the OC/EC organic carbon analyzer (sunset lab model
- 168 5l) and the temporal resolution for carbon detection was an hour. The in-depth analysis of PM_{2.5}
- 169 component provides significant reference for understanding the evolution and sources of PM_{2.5}
- during the pollution episodes.

171 **2.3 Method**

172 2.3.1 Simulation Models

- 173 We employed the WRF-CAMx model for simulating the effects of emission reduction measures
- on the reduction of major airborne pollutants. The WRF-CAMx includes three models: the

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175 middle-scale meteorology model (WRF), the source emission model (SMOKE) 176 (https://www.cmascenter.org/cmaq/) and air quality model (CAMx) (http://www.camx.com/). The WRF model provided the meteorological field for the analysis. The CAMx model has been widely 177 used for simulating the evolution of air pollution episodes (An et al., 2007; Liu et al., 2010) and 178 179 was employed for simulating the variations of airborne pollutants in this study (ENVIRON, 2013). In this research, the central point for the CAMx was set at the coordinate (35 N, 110 E) and 180 181 bi-directional nested technology was employed, producing two layers of grids with a horizontal 182 resolution of 36 km and 12 km respectively. The first layer of the grids has a 36km resolution and 183 200*160 cells covering most areas in East Asia (including Japan, South Korea, China, North 184 Korea, and other countries). The second layer of the grids has a 12km resolution and 120*102 cells, based on the lambert map projection and standard latitude lines 24 N and 46 N, covering the 185 186 North China Plain, which includes the Beijing-Tianjin-Hebei region, Shandong and Henan 187 Provinces. The vertical layer was divided into 20 unequal layers, eight of which were of less-than-1km distance to the ground for better featuring of the structure of atmospheric boundary. 188 Airborne pollutants in CAMx were simulated according to some physical and chemical 189 190 mechanisms, including: a) horizontal advection scheme (PPM), b) implicit Euler vertical 191 convection scheme, c) the horizontal diffusion of K theory, d) Saprc99 gas-phase chemical 192 mechanism, and e) EBI calculation method. The initial and boundary conditions for simulating 193 airborne pollutants were set using the default CAMx profiles. For better simulating the pollution 194 process with longer time series, the simulation period was set as the entire March 2013, November 195 2016, November 2017, and March 2018. For the first running of this model, a spin-up period of 5 196 days was set to simulate the initial field and the following initial field was decided by the output of 197 previous simulations. Hence, the accumulation effects of emission sources have been 198 comprehensively considered and the influence of uncertain initial conditions has been reduced 199 significantly.

We employed ARW-WRF3.2 to simulate the meteorological field. The setting of the center and the bi-directional nest for the WRF was similar to that of the CAMx as mentioned above. There were 35 vertical layers for the WRF and the outer layer provided boundary conditions of the inner layer. The meteorological background field and boundary information with a GFS resolution of 1 °×1 ° and temporal resolution of 6h were acquired from NCAR (National Center for Atmospheric Research, https://ncar.ucar.edu/) and NCEP (National Centers for Environmental Prediction) respectively. The terrain and underlying surface information was obtained from the USGS 30s global DEM (https://earthquake.usgs.gov/). The output from the WRF model was interpolated to the region and grid for the CAMx model using the Meteorology-Chemistry Interface Processor (MCIP, https://www.cmascenter.org/mcip). The meteorological factors used for this model include temperature, air pressure, humidity, geopotential height, zonal wind, meridional wind, precipitation, boundary layer heights and so forth. An estimation model for terrestrial ecosystem MEGAN (http://ab.inf.uni-tuebingen.de/software/megan/) was employed to process the natural emissions. Anthropogenic emission data were from the Multi-resolution Emission Inventory for China, MEIC 0.5 °×0.5 ° emission inventory (http://www.meicmodel.org/) and Beijing emission inventory (http://www.cee.cn/). We input the processed natural and anthropogenic emission data into the SMOKE model and acquired comprehensive emission source files.

