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Cross-boundary transport and source apportionment for $PM_{2.5}$ in a typical industrial city in the Hebei Province, China: A modeling study

Xiaoyong Liu[1,2,4], *Xiaole Pan*[1,*], *Jie Li*[1], *Xi Chen*[1,4], *Hang Liu*[1,4], *Yu Tian*[1,4], *Yuting Zhang*[1,4], *Shandong Lei*[1,4], *Weijie Yao*[1,4], *Qi Liao*[1,4], *Yele Sun*[1,2,5], *Zifa Wang*[1,2,5], *Hong He*[2,3]

[1] *State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China*
[2] *Center for Excellence in Regional Atmospheric Environment, Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, China*
[3] *Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China*
[4] *University of Chinese Academy of Sciences, Beijing 100049, China*
[5] *College of Earth Sciences, University of Chinese Academy of Sciences, Beijing 100049, China*

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ABSTRACT

Cross-boundary transport of air pollution is a difficult issue in pollution control for the North China Plain. In this study, an industrial district (Shahe City) with a large glass manufacturing sector was investigated to clarify the relative contribution of fine particulate matter ($PM_{2.5}$) to the city's high levels of pollution. The Nest Air Quality Prediction Model System (NAQPMS), paired with Weather Research and Forecasting (WRF), was adopted and applied with a spatial resolution of 5 km. During the study period, the mean mass concentrations of $PM_{2.5}$, SO_2, and NO_2 were observed to be 132.0, 76.1, and 55.5 $\mu g/m^3$, respectively. The model reproduced the variations in pollutant concentrations in Shahe at an acceptable level. The simulation of online source-tagging revealed that pollutants emitted within a 50-km radius of downtown Shahe contributed 63.4% of the city's total $PM_{2.5}$ concentration. This contribution increased to 73.9±21.2% when unfavorable meteorological conditions (high relative humidity, weak wind, and low planetary boundary layer height) were present; such conditions are more frequently associated with severe pollution ($PM_{2.5} \geq 250$ $\mu g/m^3$). The contribution from Shahe was 52.3±21.6%. The source apportionment results showed that industry (47%), transportation (10%), power (17%), and residential (26%) sectors were the most important sources of $PM_{2.5}$ in Shahe. The glass factories (where chimney stack heights were normally < 70 m) in Shahe contributed 32.1% of the total $PM_{2.5}$ concentration in Shahe. With an increase in $PM_{2.5}$ concentration, the emissions from glass factories accumulated vertically and narrowed horizontally. At times when pollution levels were severe, the horizontally influenced area mainly covered Shahe. Furthermore, sensitivity tests indicated that reducing

* Corresponding author.
E-mail: panxiaole@mail.iap.ac.cn (X. Pan).

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emissions by 20%, 40%, and 60% could lead to a decrease in the mass concentration of $PM_{2.5}$ of of 12.0%, 23.8%, and 35.5%, respectively.

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Introduction

In past decades, China has experienced rapid economic development and urbanization, which has resulted in the release of excessive emissions into the atmosphere. In the North China Plain (NCP), haze caused by fine particulate matter ($PM_{2.5}$) occurs frequently (Zhao et al., 2013; Ji et al., 2016; Sun et al., 2020a, 2020b; Yang et al., 2020). High concentrations of $PM_{2.5}$ have an adverse effect on air quality and human health. Furthermore, $PM_{2.5}$ can also affect agricultural ecosystems and the climate (Feng et al., 2018; Huang et al., 2014). To determine the features and mechanisms of haze formation in the NCP, many studies have been conducted that use a combination of field campaigns and numerical models (Cheng et al., 2019; Li et al., 2017a, 2017b; Li et al., 2019; Yang et al., 2015). In the NCP, issues related to cross-boundary transport and source apportionment of $PM_{2.5}$ have been hot topics. Air quality modeling systems, such as weather research and forecast modeling with online chemistry (WRF-Chem), community multiscale air quality (CMAQ), Air Quality Model with Extensions (CAMx), and the nested air-quality prediction modeling system (NAQPMS), are commonly used to examine air quality issues (Gao et al., 2016, 2020; Wang et al., 2018). Previous studies indicate that regional transport of $PM_{2.5}$ plays an important role in regional haze episodes in Hebei, Beijing, and Tianjin (Wang et al., 2014). Wang et al. (2015) employed the CMAQ model to study the sources of $PM_{2.5}$ in Hebei Province. They found that the regional source contributions to the total concentrations of $SO_4{}^{2-}$, $NO_3{}^{-}$, and $NH_4{}^{+}$ were 40.9%, 62.0%, and 59.1% in Shijiazhuang, Xingtai, and Handan, respectively. Liu et al. (2020) determined that local sources were the dominant contributors to $PM_{2.5}$ in Beijing, and that the main external contributions originated from surrounding areas, such as Hebei and Shandong. To the best of our knowledge, applications of models to study $PM_{2.5}$ in the NCP have mostly been restricted to studies of a megacity or at the regional scale. Research that focuses on typical industrial cities in Hebei Province has been rare; and these well known industrial cities have been largely ignored.

