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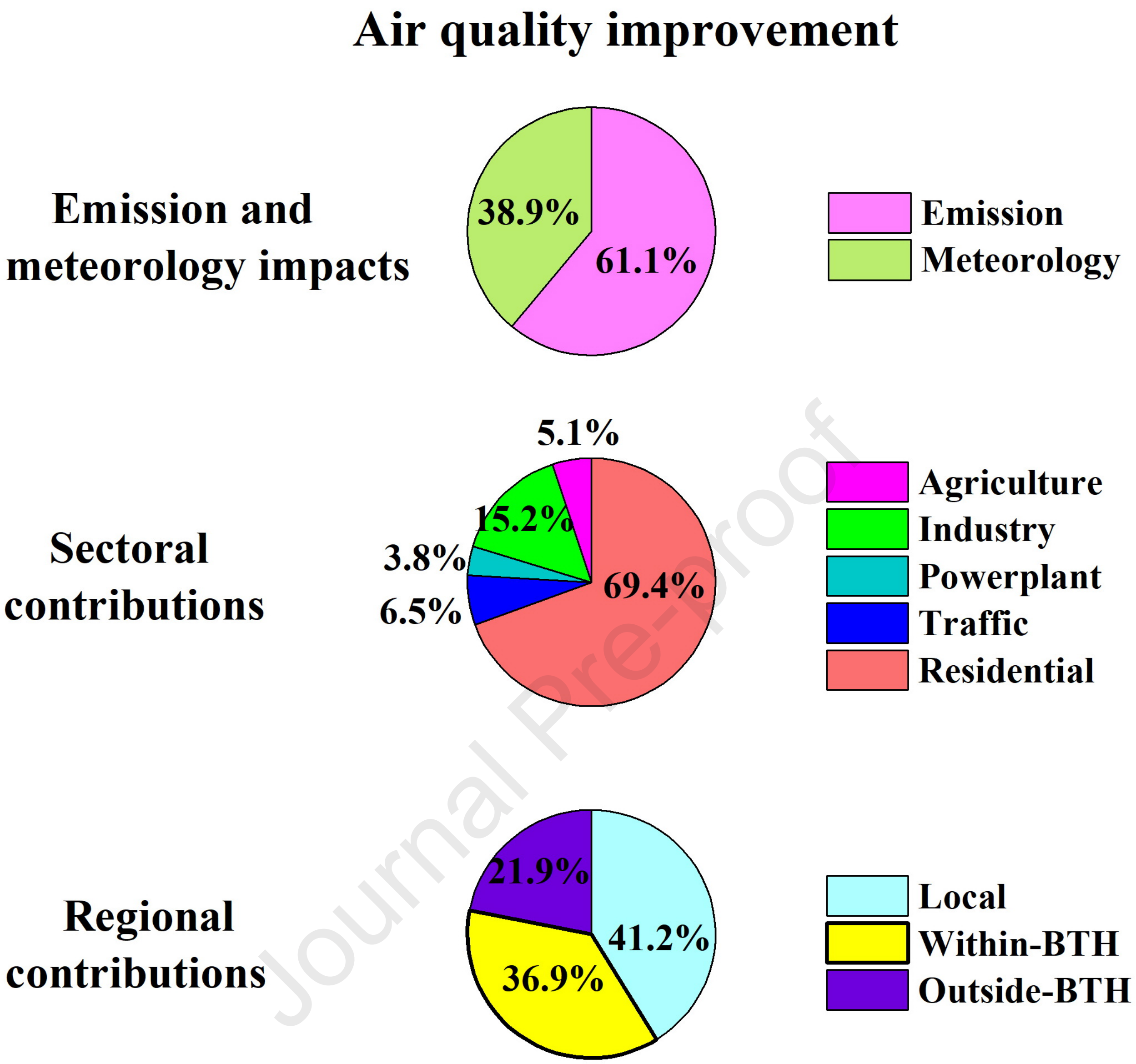
## Credit authorship contribution statement

S.Y., and Y.Z. conceived and designed the research. Y.Z and S.Y. performed model simulations and the data analyses. X.C., M.L., L.W., P. L. Z.L., W. L., and D.R. contributed to the scientific discussions. S.Y., Y.Z. and J.H.S wrote the paper.


Air quality improvement
Emission and meteorology impacts
38.9%
61.1%
Emission
Meteorology
Sectoral contributions
5.1%
15.2%
3.8%
6.5%
69.4%
Agriculture
Industry
Powerplant
Traffic
Residential
Regional contributions
21.9%
41.2%
36.9%
Local
Within-BTH
Outside-BTH

# City-level air quality improvement in the Beijing-Tianjin-Hebei region from 2016/17 to 2017/18 heating seasons: attributions and process analysis

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# Abstract

With the implementation of clean air strategies, $PM_{2.5}$ pollution abatement has been observed in the "2+26" cities in the Beijing-Tianjin-Hebei (BTH) region (referred to as the BTH2+26) and their surrounding areas. To identify the drivers for $PM_{2.5}$ concentration decreases in the BTH2+26 cites from the 2016/17 heating season (HS1617) to the 2017/18 heating season (HS1718), we investigated the contributions of meteorological conditions and emission-reduction measures by Community Multi-Scale Air Quality (CMAQ) model simulations. The source apportionments of five sector sources (i.e., agriculture, industry, power plants, traffic and residential), and regional sources (i.e., local, within-BTH: other cities within the BTH2+26 cities, outside-BTH, and boundary conditions (BCON)) to the $PM_{2.5}$ decreases in the BTH2+26 cities were estimated with the Integrated Source Apportionment Method (ISAM). Mean $PM_{2.5}$ concentrations in the BTH2+26 cities substantially decreased from 77.4-152.5 μg $m^{-3}$ in HS1617 to 52.9-101.9 μg $m^{-3}$ in HS1718, with the numbers of heavy haze (daily $PM_{2.5}$ ≥150 μg $m^{-3}$) days decreasing from 17-77 to 5-30 days. The model simulation results indicated that the $PM_{2.5}$ concentration decreases in most of the BTH2+26 cities were attributed to emission reductions (0.4-55.0 μg $m^{-3}$, 2.3-81.6% of total), but the favorable meteorological conditions also played important roles (1.9-25.4 μg $m^{-3}$, 18.4-97.7%). Residential sources dominated the $PM_{2.5}$ reductions, leading to decreases in average $PM_{2.5}$ concentrations by more than 30 μg $m^{-3}$ in severely polluted cities (i.e., Shijiazhuang, Baoding, Xingtai, and Beijing). Regional source analyses showed that both local and within-BTH sources were significant contributors to $PM_{2.5}$ concentrations for most cities. Emission controls in local and within-BTH sources in HS1718 decreased the average $PM_{2.5}$ concentrations by 0.1-47.2 μg $m^{-3}$ and 0.3-22.1 μg $m^{-3}$, respectively, relative to those in HS1617. Here we demonstrate that a combination of favorable meteorological conditions and anthropogenic emission reductions contributed to the improvement of air quality from HS1617 to HS1718 in the BTH2+26 cities.