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218 Table 1 Sources of Emission inventory

Airborne Pollutants	Sources	Data description
PM _{2.5} , BC, OC	MEIC	Resolution: 0.5 °×0.5 °
SO_2	Survey of Emission sources	Point sources, Polygon sources
NOx	Survey of Emission sources	Point sources, Polygon sources
PM_{10}	Survey of Emission sources	Point sources, Polygon sources
NH_3	MEIC	Resolution: 1 °×1 °
Anthropogenic VOCs	MEIC	Resolution: 0.5 °×0.5 °
Natural VOCs	MEGAN	Corresponding Grid data

2.3.2 Source Apportionment

PSAT (Particulate Matter Source Apportionment Technology) is one major extension of the CAMx model. PSAT was developed from the related ozone source apportionment method and provided PM source apportionment for specific geographic regions and source categories (Huang, Q. et al.,2012). Furthermore, PSAT can be used to analyze the source-acceptor relationship of PM_{2.5} pollutants, and trace SO₂, SO₄²⁻, NO₃-, NH₄+, SOA, Hg, EC, dust particles, and other primary and secondary particles. As a species tagging method, PSAT tracks the regional source and industry source of environmental receptor PM2.5 and its main chemical components, and then evaluates the contribution of initial conditions and boundary conditions to PM generation. By identifying and tracking the transport, diffusion, transformation and decomposion of pollutants emitted from various sources, PSAT estimates the relative contribution of different emission sources to the spatial distribution of PM concentrations based on the analysis of mass balance. PSAT-based source apportionment is conducted using reactive tracers that simulate the nonlinear transformation between primary PM and secondary PM and are highly efficient and flexible for source apportionment from the perspective of geographical source regions, emissions source categories and individual sources (Burr, M. et al., 2011). PSAT effectively avoids the concentration biases caused by Brute-force based source-closure methods that ignores non-linear chemical processes and has been widely in previous studies (Xing, J. et al., 2011; Huang, Q. et al., 2012; Wu, D. et al., 2013; Li, X. et al., 2015; Li, Y. et al., 2015). For this research, we established a regional transport matrix between pollution sources and environmental receptors. According to the provincial administrative division, the national grid is divided into 17 divisions, each of which represents a provincial unit, and all other cells outside the national boundary are classified as Class I, including the ocean and other areas. According to the scope of the Beijing-Tianjin-Hebei Region and the "2+26" network, we further divide the study area into 13 sub-divisions, including Beijing, Tianjin, eight cities in Hebei Province, Henan Province, Shandong Province and Shanxi Province, for quantifying the influence of local emission and regional transport on the variations in PM2.5 concentrations in Beijing during the four pollution episodes.

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2.4 Model verification

For the mean PM_{2.5} concentrations in Beijing, we compared the observed and model estimated PM_{2.5} concentrations during the four pollution episodes to verify the accuracy of the WRF-CAMx model (Fig 2). According to Fig 2, a general agreement was found between the simulated and observed data with more than 85% of data points falling into the siege area of 1:2 and 2:1 lines. WRF-CAMx slightly underestimated PM_{2.5} concentrations due to the uncertainty in the emission inventory, meteorological field simulation errors and insufficient chemical reaction mechanisms. For the four pollution episodes, the correlation coefficient R, normalized mean bias (NMB), normalized mean error (NME), mean fractional bias (MFB) and mean fractional error (MFE) between observed and simulated data ranged from 0.65~0.80, -21%~-10%, 19%~32%, -31%~-8%, and 12%~34% respectively, indicating a satisfactory simulation output (Boylan et al., 2006).

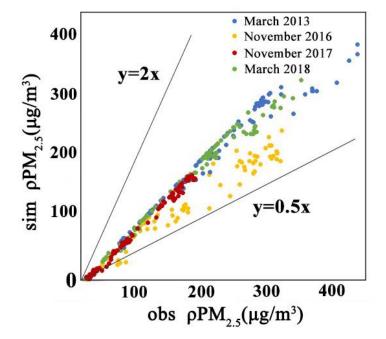


Fig 2. The comparison of observed and model simulated $PM_{2.5}$ concentrations in Beijing during four pollution episodes.