In 2017, a regional integrated air pollution prevention and management project was implemented in the Jing-Jin-Ji Region and surrounding areas (Chen et al., 2019, 2020). The project includes Beijing, Tianjin, Shijiazhuang (the capital city of Hebei Province), and other cities, such as Shahe, an industrial city characterized by glass manufacturing. The annual production of glass in Shahe accounted for approximately 60% of the gross product in the NCP. Shahe suffers from serious air pollution due to the substantial emission of pollutants. Therefore, it is urgent and necessary to conduct relevant research in Shahe. The results of such research can broaden our knowledge of haze issues in the NCP and can provide useful knowledge to inform environmental policy. In this study, a three-domain nested simulation using the NAQPMS model was applied to analyze the cross-boundary transport and source apportionment of $PM_{2.5}$ in Shahe in the winter of 2017–2018. Additionally, the contribution of pollutant emissions from glass-making factories to total $PM_{2.5}$ concentrations in Shahe was quantified. Finally, sensitivity tests were conducted to evaluate the effect of reduced levels of emissions.

1. Methods and model configurations

1.1. *Description of the study area and observation site*

Shahe, located in southern Hebei Province, has a population of 450,000, and covers an area of approximately 1000 km^2. A field campaign was conducted in downtown Shahe from January 1 to 20, 2018. Hourly mean mass concentrations of $PM_{2.5}$, SO_2, and NO_2 were obtained. The sampling site (114.52° N, 36.88° E) was located in the southeastern part of the city (Fig. 1**b**), on the eighth floor of a building situated approximately 28 m above the ground. There are many glass factories to the northeast of the sampling site (Fig. 1**b**).

1.2. *Instruments*

The hourly mean mass concentrations of NO_2 and SO_2 were determined with two integrated gas monitors (NA-721 and SA-731, Kimoto Electric, Ltd., Japan), which use bioluminescence and ultraviolet fluorescence methods, respectively. The mass concentration of $PM_{2.5}$ was determined by a dichotomous monitor (PM-712, Kimoto Electric, Ltd., Japan) at a time-resolution of 1 hr and a flow rate of 16.7 L/min. More detailed descriptions of the above instruments can be found in published literature (Li et al., 2017a, 2017b; Yang et al., 2018). The hourly relative humidity (RH), atmospheric temperature (T), wind speed (WS), and wind direction (WD) were determined by an automatic meteorological station.

1.3. *Model description and configurations*

The Nested Air Quality Prediction Model System (NAQPMS) is a model developed by the Institute of Atmospheric Physics, Chinese Academy of Sciences and is a three-dimensional Eulerian terrain-following chemical transport model; this model has been widely used to study air pollution in China (Chen et al., 2018; Du et al., 2019; Kong et al., 2019). The Carbon Bond Mechanism version Z (CBMZ) and thermodynamic model ISORROPIA1.7 are used in the NAQPMS model. An online tracer-tagging module is incorporated into NAQPMS, which can be used to identify the source apportionment of pollutants and resolve the contributions from the source-tagging region (Li et al., 2014; Wu et al., 2017). The three-dimensional Weather Research and Forecasting (WRF) model version 3.6.1

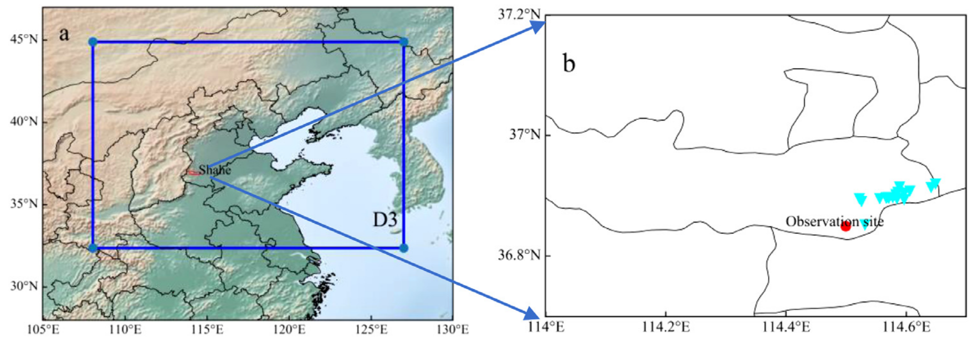

Fig. 1 – (a) The third model domain of WRF and NAQPMS; (b) The location of the observation site (red dot) and glass factories (light blue inverted triangles) in Shahe.