**Key words** BTH2+26 cities, air quality improvement, source apportionment, emission reduction, meteorological contribution

# 1. Introduction

Along with rapid urbanization, population growth and industrialization, air pollution has become a prominent environmental problem in China, which poses an urgent threat to social economy, ecological environment and human health (Huang et al., 2015; Van Donkelaar et al., 2015; Rosenfeld et al., 2019; Tilt, 2019). High concentrations of $PM_{2.5}$ (particulate matter with aerodynamic diameter ≤ 2.5 μm) are especially prevalent. The Beijing-Tianjin-Hebei (BTH) urban agglomeration in China, has faced persistent and serious air pollution for a long time. For example, in January 2010, the maximum hourly concentrations of $PM_{2.5}$ in Tianjin in the BTH region exceeded 400 μg $m^{-3}$ (Zhao et al., 2013). In a heavy and persistent pollution episode in January 2013, the observed maximum hourly concentrations of $PM_{2.5}$ in Beijing, Tianjin and Shijiazhuang were 680, 500 and 660 μg $m^{-3}$, respectively (Wang et al., 2014). The $PM_{2.5}$ mean concentrations in Shijiazhuang in winter during 2012-2014 reached 105 μg $m^{-3}$, three times the national standard of 35 μg $m^{-3}$ in China (Liu et al., 2018). Frequent and persistent severe $PM_{2.5}$ pollutions in the BTH region have been well documented (Wang et al., 2014; Zhang et al., 2014; Zheng et al., 2016; Zhang et al., 2019a, d).

To mitigate severe air pollution problems, the government of China launched the Air Pollution Prevention and Control Action Plan in 2013. Under the implementation action, the BTH region, Yangtze River Delta, and Pearl River Delta were designated as three key target regions to decrease $PM_{2.5}$ concentrations. A series of strict air pollutant controls and emission reduction measures were enacted, such as eliminating industries and enterprises with high pollution and emissions, promoting desulfurization in industrial facilities, adopting clean fuels to replace coal combustion, optimizing industrial and energy structures, and enhancing industrial and vehicle emission standards. In response to these emission-reduction efforts, significant air quality improvements are evident from satellite data and field observations (Cheng et al., 2019; Li et al., 2019a; Xue et al., 2019). For instance, the annual mean concentrations of $PM_{2.5}$ in Beijing declined from 89.5 μg $m^{-3}$ to 58 μg $m^{-3}$ from 2013 to 2017, and the annual mean concentrations of $PM_1$ (particulate matter with aerodynamic diameter ≤ 1 μm) decreased from 66.2 μg $m^{-3}$ to 33.4

μg $m^{-3}$ in winter from 2014 to 2017 in Beijing (Cheng et al., 2019; Li et al., 2019a). From 2013 to 2017, the mean population-weighted $PM_{2.5}$ concentrations in the BTH region decreased by 37% (from 102.8 to 66.1 μg $m^{-3}$) with the proportion of polluted days (daily $PM_{2.5}$ concentration >75 μg $m^{-3}$) declining from 56.1% to 27.0% (Xue et al., 2019). Nonetheless, heavy $PM_{2.5}$ pollutions still frequently occurred in the BTH region. In light of the influences of meteorological conditions on $PM_{2.5}$ variations, significant contributions of regional transports to $PM_{2.5}$ in the downwind areas occurred. For example, on the basis of simulations for assessment of inter-city transports, Chang et al. (2018) found that Zhangjiakou and Baoding contributed 57% to the total inflow influx of $PM_{2.5}$ in Beijing in winter. To accomplish the integrated pollution source control targets, the government of China issued the “2017 Air Pollution Prevention and Management Plan for the Beijing-Tianjin-Hebei region and its surrounding Areas”, in which the “BTH2+26” cities were defined as the BTH air pollution transport channel cities (MEP, 2017). The BTH2+26 cities cover less than 3% of China’s area, but emit more than 15% of the entire country’s primary $PM_{2.5}$, 10% of $SO_2$ and $NO_x$, and 8% of VOCs (MEP, 2017). With high population density and intensive distributions of industries, the BTH2+26 city cluster has become the most polluted area across China, especially during wintertime (Zhang et al., 2013; Zhong et al., 2019). Based on observations of 169 cities carrying out air quality monitoring stations in China in 2018, 13 of 20 cities with the worst air quality in China belong to the BTH2+26 cities (MEE, 2018).

$PM_{2.5}$ concentrations are closely associated with meteorological conditions and emissions (He et al., 2017; Bao et al., 2019; Ma et al., 2017; Zhang et al., 2019c, d). Huang et al. (2017) suggested that the weather conditions (such as weak winds) during the 2nd Youth Olympic Games increased the mean $PM_{2.5}$ concentrations by 7.8% in Nanjing. Favorable meteorological conditions associated with higher wind speeds, higher boundary layer height and lower humidity contributed to the $PM_{2.5}$ decreases. By comparing influences of different synoptic patterns on wintertime haze formation in the BTH region, Bei et al. (2020) found that when northeast or northwest winds prevailed, $PM_{2.5}$ concentrations were more likely to decrease. In comparison with meteorology, emission reductions can be regarded as the more critical driving force for $PM_{2.5}$ decreases. For example, Cheng et al. (2019) showed that rapid $PM_{2.5}$ decreases in Beijing from 2013 to 2017 were dominantly contributed by local emission reductions. Xu et al. (2020) confirmed the major role of emission reductions in the continuous decreases of $PM_{2.5}$

concentrations during 2014-2017 across China. Similar conclusions are obtained from the ground-based observations and model simulations (Guo et al., 2018; Li et al., 2019a; Zhang et al., 2019b, c; Zhang and Geng, 2019). Annual mean concentrations of $PM_{2.5}$ in the BTH region decreased by 9.9% in 2017 relative to 2016 and 11.8% in 2018 relative to 2017 (MEE2017; MEE, 2018). Air quality improvements in the BTH region have been reported, largely focused on short heavy pollution episodes or a single city (e.g., Chen et al., 2019a; Gao et al., 2018). Quantitative analyses of driving factors for air pollution abatements in the BTH region during long time periods have remained limited. Here, we evaluate evidences of $PM_{2.5}$ decreases in the BTH2+26 cities from 2016/17 heating season (labeled as HS1617) to 2017/18 heating season (labeled as HS1718), and quantitatively estimate the contributions of meteorological condition changes and emission reductions to $PM_{2.5}$ abatements. We further investigate the contributions of emission controls associated with sectoral and regional sources to the $PM_{2.5}$ concentrations in the BTH2+26 cities.