3 Results

3.1 Temporal variations in PM_{2.5} concentrations during the four pollution episodes

Chen et al. (2017, 2018) suggested that wind speed and relative humidity were the major meteorological factors that influenced wintertime $PM_{2.5}$ concentrations in Beijing. Based on the

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ground observation data, we found that the two meteorological factors during the pollution episode in November 2016 was fairly similar to that during the orange alert period in November 2017, while the meteorological condition during the pollution episode in March 2013 was fairly similar to that during the orange alert period in March 2018 (as shown in Table 2). According to Table 2, all the four pollution episodes experienced a high-humidity and weak-wind condition. Specifically, the fairly high relative humidity for the "2+26" orange alert period in November 2017 and the fairly low wind speed for the "2+26" orange alert period in March 2018 led to extremely unfavorable conditions for the dispersion of airborne pollutants.

Table 2 Major meteorological conditions during the four pollution episodes.

Pollution Episodes	Mean Relative	Mean Wind	
	Humidity(%)	Speed(m/s)	
March, 2013 (No emission-reduction)	56.25	2.32	
March, 2018 ("2+26" strategy)	57.25	1.72	
November, 2016 (Local emission-reduction)	58.25	2.37	
November, 2017 ("2+26" strategy)	72.25	2.09	

When the meteorological influences on the variations of $PM_{2.5}$ concentrations were limited, a comparison between the $PM_{2.5}$ concentrations during these two orange alert periods and that during the two corresponding pollution episodes provides useful reference for evaluating the effects of "2+26" strategy on $PM_{2.5}$ reductions during the pollution episodes, which are usually observed under a stagnant atmospheric condition, with high relative humidity and low wind speed. The temporal variations in $PM_{2.5}$ concentrations during the two "2+26" orange alerts and the two corresponding pollution episodes are shown in Table 3 and Fig 3.

Table 3 Characteristics of PM_{2.5} concentrations during four pollution episodes

Pollution	Mean	Peak	Duration of	Duration of	Duration of	Period with	
episode	$(\mu g/m^3)$	$(\mu g/m^3)$	PM _{2.5} >100	$PM_{2.5} > 150$	PM _{2.5} >200	PM _{2.5} >300	
			$\mu g/m^3$ (h)	$\mu g/m^3$ (h)	$\mu g/m^3$ (h)	$\mu g/m^3 (h)$	
March, 2013	208.49	426.12	10	24	37	12	
(No emission-reduction)							
March, 2018	174.24	325.91	6	10	44	5	
("2+26" strategy)							
November, 2016	138.05	138.05 310.30	310.30	14	9	26	5
(Local emission-reduction)		510.50	14	9	20	3	
November, 2017	92.91	176.20	24	24	0	0	
("2+26" strategy)							

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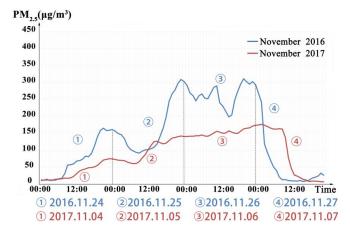


Fig 3. Variations of $PM_{2.5}$ concentrations during four pollution episodes with different emission-reduction measures in Beijing

As shown in Table 3 and Figure 3, both the peak and average $PM_{2.5}$ concentrations during the two orange alert periods were remarkably lower than those during the two corresponding pollution episodes with similar initial $PM_{2.5}$ levels and meteorological conditions. For the pollution episode in March 2013 and March 2018, $PM_{2.5}$ concentrations were both around $50\mu g/m^3$ at the beginning of both periods. Similarly, for the pollution episode in November 2016 and November 2017, the initial $PM_{2.5}$ concentrations were both around 15 $\mu g/m^3$ at the beginning of both periods. Following the similar initial $PM_{2.5}$ concentrations, it is noted that $PM_{2.5}$ concentrations increased at a much lower rate and further led to a lower peak and average $PM_{2.5}$ concentrations during the two orange alert periods.