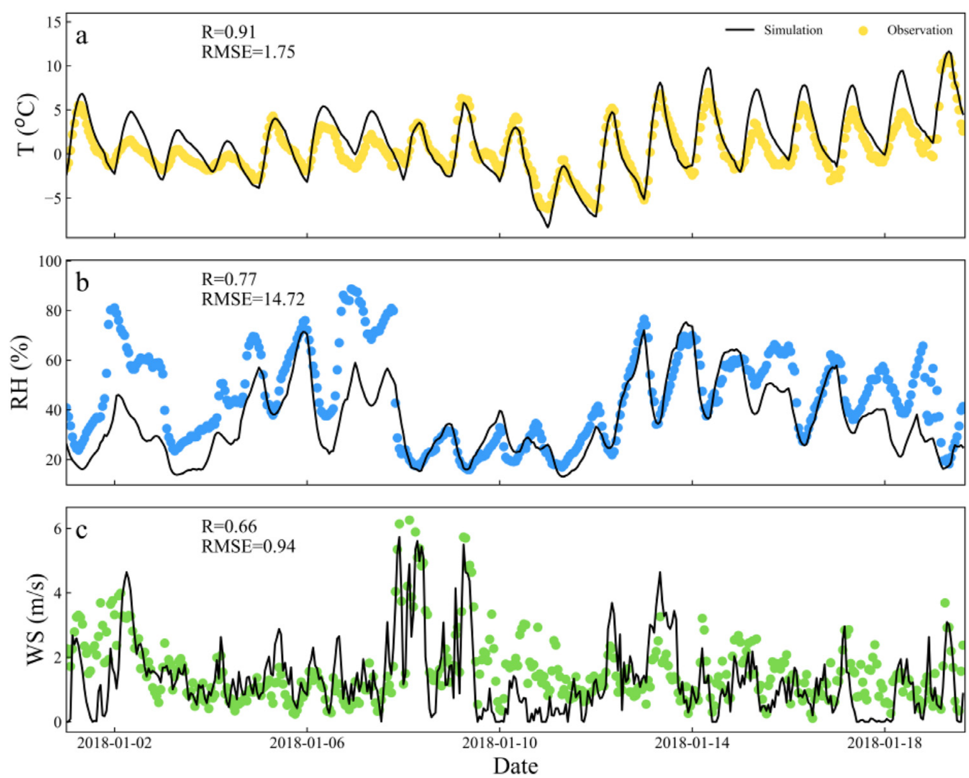

Fig. 2 – Comparison of simulated and observed meteorological conditions in Shahe.

was run to provide the meteorological conditions that were input into the NAQPMS. The NAQPMS and WRF have the same horizontal resolutions and three nested domains (45-, 15-, and 5-km). Shahe and other cities within the NCP were included in the third domain (Fig. 1a). Vertically, the NAQPMS model used 20 terrain-following layers that range from the surface up to 20-km; there were approximately ten layers under 2.5-km. The simulation was conducted from December 26, 2017 to January 20, 2018; the first six days were used as the model spin-up time. Emissions data from the glassmaking factories in Shahe was used to update the Multiresolution Emission Inventory for China (MEIC, http://www.meicmodel.org/). The updated anthropogenic emissions were used in this study.

2. Results and discussion

2.1. *Model evaluation of meteorological data and air pollutant measurements*

The hourly observed and simulated air temperature (T), RH, and WD are displayed in Fig. 2. The WRF model reproduced the meteorological conditions at an acceptable level. The correlation coefficients of T, RH, and WD between the observed and simulated data were 0.91, 0.77, and 0.66, respectively. The root mean square error (RMSE) of T, RH, and WS was 1.75 °C, 14.72%, and 0.94 m/sec, respectively. The NAQPMS model was run to reproduce the change in pollutant concentrations in Shahe. As