## 2. Experimental

### 2.1 Study areas

The major 28 cities (i.e., 2+26 cities) in the BTH region (referred to as the BTH2+26 cities) located in the air pollution transport channels are listed as follows: 2 (Beijing, Tianjin), 26 (Anyang, Baoding, Binzhou, Cangzhou, Changzhi, Dezhou, Hebi, Handan, Hengshui, Heze, Jincheng, Jinan, Jining, Jiaozuo, Kaifeng, Liaocheng, Langfang, Puyang, Shijiazhuang, Tangshan, Taiyuan, Xingtai, Xinxiang, Yangquan, Zibo, and Zhengzhou). The location of each city in the BTH2+26 cities is presented in Fig. 1.

### 2.2 Observation data

In this study, October, November, and December in the current year, and January, February, and March in the following year are defined as the autumn-winter heating season. Hourly $PM_{2.5}$ concentrations in HS1617 and HS1718 in the BTH2+26 cities were obtained from National Environmental Monitoring Center (CNEMC, http://www.cnemc.cn/). Official $PM_{2.5}$ data were released by Ministry of Ecology and Environment of the People's Republic of China.

Observations from 154 monitoring sites in the BTH2+26 cities were used for model performance evaluation and data analyses. For the evaluation of model performances, the model results at each monitoring site in each city were retrieved and the comparisons between mean values of observations and the model results for all monitoring sites in each city were carried out as in Zhang et al. (2019) and Chang et al. (2018). The hourly measurements of meteorological parameters (temperature, wind speed and relative humidity (RH)) used in this study were obtained from the National Climate Data Center (NCDC) (ftp://ftp.ncdc.noaa.gov/pub/data/noaa/).

## 2.3 Model configuration

The WRF/CMAQ-ISAM model was applied to simulate $PM_{2.5}$ concentrations and provided information about $PM_{2.5}$ source apportionments. The WRFv3.7（Weather Research and Forecast）model was developed by the National Center for Atmospheric Research and was used to provide meteorological fields for chemical simulations. The CMAQ (Community Multiscale Air Quality) model was selected to simulate pollutant concentration evolutions and has been widely used in simulations and predictions for ozone, particulate matter, toxics, and acid deposition (Kwok et al., 2013; Yu et al., 2014, Wu et al., 2018). Physical parameterizations and chemical options in the WRF/CMAQ model were the same as those in Wu et al. (2018). ISAM (Integrated Source Apportionment Method) was used to track contributions of industrial and regional sources to atmospheric pollutant concentrations (Kwok et al., 2013).

Anthropogenic emissions were obtained from MEIC (Multi-resolution emission inventory for China) made by Tsinghua University for the base year of 2016. Primary atmospheric species, including $PM_{2.5}$, $PM_{10}$ (particulate matter with aerodynamic diameter $\leq$ 10 μm), $SO_2$, $NO_x$, CO, VOCs, $NH_3$, BC, and OC emitted from five emission sectors (i.e., industry, power plants, transportation, agriculture and residential), were considered. Emissions in the BTH2+26 cities and their surrounding regions were updated to 2017 and 2018 based on the bottom-up inventory supplied by Beijing Municipal Research Institute of Environmental Protection (Zheng et al., 2018).

A two-way nesting simulation was employed at a horizontal resolution of 36 km in the outer domain (D01) and 12 km in the inner domain (D02). D01 covered eastern and central parts of China and D02 covered the BTH regions including the BTH2+26 cities as shown in Fig. 1. Meteorological and initial lateral boundary conditions were provided by global tropospheric analyses data (FNL) with a spatial resolution of 1° and temporal resolution of 6 h. The default initial and boundary chemical conditions were used for the D01 domain, and the simulation results of the D01 domain were used for the D02 domain.

Source apportionments of $PM_{2.5}$ in HS1617 and HS1718 were performed using the WRF-CMAQ/ISAM. The flow chart of the attributions and process analyses for the air quality improvements in the BTH region from HS1617 to HS1718 is shown in Fig. 2. To examine the contributions of meteorological conditions and emission reductions to $PM_{2.5}$ changes, two base case (Case A and Case B) and one fixed-emission sensitivity experiments (Case C) were employed. In Case A (Case B), actual anthropogenic emission inventory and meteorological conditions of HS1617 (HS1718) were used. In Case C, the same anthropogenic emissions as the Case A were used but with the meteorological fields of Case B. Therefore, the differences between Case A and Case C (Case B and Case C) can be used to represent the impacts of meteorological changes (emission reductions) on $PM_{2.5}$ (Chen et al., 2019a; Cheng et al., 2019). For each simulation, the results of 6 days as model spin-up time were discarded.

# 3. Results and discussion

## 3.1. Model evaluation

The comparisons of observed and simulated values for daily mean meteorological parameters (temperature, wind speed and relative humidity (RH)) and $PM_{2.5}$ concentrations in HS1617 and HS1718 in six major cities are presented in Fig. 3. The six major cities refer to two municipalities (Beijing and Tianjin), and four provincial capital cities (i.e., Shijiazhuang, Jinan, Taiyuan, and Zhengzhou). Fig. 3 shows that the model well captured the variations of temperature at two meters (T2) with normalized mean bias (NMB) (Yu et al., 2006) of 0.07%, -0.49%, -0.60%, -0.33%, -0.30% and -0.01% for Beijing, Tianjin, Shijiazhuang, Jinan, Taiyuan and Zhengzhou, respectively. The correlation coefficients (R) ranged from 0.96 to 0.98, showing the good correlations between observations and simulations for T2. The model overestimated RH for

Tianjin in HS1718, but showed good performance in the simulations of RH for other five major cities, with NMB values ranging from -23.77% to -1.70%. The simulations showed good correlations with the observed RH with R values ranging from 0.72 to 0.89. Wind speeds (WS) in the six major cities were overestimated, which is a common phenomenon in model simulations for meteorology (with NMB values of 5-50%) (Chen et al., 2019a; Zhang et al., 2006). The time series of simulations for T2, RH and WS in Fig. 3 showed good agreements with observations, indicating that the WRF model reproduced reasonable meteorological conditions for the $PM_{2.5}$ simulations.