According to Table 3, the mean and peak PM_{2.5} concentrations during the "2+26" orange alert period in March, 2018 was 16.43% and 23.52% lower than those during the pollution episode in March 2018 respectively. Meanwhile, the duration with extremely high PM_{2.5} concentrations was notably shorter during the orange alert period. The "2+26" strategy implemented during the

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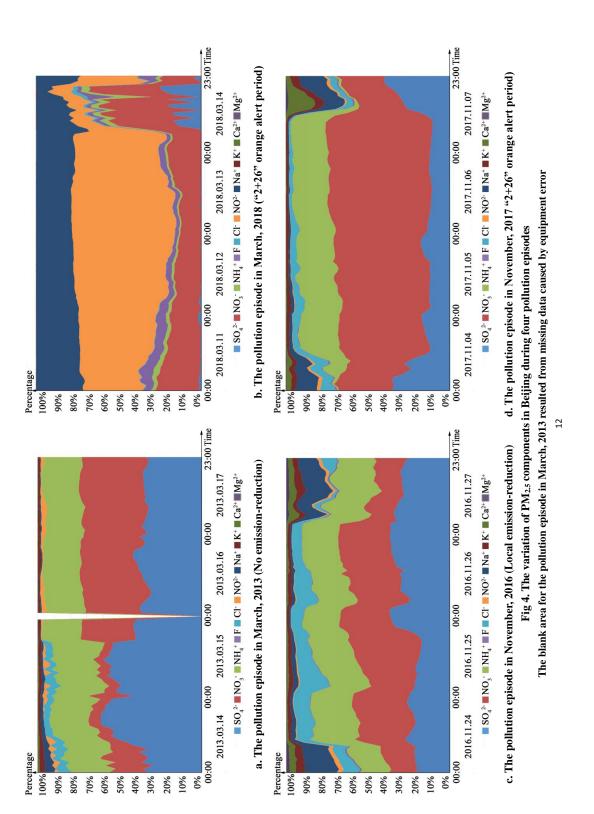


298	orange alert period in November 2017 led to even better effects on PM _{2.5} reductions. The mean
299	and peak PM _{2.5} concentrations during this period was 32.70% and 43.22% lower than those during
300	the pollution episode in November 2016 respectively. More importantly, during the entire orange
301	alert period, PM _{2.5} concentrations were constantly lower than 200 µg/m³, indicating a highly
302	efficient control of high PM _{2.5} concentrations.
303	3.2 PM _{2.5} component analysis during four pollution episodes
304	The temporal variations of different PM _{2.5} components during the four pollution episodes are
305	shown in Fig 4. As the figure indicates, the components of PM _{2.5} in Beijing during the four
306	pollution episodes had notable variations.

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For the four pollution episodes with different emission-reduction measures, the main components for $PM_{2.5}$ were all SO_4^{2-} , NO_3^- , and NH_4^+ . However, some major differences existed. With no or only local emission-reduction measures implemented, the dominant $PM_{2.5}$ components was SO_4^{2-} for the pollution episode in March 2013 and November 2016. During two "2+26" orange alert periods, NO_3^- became the dominant $PM_{2.5}$ components. Except for the pollution episode in March 2018, the proportion of another major ion NH_4^+ was generally consistent during the four pollution episodes. The mean mass concentrations and proportions of SO_4^{2-} , NO_3^- , and NH_4^+ during the four pollution episodes are shown in Table 4.

Table 4. The mean mass concentration and percent of three major $PM_{2.5}$ components during four pollution episodes

SO₄²⁻ NO₃ NH_4^+ Pollution Concentration Concentration Concentration **Episode** Percent Percent Percentage $(\mu g/m^3)$ (μg/m³) $(\mu g/m^3)$ March, 2013 (No emission-reduction) 45.11 39.92% 34.63 30.65% 27.14 24.02% March, 2018 ("2+26" strategy) 20.24 14.66% 50.17 36.34% 14.00 10.14% November, 2016 (Local emission-reduction) 14.96 30.10% 16.19 32.57% 11.66 23.45% November,2017 ("2+26" strategy) 7.37 13.03% 33.42 59.06% 12.33 21.80%