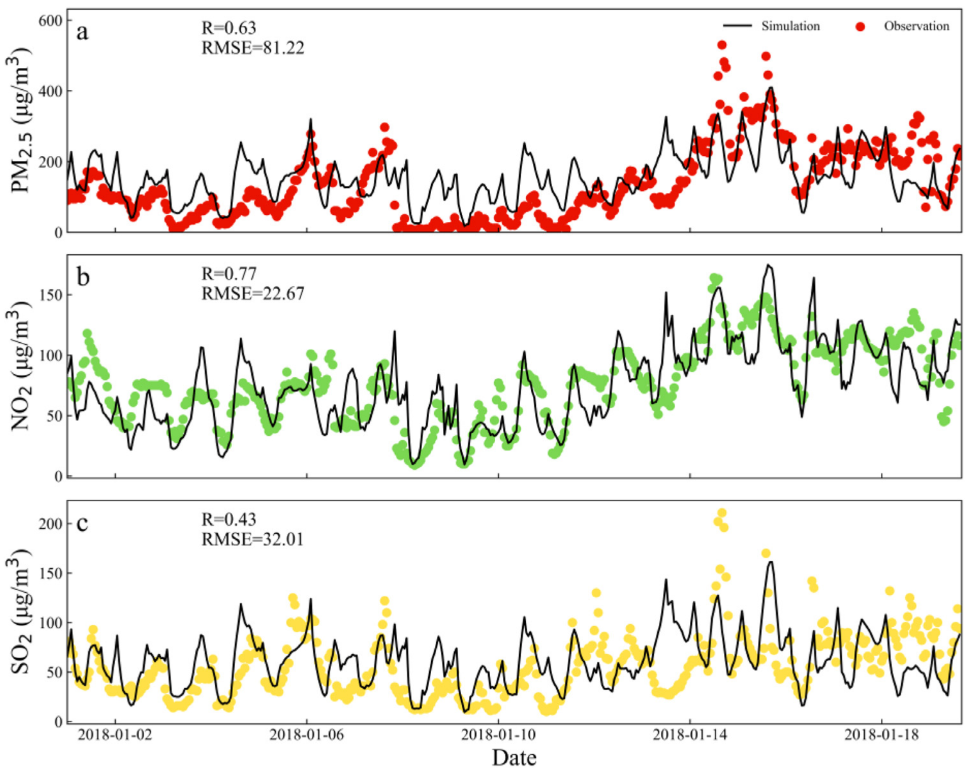

Fig. 3 – Comparison of simulated and observed pollutants in Shahe.

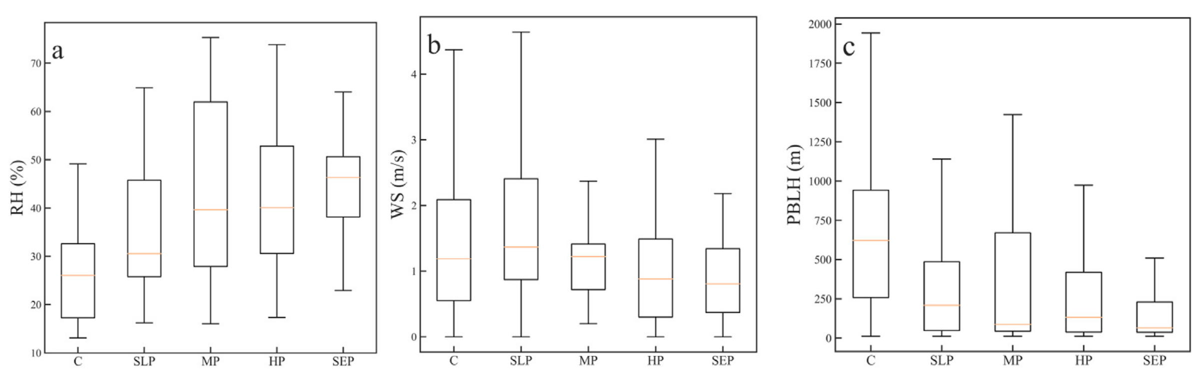

Fig. 4 – Variations in (a) RH, (b) wind speed, and (c) planetary boundary layer height under different $PM_{2.5}$ levels in Shahe from January 1 to 20, 2018.

shown in Fig. 3, the observed and simulated $PM_{2.5}$, NO_2, and SO_2 demonstrated strong correlation, with coefficients of 0.63, 0.77, and 0.43, respectively. There was also a strong agreement between the mass concentrations of the simulated and observed concentrations of $PM_{2.5}$, NO_2, and SO_2, which recorded RMSE values of 81.22, 22.67, and 32.01 $\mu g/m^3$, respectively. The mean mass concentrations of observed $PM_{2.5}$, NO_2, and SO_2 during the study period were 132.0, 76.1, and 55.5 $\mu g/m^3$, respectively.

2.2. *Impact of meteorological conditions on pollution*

Previous studies revealed that meteorological conditions of weak wind, high RH, and shallow planetary boundary layer height (PBLH) can promote the formation or accumulation of $PM_{2.5}$ (Wang et al., 2016; Zhang et al., 2018). We investigated the variations in $PM_{2.5}$ concentrations under different meteorological conditions in Shahe. Each hour within the study period was classified by pollution level, according to the hourly simulated $PM_{2.5}$ mass concentrations; the classification categories were designated as follows: clean (C, $PM_{2.5} < 75$ $\mu g/m^3$), slightly polluted (SLP, $75 \leq PM_{2.5} < 115$ $\mu g/m^3$), moderately polluted (MP, $115 \leq PM_{2.5} < 150$ $\mu g/m^3$), heavily polluted (HP, $150 \leq PM_{2.5} < 250$ $\mu g/m^3$), and severely polluted (SEP, $PM_{2.5} \geq 250$ $\mu g/m^3$). As shown in Fig. 4a, the $PM_{2.5}$ concentration level increased with rising RH. High RH can promote the oxidation of SO_2 and NO_x to sulfate and nitrate, respectively (Huang et al., 2014; Liu et al., 2020). In general, the $PM_{2.5}$ level was negatively related to wind speed (Fig. 4b). Weak wind is not conducive to pollutant diffusion. The planetary boundary layer (PBL) is another important meteorological factor in $PM_{2.5}$ pollution levels. PBL dynamics can influence wind shear, turbulence, and the vertical mixing of air pollutants (Lv et al., 2020). The shallow planetary boundary layer height (PBLH) increased the concentration of $PM_{2.5}$ in Shahe (Fig. 4c). Under SEP conditions, the average simulated PBLH was only 187 m, which is approximately one-fourth that of the clean period's PBLH. The correlation of RH, WS, and PBLH with $PM_{2.5}$ concentration was 0.49, −0.28, and −0.59, respectively. This suggests that PBLH was the most important factor in influencing $PM_{2.5}$ concentration. Overall, high RH, weak WS, and low PBLH represent unfavorable meteorological conditions for $PM_{2.5}$ pollution in Shahe.