The NMB (R) values for $PM_{2.5}$ were 5.61% (0.71), 2.43% (0.78), -3.54% (0.79), 2.12% (0.79), 8.78% (0.59) and 3.42% (0.84) for Beijing, Tianjin, Shijiazhuang, Jinan, Taiyuan and Zhengzhou, respectively. The daily time series and evaluation results showed that the model reproduced the temporal and spatial variations of the observed $PM_{2.5}$ concentrations well in HS1617 and HS1718 in the six major cities.

## 3.2. $PM_{2.5}$ improvement evidences from observations and simulations

Observed and simulated mean concentrations of $PM_{2.5}$ and the numbers of heavy haze (daily $PM_{2.5}$ concentrations higher than 150 μg $m^{-3}$) days in each city in HS1617 and HS1718 are presented in Fig. 4. According to observations and simulations, the BTH2+26 cities suffered from severe $PM_{2.5}$ pollution with higher $PM_{2.5}$ concentrations and more frequently occurring heavy haze episodes in HS1617 relative to those in HS1718. The observations showed that in the BTH2+26 cities, the mean $PM_{2.5}$ concentrations declined dramatically from 77.4-152.5 μg $m^{-3}$ in HS1617 to 52.9-101.9 μg $m^{-3}$ in HS1718, with the percentage decreasess of 5.7%-42.5%. Noticeable decreases of the heavy haze pollution episodes were observed in the BTH2+26 cities. In HS1617, there were 17-77 (7.1-42.3%) heavy haze days, while in HS1718, the numbers of heavy haze days decreased to 5-30 (2.8-16.5%), as shown in Fig. 4. By comparing $PM_{2.5}$ concentrations in HS1718 to those in HS1617, the most notable improvements were observed in Shijiazhuang, Baoding, Beijing and Langfang, with the average $PM_{2.5}$ concentrations decreased by 61.6, 56.2, 39.2 and 37.6 μg $m^{-3}$ (40.4%, 39.8%, 42.6% and 39.2%), respectively, and the frequencies of heavy haze days decreased by 26.4%, 26.4%, 15.4% and 17.0%, respectively.

However, in Jincheng and Jining, the average $PM_{2.5}$ concentrations only decreased by 4.7 and 8.1 μg $m^{-3}$, respectively, with the percentages of heavy haze days remaining the same, suggesting the need for continuous pollution control efforts in these two cities.

Fig. 4 also shows the average differences between the observed and simulated $PM_{2.5}$ concentrations and frequencies of heavy haze days in HS1617 and HS1718. The average $PM_{2.5}$ concentrations in most of the 28 cities were underestimated and Shijiazhuang showed the largest negative bias with value of -17.5 μg $m^{-3}$ in HS1617. The numbers of heavy haze days also showed underestimations in most cities and the maximum negative bias (-16 days) occurred in Handan in HS1718. Overall, the simulated $PM_{2.5}$ concentration variations and the days of heavy haze in the BTH2+26 cities in HS1617 and HS1718 were acceptable and could be used for further analyses.

Although remarkable decreases of $PM_{2.5}$ concentrations and frequencies of heavy haze episodes were observed in most of the BTH2+26 cities, the average $PM_{2.5}$ concentrations were still at fairly high levels and the BTH2+26 cities were still facing considerable pressures in mitigating $PM_{2.5}$ pollution. The decomposition of $PM_{2.5}$ concentration decreases will provide scientific information about the formation of $PM_{2.5}$.

## 3.3 Contributions from meteorological conditions and emission reductions

As discussed in Section 3.2, both the observations and the simulations confirmed the $PM_{2.5}$ abatements in the BTH2+26 cities in HS1718 relative to those in HS1617. The decreases of $PM_{2.5}$ concentrations can be attributed to favorable meteorological conditions and anthropogenic emission reductions. According to previous reports, northwest winds were more favorable for the dilution of $PM_{2.5}$ than the southwest winds in the BTH region (Chang et al., 2018; Wang et al., 2019). The impacts of meteorological conditions on $PM_{2.5}$ concentrations varied with synoptic background and meteorological fields. To identify the effects of meteorological conditions and emission-reduction measures on the $PM_{2.5}$ abatements in the BTH2+26 cities, we designed two base case simulations (Cases A and B) and one emission-fixed sensitive simulation (Case C), as described in Section 2.3. $PM_{2.5}$ decreases due to meteorology were obtained by subtracting the results of Case C from those of Case A and the contributions of emission reductions were got by subtracting the results of Case C from those of Case B.

### 3.3.1. Meteorological conditions and emission differences

We compared the changes in main meteorological parameters between HS1617 and HS1718. Fig. 5 shows the average values of T2, RH, WS and planetary boundary-layer height (PBLH) in the BTH2+26 cities in HS1617 and HS1718. Compared with HS1617, the lower T2 and RH, and stronger atmospheric diffusion capacity associated with larger WS and higher PBLH values (Su et al., 2018) in HS1718 were not favorable for the heterogeneous formations of secondary aerosols, but were conductive to the dispersion of aerosols. The more favorable meteorological conditions in HS1718 than those in HS1617 contributed to $PM_{2.5}$ concentration decreases in the BTH2+26 cities.