Through comparison, we found a dramatic decrease of SO_4^{2-} and a notable increase of NO_3^{-} during two orange alert periods. The main source for SO_4^{2-} is the combustion of fossil fuels (Shimano, S. et al.,2006; Kuenen, J. et al.,2013), especially the intensive burning of sulfur coals for wintertime central-heating, manufacturing and household use. The main source for NO_3^{-} is vehicle exhaust (Rodr guez, S. et al.,2004; Watson, J. G. et al.,2007; Zeng, F. et al.,2010). NH_4^{+} is the secondary pollutant of urban NH_3 , the main source of which is the decomposition of organic elements (Frank, D. S. et al.,1980; Watson, J. G. et al.,2007) and the combustion of fossil fuels (Frank, D. S. et al.,1980; Watson, J. G. et al.,2007; Pan et al., 2016). Through a novel approach, Pan et al (2016) quantified that more than 90% NH_3 in the Beijing-Tianjin-Hebei Region during heavy pollution episodes resulted from the combustion of fossil fuels. The large variations of $PM_{2.5}$ components during these episodes was mainly attributed to long-term environmental

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policies and contingent emission-reduction measures. A large number of small polluting factories in Beijing and its surrounding areas have been shut down, and the use of household coal, especially coarse coal that produces large amounts of sulfate-related pollutants, has been restricted significantly. In addition to long-term environmental protection policies, contingent emission-reduction measures, including the temporal shut-down of many factories that consumes a large amount of coal, were implemented during air pollution alert periods. Furthermore, the recently launched "2+26" plan requires that areas surrounding Beijing, including many cities in Hebei Province (e.g., Tangshan) well-known for their coal-based iron industries, should take simultaneous emission-reduction actions during regional pollution episodes. These long-term and contingent strategies led to a notable decrease of SO₄²⁻ through local emission-reduction measures and a further decrease of SO₄²⁻ through "2+26" regional emission-reduction measures. Meanwhile, the much lower proportion of NH₄⁺ in March 2018 also indicated a sudden reduction in coal fuel usage in Beijing and its neighboring areas in 2018, the major driver for which will be explained in the following section. Conversely, during the four pollution episodes, no strict regulation was placed on the control of vehicle exhaust. Hence, the notable decrease of SO₄²⁻ and generally constant mass concentration of NO₃ led to a rapidly rising proportion of NO₃ among the PM_{2.5} components during the two orange alerts.

3.3 Source apportionment during the four pollution episodes

Based on PM_{2.5} component analysis and PSAT-based source apportionment, we further quantified the relative contributions of different sources to PM_{2.5} concentrations in Beijing during the four pollution episodes (Fig 5). A major difference between the pollution episode in March 2013 and in March 2018 was the dramatic decrease in the relative contribution of coal combustion from 40% to 11%. Meanwhile, the relative contribution of vehicle exhaust increased significantly from 19% to 54%, indicating that vehicle exhaust became the dominant source for the pollution episode in March 2018 with the "2+26" regional emission-reduction measures. On the other hand, the difference in the relative contributions of different sources between the two pollution episodes in November 2016 (with local emission-reduction measures) and November 2017 (with "2+26" regional emission-reduction measures) were much smaller. The major differences lied in the notable increase in the relative contribution of vehicle exhaust from 29% to 40% and the decrease in the relative contribution of coal combustion from 34% to 28%.

As described above, the continuous decrease in the relative contribution of coal combustion from the pollution episodes in 2013 to the episode in 2018 resulted from the combination of long-term

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and contingent local and regional emission-reduction measures. Note that despite a similar "2+26" strategy implemented, the relative contribution of coal combustion during the orange alert period in November 2017 was much higher than that in March 2018. A major reason for this dramatic change in a short period was the implementation of a large-scale environmental project. Before November 2017, the starting point of central heating in Beijing, a regional project called "Coal to Gas" had finished replacing coal-based central heating systems by gas-based systems for 1.9 million households in the Beijing-Tianjin-Hebei Region, leading to a 2 million-ton decrease in coal consumption in the region. As a result, the relative contribution of coal combustion, which was the dominant emission source for PM_{2.5} in Beijing during the central-heating season from November to March, decreased to a fairly low level during the orange alert period in March 2018.