2.3. Source identification of $PM_{2.5}$

2.3.1. Geographical origins of $PM_{2.5}$

To identify the geographical origins of $PM_{2.5}$ in Shahe, seven regions were demarcated (**Fig. S1**). Assuming the observation site is situated at the 0-km position, the distances of the seven demarcated regions to the observation site are as follows: 0–50 km, 50–100 km, 100–150 km, 150–200 km, 200–250 km, 250–300 km, and > 300 km, respectively. Following the demarcation of the regions, the NAQPMS model was run with an online source-tagging module (Lu et al., 2017). Fig. 5a shows the contributions of each of the seven regions to $PM_{2.5}$ pollution in Shahe during the study period. The contribution from the region located 0–50 km from the sampling site was much higher than the contributions from other regions, providing an average contribution of 63.4% to the total $PM_{2.5}$ levels. At times during the study that were characterized by severe pollution ($PM_{2.5} \geq 250$ μg/m^3), this contribution increased to 73.9%±21.2%. The 0–50 km region covers an area of approximately 8000 km^2, and includes southern Xingtai and northern Handan. The total contributions of the first six regions account for 89.2% of total $PM_{2.5}$ pollution in Shahe. This indicates that Shahe is influenced by local emissions and regional transport across a relatively short distance. When the mass concentration of $PM_{2.5}$ was 50–325 μg/m^3 in Shahe, the contribution from > 300 km away from the observation site increased. **Fig. S2** displays the contributions of seven regions to $PM_{2.5}$ concentrations in Shahe under different scenarios of $PM_{2.5}$ pollution. The contribution of the first region to sulfate, nitrate, ammonium, and primary $PM_{2.5}$ were 42.6%, 35.9%, 40.6%, and 76.4%, respectively. As $PM_{2.5}$ concentration increased, the contribution to sulfate, nitrate, and ammonium pollution from the most remote area (> 300 km) increased.

To reveal the contribution of local emissions to $PM_{2.5}$, the emissions from Shahe were tagged, and the contribution was traced. During the study, the hourly contribution of local emissions to $PM_{2.5}$ in Shahe ranged from 4.2% to 91.4% (**Fig. S3**), with an average of 52.3±21.6%. The average local contribution in the C, SLP, MP, HP, and SEP categories was 62.0%, 52.7%, 48.6%, 51.7%, and 52.2%, respectively (**Fig. S4**). This indicates that regional transport from outside Shahe contributes to an increase in haze. The primary and secondary components contributed 36.1% and 16.2% to $PM_{2.5}$ concentration in Shahe from local emissions during the entire study (Fig. 5b). With the increase of $PM_{2.5}$ concentrations, the contribution from the primary components decreased, while that from the secondary components increased. We inferred that haze conditions occurred when RH was high and WS was relatively weak (Fig. 4a and b); these conditions were not conducive to the diffusion of primary pollutants but were conducive to the formation of secondary components.

2.3.2. Source apportionment

Four categories of emissions sources were used to identify the source apportionment of $PM_{2.5}$ in Shahe; the categories include: residential (residential combustion and biomass combustion), industry, power plant, and transportation (on-road and off-road vehicles). Fig. 6 displays the source apportionment for the primary and secondary components of $PM_{2.5}$ in Shahe. Industry emissions were the largest contributor to the primary components in Shahe, accounting for an average of approximately 62% of the primary components. The second greatest contributor to the primary components was the residential sector, with a mean contribution of 25%. When the mass concentration of $PM_{2.5}$ increased, the contribution to the primary components from the industry and power plant sectors decreased, while that from residents and transportation increased. Industry emissions were also the largest contributor to the secondary components. In addition to industry emissions, both residential emissions and power plant emissions were important, contributing an average of 27% and 24%, respectively, to the secondary components of $PM_{2.5}$ in Shahe. In sum, residential, industry, power plant, and transportation emissions contributed an average of 26%, 47%, 17%, and 10%, respectively, to the $PM_{2.5}$ concentration in Shahe.