Fig. 6 presents anthropogenic emissions of $SO_2$, $NO_x$, $NH_3$ and primary $PM_{2.5}$ in the BTH2+26 cities in HS1617 and HS1718 based on the bottom-up emission inventory. Anthropogenic emission amounts of $SO_2$ and primary $PM_{2.5}$ in the BTH2+26 cities from HS1617 to HS1718 were reduced by 3.5-69.5% and 2.2-63.4%, respectively. For Beijing, Tianjin, Shijiazhuang, Jinan, Taiyuan and Zhengzhou, $SO_2$ (primary $PM_{2.5}$) emissions from HS1617 to HS1718 were estimated to be reduced by 58.9% (18.1%), 35.7% (43.5%), 17.0% (27.8%), 63.2% (31.5%), 28.3% (12.5%), and 48.6% (23.7%), respectively. For $NO_x$ and $NH_3$ emissions, most cities experienced obvious decreases and specific cities experienced increases. The control of traffic had caused the $NO_x$ emissions to decrease by 13.1%, 8.7%, 4.7%, 27.4% and 18.7% in Beijing, Shijiazhuang, Jinan, Taiyuan and Zhengzhou, respectively, while $NO_x$ emissions in Tianjin increased by 2.28%. Due to the lack of effective control measures, $NH_3$ emissions vary greatly from city to city from HS1617 to HS1718. In some cities, $NH_3$ emissions were largely reduced, such as in Beijing (6.4%), Taiyuan (33.0%), and Zhengzhou (19.6%), while the $NH_3$ emissions showed increases in some other cities, such as Tianjin (5.0%), Shijiazhuang (0.2%) and Jinan (35.2%). Increases in $NH_3$ emissions partly counteracted the effects of reducing other emissions on decreasing $PM_{2.5}$ concentrations.

### 3.3.2. Contributions of meteorological conditions and emission reductions

Fig. 7a presents the concentration changes of $PM_{2.5}$, and the contributions from meteorological conditions and emission reductions averaged over six months in the BTH2+26 cities on the basis of model simulations. The spatial distributions of $PM_{2.5}$ concentration changes showed that the average $PM_{2.5}$ concentrations decreased in all BTH2+26 cities, being consistent with analyses presented in 3.1. The notable $PM_{2.5}$ concentration average changes were found in the western part of the BTH region, especially in Shijiazhuang due to the high $PM_{2.5}$ concentrations, as shown in Fig. 7a.

As discussed in 3.3.1, the meteorological conditions in HS1718 were beneficial for air quality improvement in the BTH2+26 cities relative to those of HS1617. The decreases of $PM_{2.5}$ concentrations in HS1718 contributed by favorable meteorological conditions and emission-reduction measures varied substantially in different cities, as shown in Fig. 7b. In the BTH2+26 cities, the average changes of $PM_{2.5}$ concentrations due to meteorological conditions and emission-reduction measures in HS1718 were estimated to be 1.9-25.4 μg $m^{-3}$ and 0.4-55.0 μg $m^{-3}$, respectively. The contributions of emission reductions to the $PM_{2.5}$ abatements in Beijing, Tianjin, Shijiazhuang, Jinan, Taiyuan and Zhengzhou were 26.6, 21.6, 55.0, 7.0, 13.9 and 15.1 μg $m^{-3}$, accounting for 68.3%, 71.3%, 68.4%, 50.4%, 44.6% and 54.7% of the total $PM_{2.5}$ reductions, respectively. The contributions of favorable meteorological conditions to the $PM_{2.5}$ decreases in these cities were 12.3, 8.7, 25.4, 6.9, 17.3 and 12.5 μg $m^{-3}$, respectively. Compared with the contribution of favorable meteorological conditions, emission-reduction measures were the dominant contributor for the $PM_{2.5}$ abatements in most cities, and the average $PM_{2.5}$ concentrations in Beijing, Tianjin, Anyang, Baoding, Handan, Shijiazhuang, and Xingtai were decreased by more than 20 μg $m^{-3}$ attributed to emission-reduction measures. Without emission reductions, the average $PM_{2.5}$ concentrations in Beijing, Tianjin, and Shijiazhuang in HS1718 would have increased to 83.6, 87.0, and 144.6 μg $m^{-3}$, respectively, leading to more economic loss and health risks to the public. However, for Heze, Jincheng and Jining, emission-reduction measures played a minor role and caused the $PM_{2.5}$ concentrations decreased by lower than 1 μg $m^{-3}$. It should be noted that the contributions of meteorological conditions might be overestimated due to the overestimations of the WS simulated by the model. The meteorology contribution results in this study agree well with the analysis of Zhang et al. (2019d), who proposed that the contributions of favorable meteorological conditions to the $PM_{2.5}$ abatements in the BTH region were 13% in 2017 relative to 2013 and 50% in 2017 relative to 2016.

In our study, the contribution of meteorology to $PM_{2.5}$ abatement in Beijing (with average concentration of 12.3 μg $m^{-3}$ and percentage of 31.7%) was higher than the result of Cheng et al. (2019) (with average concentration of 4.4 μg $m^{-3}$, and percentage of 29.5%). The large difference in the contributions of meteorological conditions was caused by the differences in the analysis periods. The study of Cheng et al. (2019) focused on the whole year, while our study focused on the heating season only. The effects of meteorological conditions on $PM_{2.5}$ variations were more striking in winter than in other seasons due to the meteorological conditions (Zhang et al., 2019d). In summary, the emission reductions in HS1718 were the dominant factor for the improvement of air quality in most of the BTH2+26 cities, and the favorable meteorological conditions played leading roles in reducing $PM_{2.5}$ concentrations in specific cities, such as Changzhi, Heze, Jincheng, Jiaozuo and Taiyuan. Zhang et al. (2019d) and Cheng et al. (2019) also confirmed the dominant role of emission reductions in $PM_{2.5}$ decrements in Beijing compared to favorable meteorological conditions. Since the favorable meteorological conditions in HS1718 had large contributions to decreasing $PM_{2.5}$ concentrations in the BTH2+26 cities, it was very important to implement emission-reduction measures in the BTH2+26 cities in order to continuously mitigate severe $PM_{2.5}$ pollution.