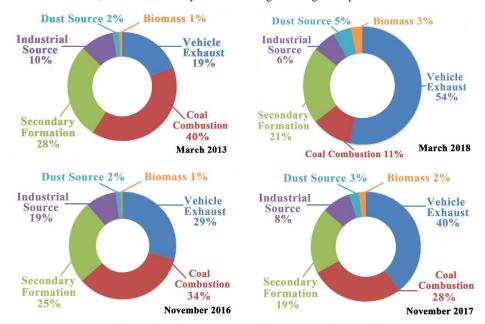


Fig 5. The relative contribution of different sources to $PM_{2.5}$ concentrations in Beijing during four pollution episodes

3.4 The relative contribution of local emission and regional transport to PM_{2.5} concentrations in Beijing during the four pollution episodes

Through the simulation of the WRF-CAMx model based on local and regional emission inventories, we quantified the relative contributions of local emission and regional transport of airborne pollutants to the variations in $PM_{2.5}$ concentrations in Beijing during four pollution episodes (Fig 6). According to Fig 6, the relative contributions of local emission and regional

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transport to PM_{2.5} concentrations in Beijing varied notably. For the pollution episode in March 2013 with no emission-reduction measures, the relative contribution of local emissions was 69.27%, much lower than the 88.35% for the "2+26" orange alert period in March 2018. On the other hand, for the pollution episode in November 2016 with local emission-reduction measures, the relative contribution of local emissions was 76.83%, much higher than the 49.46% for the "2+26" orange alert period in November 2017. Meanwhile, the relative contribution to PM_{2.5} concentrations in Beijing from specific areas also differed significantly during these pollution episodes. We found that different emission-reduction strategies did not lead to a clear pattern for the relative contributions of local emission and regional transport. One major reason for this is that the regional transport of airborne pollutants from neighboring areas to Beijing is influenced significantly by meteorological conditions, the intensity of regional emission sources and the regional distribution of PM_{2.5} concentrations, which demonstrated remarkable seasonal variations and synoptic-scale uncertainties. From this perspective, we attempted to explain the underlying drivers for the variations in local and regional contributions to PM_{2.5} concentrations during the four pollution episodes.

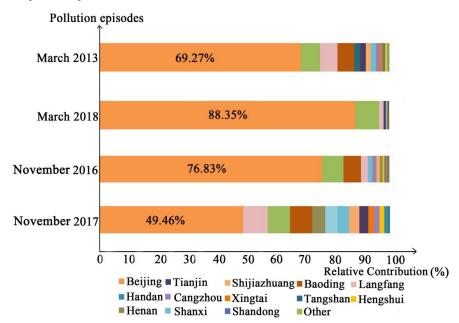


Fig 6. The relative contributions of local emission and regional transport to PM_{2.5} concentrations in Beijing during the four pollution episodes

For the pollution episode in March 2013, without long-term and contingent emission-reduction measures, the large amount of combusted coal fuels in the neighboring areas of Beijing led to a

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relatively large regional contribution. For the pollution episode in March 2018, with the implementation of the large-scale "Coal to Gas" project and "2+26" strategy, the rapidly reduced coal consumption in cities surrounding Beijing and the limited restriction on the emission of local vehicles led to a fairly high local contribution. For the pollution episode in November 2016, an inversed temperature layer was observed with high relative humidity, which was a favorable environment for the production of secondary PM_{2.5} and a relatively large local contribution. Despite the implementation of the "2+26" strategy, the abnormally high regional contribution for the pollution episode in November 2017 could be attributed to the prevailing southerly winds that brought in a large amount of air from neighboring cities (e.g., Shijiazhuang). Therefore, although this pollution episode occurred in winter, it had more similarities to a summertime pollution episode, which was characterized by prevailing southerly winds, thoroughly mixed pollutants within the Beijing-Tianjin-Hebei Region, and notable regional transport.