As shown in Fig. 7, the region from 0 to 50 km was the most important contributor to the $PM_{2.5}$ in Shahe from a geographical standpoint; this region contributed 57%, 63%, and 53% to the $PM_{2.5}$ concentration in the industry, power plant, and residential sectors. The region of 0–50 km covers Shahe, Xingtai, Handan, and Anyang, all of which are characterized by intensive industrial activity (Sun et al., 2020a, 2020b; Xiao et al., 2020). As to $PM_{2.5}$ concentration in the transportation sector, the contributions from the region of 0–50 km and > 300 km were 37% and 34%, respectively. We inferred that the two megacities (Beijing and Tianjin) located in the region > 300 km from the sampling site contributed to high emissions because of the intense traffic emissions that are located there.

2.4. Influence of industry emissions

Glass manufacturing is the most important industry in Shahe City, and its operations produce substantial emissions. To investigate the influence of glass factories' emissions discharge, the emissions from glass manufacturing were tagged and tracked. Fig. 8 displays the average $PM_{2.5}$ concentrations that the glass factories emitted in Shahe over the duration of the entire study. The glass enterprise emissions mainly influenced Shahe and its surrounding cities. It was determined that glass manufacturing contributed 32.1% of the $PM_{2.5}$ concentration to Shahe during the study period. Glass factories also increased $PM_{2.5}$ concentration in nearby cities (including Yangquan, Jinzhong, Changzhi, Handan, and Anyang) by 0–20 μg/m^3 (Fig. 8); the variable increase was dependent on

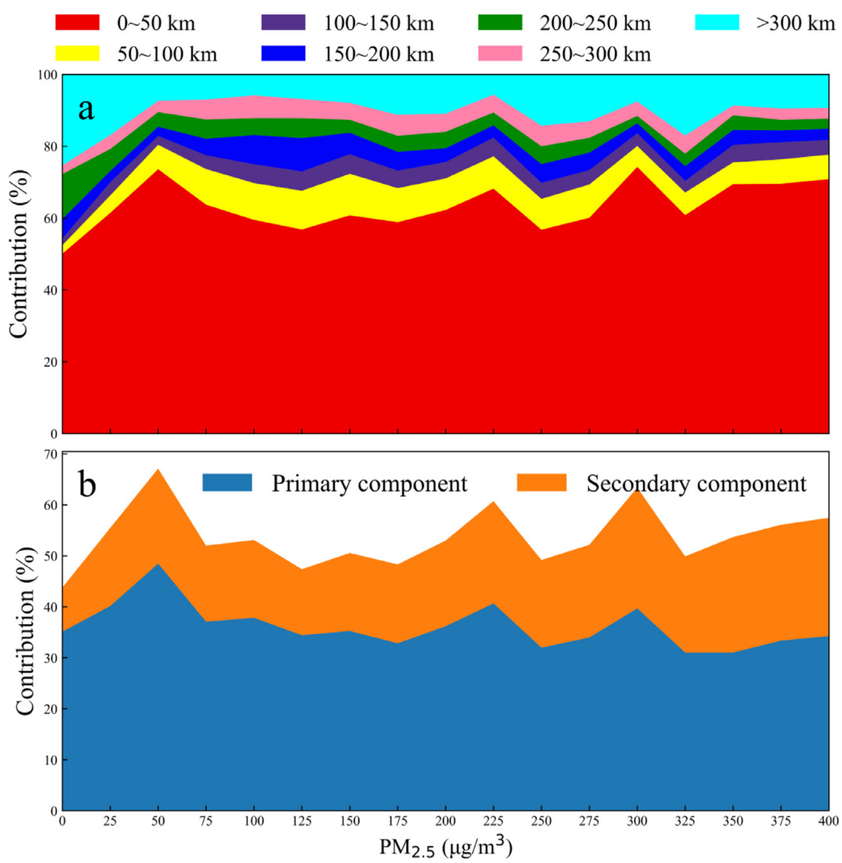

Fig. 5 – (a) The average percentage contribution of different regions to $PM_{2.5}$ concentration in Shahe, (b) The average percentage contribution of the primary component and secondary component to $PM_{2.5}$ concentration from local emissions in Shahe.

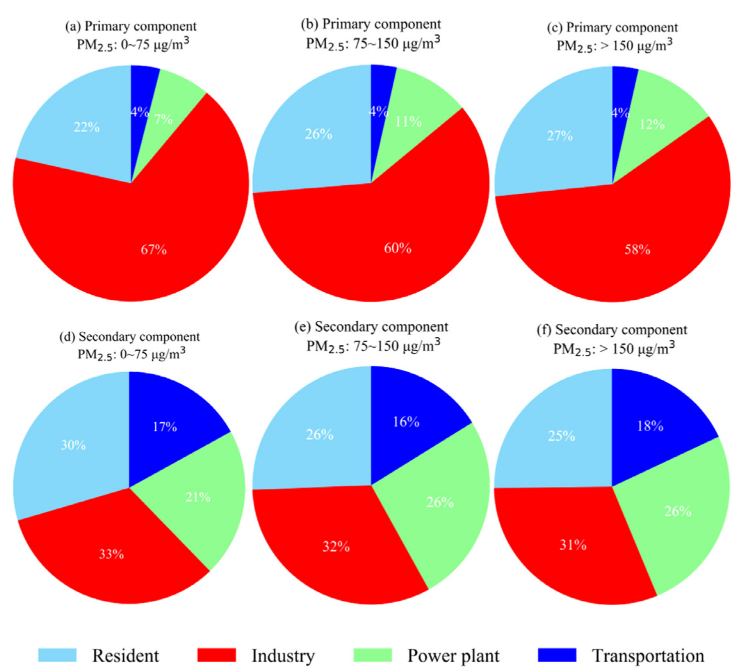

Fig. 6 – The contributions of source categories to the primary and secondary components under different $PM_{2.5}$ concentrations in Shahe.