## 3.4. Contributions from sectoral emissions and regional sources

According to the results of ISAM source apportionments, we attributed the $PM_{2.5}$ abatements in the BTH2+26 cities in HS1617 and HS1718 to sectoral emissions and regional contributions. Fig. 8a shows the spatial contributions of emission reductions from agriculture, industry, power plants, traffic and residential sources to $PM_{2.5}$ decrements in the BTH2+26 cities in HS1718 relative to those in HS1617. The $PM_{2.5}$ contributed by agriculture and power plant sources in the BTH2+26 cities decreased by less than 2 μg $m^{-3}$ in HS1718 relative to HS1617. The major differences in the $PM_{2.5}$ concentrations between HS1617 and HS1718 can be attributed to industry, traffic and residential sources. The $PM_{2.5}$ decreases from traffic contributions were more notable in Beijing, Baoding and Shijiazhuang than those in other cities, suggesting the effectiveness of vehicle emission controls. The residential sources were the largest contributor to $PM_{2.5}$ concentration decreases in the BTH2+26 cities with decreases of the average $PM_{2.5}$ concentrations by more than 30 μg $m^{-3}$ for Beijing, Baoding, Hengshui, Xingtai, Shijiazhuang and Handan. It was worth noting that the decrease of $PM_{2.5}$ concentrations attributed to residential sources was more than 60 μg $m^{-3}$ in Shijiazhuang, suggesting the important role of residential emission reductions in the $PM_{2.5}$ abatements. In the processes of residential heating and cooking, large amount of black carbon and primary $PM_{2.5}$ components were emitted, resulting in the notable increase of $PM_{2.5}$ concentrations. It was estimated that during the heavy haze periods in western China, 62.2% of black carbon and 86.5% of primary organic aerosols were contributed by the residential sources (Yang et al., 2019). The dominant contributions of the residential sources to $PM_{2.5}$ concentrations in winter, especially during the heating seasons, have been well documented in China (Archer-Nicholls et al., 2016; Liu et al., 2016; Wu et al., 2018; Zhang et al., 2019c). For example, Liu et al. (2016) proposed that without residential emissions, $PM_{2.5}$ concentrations would decrease by 28±19 μg $m^{-3}$ and 44±27 μg $m^{-3}$ in Beijing and Tianjin, respectively. Therefore, implementing emission reductions for residential sources was an effective strategy to decrease $PM_{2.5}$ concentrations. In the BTH2+26 cities, the residential emission reductions were majorly achieved through promoting clean fuels in the residential sector, i.e., replacing coal with natural gas and electricity. Meng et al. (2019) evaluated that if 60% or 100% of household solid fuels were replaced by clean fuels, the population-weighted $PM_{2.5}$ concentrations in the BTH region would drop by 62 μg $m^{-3}$ or 79 μg $m^{-3}$ from 2014 to 2021. Zhao et al. (2018) projected that the residential energy transition from coal to clean fuels would prevent 0.51 million premature deaths in China. The remarkable decrease of $PM_{2.5}$ concentrations contributed by residential sources implied the great potential of residential emission reductions to air quality improvement.

The contributions of local, within-BTH, outside-BTH sources and BCON to the $PM_{2.5}$ average concentrations in the BTH2+26 cities in HS1718 relative to HS1617 are presented in Fig. 8b. In the current study, local contributions referred to the contributions of pollutants in the city itself while the within-BTH contributions represented the contributions from other cities in the BTH2+26 cities to $PM_{2.5}$. The outside-BTH contributions expressed contributions from other regions inside the simulation domain except the BTH2+26 cities and the BCON contributions referred to $PM_{2.5}$ contributions of boundary conditions. The local, within-BTH, outside-BTH contributions as well as the total contributions in $PM_{2.5}$ concentrations in HS1617 relative to those in HS1718 are summarized in Table 1. Local, within-BTH, outside-BTH and BCON sources contributed 10.9-58.9%, 10.3-65.2%, 9.3-49.5% and 4.7-16.0%, respectively, to the $PM_{2.5}$ concentrations in the BTH2+26 cities in HS1617, and 11.3-60.9%, 10.3-58.7%, 12.2-42.1% and 6.9-14.4% in HS1718, respectively. Fig. 8b shows that $PM_{2.5}$ concentrations contributed by the BCON sources changed little in the BTH2+26 cities between HS1617 and HS1718. The BCON contributions were not a major source to the $PM_{2.5}$ concentrations. The $PM_{2.5}$ concentrations contributed by local sources decreased from 10.2-81.8 μg $m^{-3}$ in HS1617 to 7.8-39.2 μg $m^{-3}$ in HS1718 in the BTH2+26 cities. The local sources dominated the $PM_{2.5}$ concentrations in Beijing, Baoding, Changzhi, Shijiazhuang, Tangshan, Taiyuan and Xingtai, with the contributions higher than 40%. The high local contributions during the heating seasons in the BTH2+26 cities estimated from this study are consistent with previous studies (Li et al., 2019b; Zhang et al., 2019a). The high local contributions in Tangshan and Taiyuan were due to their intensively distributed industries such as steel mills, building material factories and chemical plants. In Baoding, Beijing and Shijiazhuang, the large local contributions were resulted from the abundant use of bulk coal during the heating seasons. The contributions of local emission reductions to $PM_{2.5}$ were estimated to be 19.6, 11.5, 47.2, 3.4, 13.3, and 4.6 μg $m^{-3}$ for Beijing, Tianjin, Shijiazhuang, Jinan, Taiyuan, and Zhengzhou, respectively. The substantial decreases of $PM_{2.5}$ concentrations in these cities confirmed the effectiveness of air pollution control policies.

The within-BTH contributions to $PM_{2.5}$ concentrations in the BTH2+16 cities were in the range of 9.1-55.3 μg $m^{-3}$ for HS1617 and 6.6-41.3 μg $m^{-3}$ for HS1718. As shown in Fig. 8(b), the within-BTH sources were important contributors to $PM_{2.5}$ abatements in the BTH2+26 cities and varied greatly with cities. For example, for Langfang, the within-BTH sources contributed to $PM_{2.5}$ concentrations by 56.0 μg $m^{-3}$ in HS1617 and 33.9 μg $m^{-3}$ in HS1718 with percentages of 65.2% and 56.9%, respectively. Langfang was largely influenced by the within-BTH sources mainly due to its geographic position. Langfang is located in the downwind of Beijing. When north winds blow, pollutants emitted from Beijing affect Langfang. Therefore, Langfang benefited from the emission reductions in the within-BTH sources, particularly in Beijing. The within-BTH sources also had great effects on other cities such as Handan, Xingtai, Shijiazhuang and Hebi with the contributions of 22.1, 16.1, 15.9, 15.2 and 14.9 μg $m^{-3}$ to the $PM_{2.5}$ concentration decreases, respectively. This suggests the importance of inter-city transport of pollutants to the target cities. Chang et al. (2018) revealed that under stable atmospheric conditions in winter, $PM_{2.5}$ could be transported from southwest to northeast over a long distance at an height of 300-1000 meters above the ground in the BTH region. Chen et al. (2019a) analyzed the effects of different emission control measures on $PM_{2.5}$ decreases in Beijing by comparing the variations of $PM_{2.5}$ concentrations during four heavy haze episodes. Their comparisons showed that the unified emission control strategies carried out in the BTH2+26 cities contributed 16.4% to the average $PM_{2.5}$ concentration decreases in Beijing. The local emission-reduction measures accounted for 32.7% of the total $PM_{2.5}$ decreases in Beijing (Chen et al., 2019a). Using the WRF-CMAQ model simulations, Chen et al. (2019b) found that the controls of local and regional anthropogenic emissions contributed 53.7% and 24.9%, respectively, to the $PM_{2.5}$ decrease in Beijing from 2013 to 2017. Ji et al. (2019) also confirmed the important contributions of local emissions and regional transports to carbonaceous matter in Beijing from 2013 to 2017 through performing function analyses for potential contribution sources. Therefore, to further reduce the $PM_{2.5}$ concentrations in the BTH2+26 cities, emission reduction measures should be simultaneously carried out in all the major 28 cities, especially during periods of serious pollution.