4 Discussion

411 Through the comparison of the components of PM_{2.5} during the pollution episode in 2013 and those in 2016, 2017 and 2018, we found that the proportion of sulfate ions decreased significantly 412 413 while nitrite ions became the dominant component of PM_{2.5} during the pollution episodes. This 414 result is consistent with findings from some recent studies (Fromme H et al., 2008; Tan J et al., 2016; Shang X et al., 2018). The dramatic decrease in the proportions of SO₄² and NH₄⁺ ions in 415 416 PM_{2.5} during the pollution episode in March 2018 suggested a satisfactory effect of long-term 417 pollution control measures, especially the "Coal to Gas" project, and contingent "2+26" regional emission-reduction measures on the reduction of coal combustion in the Beijing-Tianjin-Hebei 418 419 Region. As revealed by previous studies (Zhang R et al., 2013; Liu Y et al., 2018; Shang X et al., 420 2018) and the source apportionment from this research, the use of coal fuels has been the 421 dominant source for the formation and mass concentration of PM_{2.5} in Beijing since 2013. 422 However, the remarkable decrease in coal combustion since the winter of 2017 has greatly 423 reduced the contribution of coal combustion to local PM2.5 concentrations, which directly 424 improved the wintertime air quality and led to the cleanest winter in Beijing since 2013. The mean 425 wintertime (the winter for Beijing here refers to the central-heating season from November 15th to 426 March 15th) PM_{2.5} concentration in Beijing for 2013, 2014, 2015, 2016 and 2017 was 88.19, 84.41, 89.39, 92.39 and 47.31 μ g/ m^3 respectively. 427

The implementation of the "2+26" strategy led to different effects on PM_{2.5} reductions during

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specific pollution episodes. In addition to different emission-reduction strategies, the improvement of air quality in Beijing is controlled by a diversity of factors. First, meteorological conditions exert a strong influence on the accumulation and dispersion of local airborne pollutants in Beijing and the long-distance transport of airborne pollutants from neighboring areas. Second, the distribution of PM2.5 concentrations in the "2+26" region determines whether the air brought into Beijing from neighboring areas increases or decreases PM_{2.5} concentrations there. Third, the PM_{2.5} level during pollution episodes influences the relative contributions of local and regional contributions. The mean PM_{2.5} concentrations during the "2+26" orange alert period in March 2018 was 174.24 μg/m³. High-concentration PM_{2.5} during pollution episodes led to a stagnant condition with high humidity and low wind speed (Chen et al. 2017, 2018), which was an unfavorable condition for the regional transport of airborne pollutants. Therefore, the relative contribution of local emission to this extremely high PM_{2.5} concentrations was 88.35% while the relative contribution of regional transport was 11.65%. In this case, although unified emission-reduction measures were implemented in its neighboring areas, the significantly restricted regional transport did not fully project the effects of the "2+26" strategy to the local PM_{2.5} concentrations in Beijing. Conversely, the mean PM_{2.5} concentrations during the "2+26" orange alert period in November 2017 was 92.91 µg/m³, which was not high enough to significantly prevent the regional transport of airborne pollutants. Therefore, the "2+26" strategy led to a simultaneous reduction in PM2.5 concentrations in this region and a large amount of clean air from its neighboring cities that significantly diluted the local PM_{2.5} in Beijing. Consequently, the relative contribution of regional transport was larger than 50% and thus the "2+26" strategy achieved a much better effect on PM_{2.5} reductions than that in March 2018. Another dominant factor that influences the effects of the "2+26" strategy is the level of air pollution alert and its corresponding emission-reduction measures. With the launch of orange air pollution alerts, a series of restrictions are placed on the temporary shut down of polluting factories and the emission of fossil fuels can be reduced significantly. However, during orange alert periods, only the use of a small proportion of vehicles that cannot meet Environmental Standards Level I and II are forbidden whilst no additional regulation is implemented on the use of more than 5 million private cars in Beijing. As a result, the relative contribution of vehicle exhaust increased rapidly during two of the "2+26" orange alert periods. Especially for the orange alert period in March 2018, vehicle exhaust contributed to more than 50% of the high PM_{2.5} concentrations that were higher than 174.24 µg/m³. With dramatically reduced use of coal fuels in