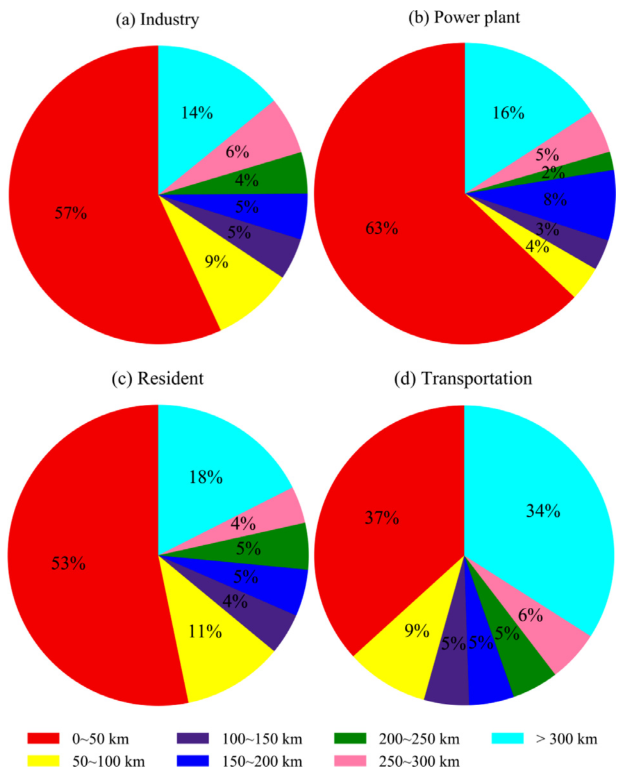

Fig. 7 – The contributions of regions to source categories in Shahe.

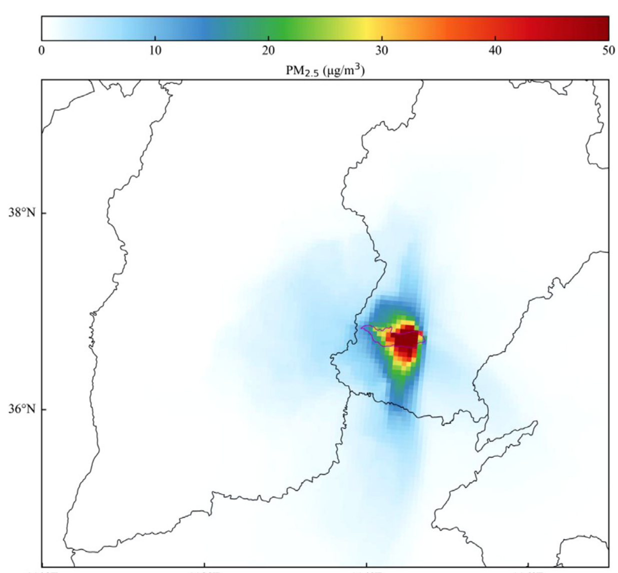

Fig. 8 – The average $PM_{2.5}$ concentrations contributed by the glass factories in Shahe from January 1 to 20, 2018.

the cities' distance from the emissions source and the meteorological conditions. Under different pollution levels, the $PM_{2.5}$ contributed by the glass factories showed different distribution characteristics (Fig. 9). In the clean period ($PM_{2.5} \leq 75$ $\mu g/m^3$), the mean simulated wind speed and PBLH were 2.3±1.6 m/sec and 1260.0±441.8 m, respectively (Fig. 4). The

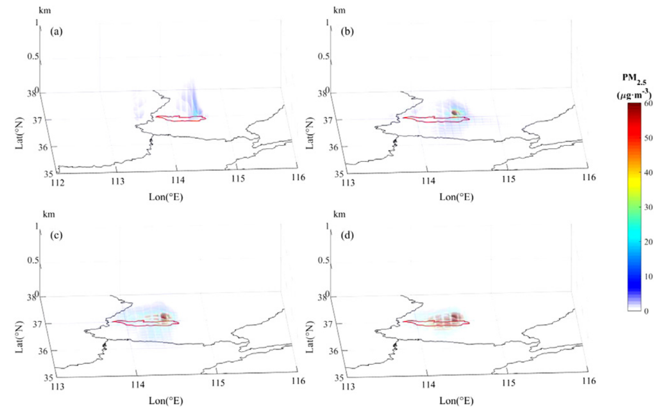

Fig. 9 – Three-dimensional distribution of $PM_{2.5}$ contributed by the glass factories during (a) $PM_{2.5} \leq 75$ $\mu g/m^3$, (b) $75 < PM_{2.5} \leq 150$ $\mu g/m^3$, (c) $150 < PM_{2.5} \leq 250$ $\mu g/m^3$, and (d) $PM_{2.5} > 250$ $\mu g/m^3$.