In addition to the within-BTH sources, the outside-BTH sources were also important contributors to the $PM_{2.5}$ concentrations in the BTH2+26 cities. The outside-BTH contributions ranged from 10.5 to 36.8 μg $m^{-3}$ in HS1617 and 7.5 to 30.6 μg $m^{-3}$ in HS1718. The average $PM_{2.5}$ concentrations attributed to the outside-BTH sources decreased by 2.6-7.3 μg $m^{-3}$ in the BTH2+26 cities in HS1718 relative to HS1617. The decreases of the average $PM_{2.5}$ concentrations caused by the outside-BTH sources in the border cities such as Zhengzhou, Kaifeng, Jining, Taiyuan were higher than those in inner cities.

The considerable contributions of regional transports to target cities have been massively studied. For example, Chen et al. (2019a) reported that regional transports contributed more than 50% to the $PM_{2.5}$ concentrations in Beijing during one pollution episode in November 2017. Hua et al. (2016) and Jiang et al. (2015) confirmed the significant contribution of regional transports from the southern neighboring region of Beijing to the high $PM_{2.5}$ pollution concentrations in Beijing, especially for heavy $PM_{2.5}$ pollution events. Li et al. (2019a) combined the field observations with the CMAQ simulations and revealed that the regional transports from the surrounding regions contributed 31.4 μg $m^{-3}$ (34.4%) and 19.0 μg $m^{-3}$ (36.4%) to the $PM_{2.5}$ concentrations in Beijing in the winters of 2014 and 2017, respectively. Regional transports of pollutants to target cities were mostly affected by prevailing wind directions. Tong et al. (2019) analyzed the effects of different energy control strategies on the $PM_{2.5}$ abatements in the BTH region, and illustrated that only by combining emission reductions with stringent energy and industrial structure adjustments could the $PM_{2.5}$ concentrations be reduced to below 35 μg $m^{-3}$ in 2030. Although notable city-level $PM_{2.5}$ abatements were observed, $PM_{2.5}$ concentrations still remained at quite high concentration levels and the frequent and persistent heavy haze episodes still occurred in the BTH2+26 cities recently. Therefore, more emission-reduction efforts are needed for continuous improvement of air quality in the BTH2+26 cities. In addition, the implementations of united and systematic emission-reduction measures are indispensable.

Among various source apportionment methods including emission reduction methods, mass-transfer methods and incremental methods (Thunis et al., 2019), the source contribution approach based on emission sources has been implemented in air quality models to determine the relationships between emission sources and air pollution levels. Model sensitivity approaches including Brute-Force Method (BFM) and Decoupled Direct Method (DDM), and tagged species source apportionment including Particulate Source Apportionment Technology (PAST) and ISAM are the two most frequently used source-oriented approaches. Compared to BFM, DDM and PAST, ISAM is an efficient and robust tool to track the contributions from multiple sectoral emissions, regional emissions and boundary conditions to primary and secondary inorganic $PM_{2.5}$ species. However, due to the complexity of the chemical mechanisms of the $PM_{2.5}$ formations and the lack of considerations of indirect effects of the interactions among inorganic $PM_{2.5}$ species, ISAM has limitations in the source apportionment of nonlinear species (Kwok et al., 2013; Thunis et al., 2019). To better understand the impacts of emissions on air quality and make scientific emission control policies, further model development is required.

# 4. Conclusion

The substantial decreases of the average $PM_{2.5}$ concentrations and the numbers of heavy haze days in the BTH2+26 cities in HS1718 (52. 9-101. 9 μg $m^{-3}$, 5-30 days) relative to HS1617 (77.3-152.5 μg $m^{-3}$, 17-77 days) were identified by both observations and simulations. Model simulations suggested the important role of both favorable meteorological conditions (1.9-25.4 μg m$^{-3}$, 18.4-97.7%) and emission-reduction measures (0.4-55.0 μg m$^{-3}$, 2.3-81.6%) in the air quality improvement in the BTH2+26 cities. The ISAM source attributions showed that residential sources dominated the $PM_{2.5}$ changes in the BTH2+26 cities and contributed more than 30 μg m$^{-3}$ to $PM_{2.5}$ decreases in Beijing, Baoding, Hengshui, Xingtai, Shijiazhuang and Handan. The local and within-BTH sources were important contributors to the $PM_{2.5}$ concentrations in the BTH2+26 cities. Emission reductions associated with the local and within-BTH sources effectively decreased the average $PM_{2.5}$ concentrations by 0.1-47.2 μg m$^{-3}$ and 0.3-22.1 μg m$^{-3}$ in HS1718 relative to HS1617, respectively.

## Credit authorship contribution statement

S.Y., and Y.Z. conceived and designed the research. Y.Z performed model simulations and the data analyses. X.C., M.L., L.W., P.L. Z.L., W.L., and D.R. contributed to the scientific discussions. S.Y., Y.Z. and J.H.S wrote the paper.