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the Beijing-Tianjin-Hebei Region due to the recent completion of the "Coal to Gas" project, the control of vehicle exhaust is increasingly crucial for managing PM2.5 concentrations during pollution episodes. In this light, red air pollution alerts, which have stricter regulations on the use of vehicles, should be employed with the "2+26" regional emission-reduction strategy during heavy pollution episodes. For instance, during the heavy pollution episode in March 2018, if a red alert instead of the orange alert was issued, the implementation of odd-even license plate policy would instantly cut the daily use of private cars in Beijing by fifty percent and significantly reduce the contribution of vehicle exhaust to PM_{2.5} concentrations. Given the growing contribution of vehicle exhaust to PM_{2.5} pollutions in Beijing, in addition to the contingent regulations during pollution episodes, long-term policies, including the improvement of the public transit system, the enhancement of petrol quality and promotion of electric cars, should be properly implemented for further reducing vehicle-exhaust induced PM_{2.5} pollutions. Although the regional transport network for air pollution in Beijing has been identified, this research suggested that only those cities adjacent to Beijing, such as Baoding, Shijiazhuang and Lang fang, made a relatively large contribution to the PM_{2.5} concentrations in Beijing whilst the relative contributions of some other areas within the "2+26" framework were very limited. Considering the substantial social and economic loss induced by the implementation of air pollution alerts, city-specific, rather than region-wide unified emission-reduction strategies, should be conducted for promoting air quality in Beijing during pollution episodes. Tight measures can be implemented in cities that make large contributions while lenient measures can

be implemented in cities that make limited contributions to PM_{2.5} concentrations. To this end, future studies should place more emphasis on quantifying the relative contributions from different

482 future studies should place more emphasis on quantifying the relative contributions from different

cities to local PM_{2.5} concentrations in Beijing and setting city-specific emission-reduction

measures for each city within the "2+26" region.

5 Conclusions

We compared the variations in $PM_{2.5}$ concentrations in Beijing during four recent pollution episodes with different emission-reduction strategies. Based on this comparison, we found that the "2+26" regional emission-reduction strategy implemented in March 2018 led to a mean $PM_{2.5}$ concentrations of only 16.43% lower than that during the pollution episode in March 2013, when no emission-reduction measure was in place. On the other hand, the same "2+26" strategy implemented in November 2017 led to a mean $PM_{2.5}$ concentrations of 32.70% lower than that

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during the pollution episode in November 2016 with local emission-reduction measures. The result suggested that the effects of the "2+26" regional emission-reduction measures on PM2.5 reductions were influenced by meteorological conditions, regional distribution of PM2.5 concentrations and local PM_{2.5} level, and could differ significantly during specific pollution episodes. Based on our PM_{2.5} component analysis, we found that the proportion of sulfate ions decreased significantly and nitrate ions were the dominant PM2.5 components during the two "2+26" orange alert periods. The source apportionment revealed that the relative contribution of coal combustion to PM_{2.5} concentrations during the pollution period in March 2013, November 2016, November 2017 and March 2018 was 40%, 34%, 28% and 11% respectively, indicating that the recent completion of the large-scale "Coal to Gas" project and contingent "2+26" regional emission-reduction measures led to a dramatic decrease in coal combustion in the Beijing-Tianjin-Hebei Region. Meanwhile, with no specific regulation on the use of private cars, the relative contribution of vehicle exhaust during the "2+26" orange alert periods in November 2017 and March 2018, was 40% and 54% respectively. The relative contribution of local emissions to PM_{2.5} concentrations in Beijing varied significantly and ranged from 49.46% to 89.35% during the four pollution episodes. With gradually reduced coal consumption in the Beijing-Tianjin-Hebei region, this research suggested that the "2+26" regional emission-reduction strategy should be implemented with red air pollution alerts to intendedly reduce the dominant contribution of vehicle exhausts to PM_{2.5} concentrations. Meanwhile, emission-reduction policies should be designed and implemented accordingly for different cities within the "2+26" regional framework. The methodology and findings from this research provided useful reference for comprehensively understanding the effects of the "2+26" strategy, and for better implementation of future long-term and contingent emission-reduction measures during heavy pollution episodes.

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524 Author contribution

- 525 Chen, Z., Xu, B and Yang, L. designed this research. Chen, Z wrote this manuscript. Chen, D.,
- Zhuang, Y, Cheng, N, Gao, B. Li, R. and Zhao, B conducted data analysis. Chen, D and Zhuang, Y.
- 527 produced the figures. Kwan, M., Yang, L. and Chen, B helped revise this manuscript.

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