Table 1 – Results of emission reduction experiment.

Case	Reduction regions	Sectors	Reduction level	$PM_{2.5}$ in Shahe ($\mu g/m^3$)
1	Shahe	All	0	155.7±69.8
2	Shahe	All	20%	137±61.6
3	Shahe	All	40%	118.7±54.3
4	Shahe	All	60%	100.5±48.5

relatively high PBLH and strong wind speed enhanced the horizontal and vertical diffusion of $PM_{2.5}$. The area influenced (112.1 °E-114.6 °E) by the glass factories covered a relatively large expanse, with a low $PM_{2.5}$ contribution of 0–10 $\mu g/m^3$ (Fig. 9a). As $PM_{2.5}$ concentration increased, the PBLH decreased and wind weakened (Fig. 4), thereby allowing the $PM_{2.5}$ that was contributed by the glass factories to start to accumulate vertically (Fig. 9b–d). The chimney heights of glass factories in Shahe are usually less than 70 m. In the hours of the study that were characterized by severe pollution, the height impacted by the glass factories increased to approximately 180 m (Fig. 9d). In the horizontal direction, the influence area of the glass factories narrowed as $PM_{2.5}$ concentration increased. In severely polluted hours (Fig. 9d), the affected area with a $PM_{2.5}$ concentration higher than 50 $\mu g/m^3$ mainly covered Shahe city. This suggests that the reduction of glass factory emissions in Shahe could improve the air quality of the local area and surrounding cities.

2.5. *Emissions reduction simulations*

As previously discussed, the $PM_{2.5}$ concentration in Shahe was greatly influenced by local emissions. To provide possible emissions reduction strategies, experiments were conducted to simulate a reduction in pollutant emissions by 20%, 40%, and 60%. As listed in Table 1, under the different emissions reduction scenarios, the mass concentrations of $PM_{2.5}$ in Shahe decreased 18.6, 37.0, and 55.2 $\mu g/m^3$, respectively. This indicates that the implementation of local emissions reductions is an effective way to reduce the $PM_{2.5}$ concentration in Shahe. We also suggest that joint emissions controls with the cities around Shahe are also needed to achieve better air quality.

3. Conclusions

In this study, the air quality of an industrial city dominated by the glass manufacturing industry in the NCP was investigated using a three-dimensional nested air quality condition model named NAQPMS. The $PM_{2.5}$ concentration from January 1 to 20, 2018, was simulated using the model. The simulated result showed that the model reproduced the measured pollutant changes in Shahe to an acceptable level. The relationship between meteorological conditions and $PM_{2.5}$ concentration was analyzed. We found that high RH, weak wind, and low PBLH represent unfavorable meteorological conditions for $PM_{2.5}$ pollution in Shahe. To identify the source of $PM_{2.5}$ in Shahe, NAQPMS was performed with an online source-tagging module. The region within a 50 km radius of downtown Shahe contributed the most $PM_{2.5}$ to the city, with an average contribution of 63.4%. During the most severely polluted ($PM_{2.5} \geq 250$ $\mu g/m^3$) hours of the study, this contribution increased to 73.9±21.2%. Furthermore, the contribution from Shahe was identified as 52.3%±21.6%. The primary and secondary components contributed 36.1% and 16.2% to the $PM_{2.5}$ concentration, respectively. Source apportionment results revealed that resident, industry, power plant, and transportation emissions contributed 26%, 47%, 17%, and 10%, respectively, to the $PM_{2.5}$ concentration in Shahe. The influence of glass factories' emissions in Shahe was also investigated. Generally, the glass factories contributed 32.1% of the $PM_{2.5}$ concentration in Shahe. With the increase of $PM_{2.5}$ concentration, the influence of the glass factories accumulated vertically and narrowed horizontally. In severely polluted hours, the horizontally affected area with a $PM_{2.5}$ concentration higher than 60 $\mu g/m^3$ which was contributed by the glass factories mainly covered Shahe. The study also identified three idealized emissions reduction scenarios. In the experiments, simulations for 20%, 40%, and 60% emissions reduction in Shahe were performed; the results indicate that the mean mass concentration of $PM_{2.5}$ in Shahe decreased by 12.0%, 23.8%, and 35.5%, respectively. We suggest that joint emissions controls between Shahe and surrounding cities are needed to improve regional air quality.

Acknowledgments

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Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2021.03.008.

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