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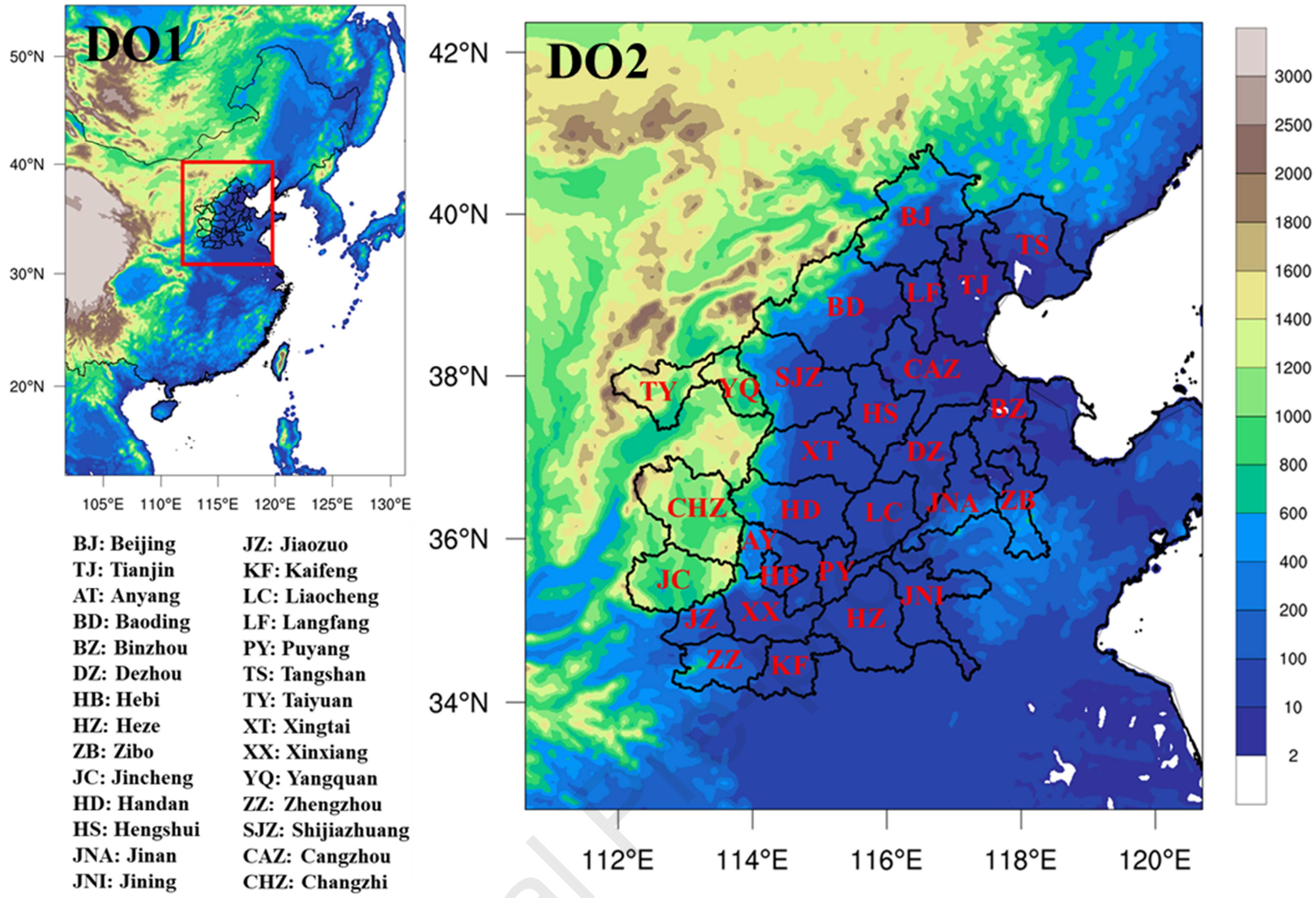

Fig. 1. Map of simulation domains (D01 and D02) and location of each city for the BTH2+26 cities.

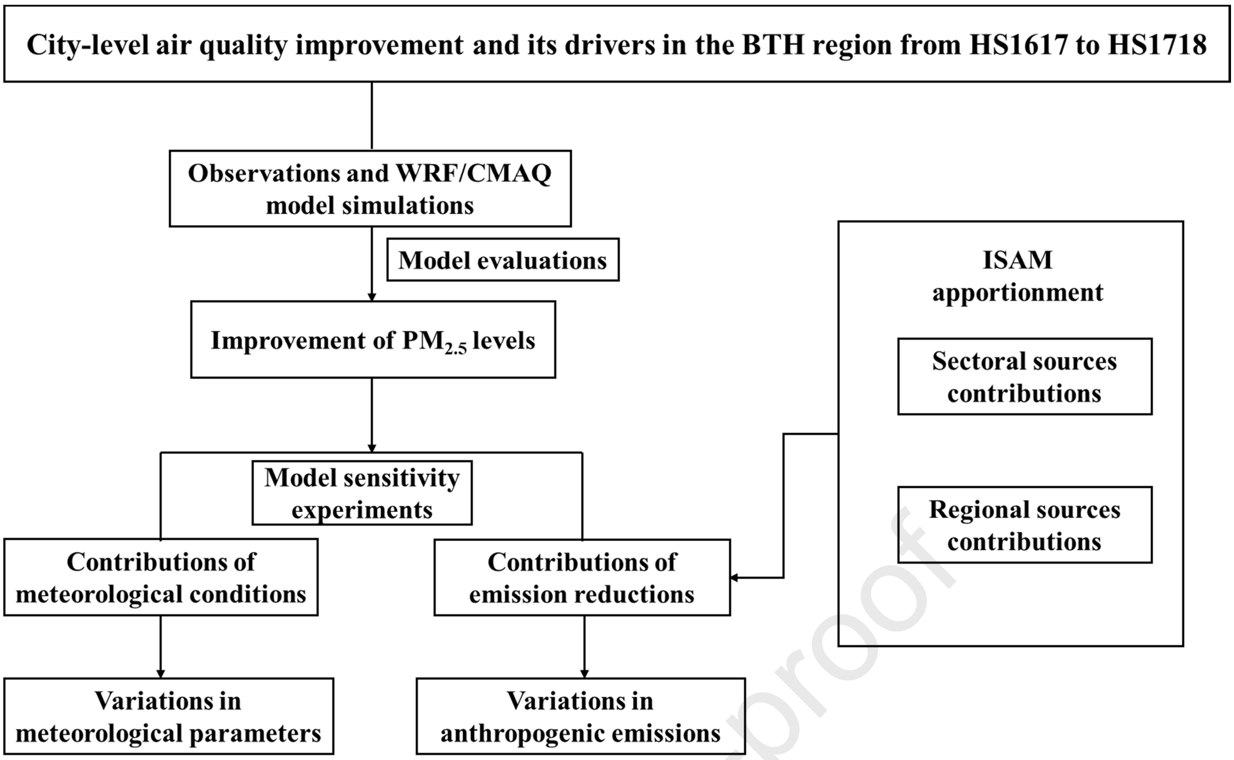

Fig.2. The technical flowchart for this study.

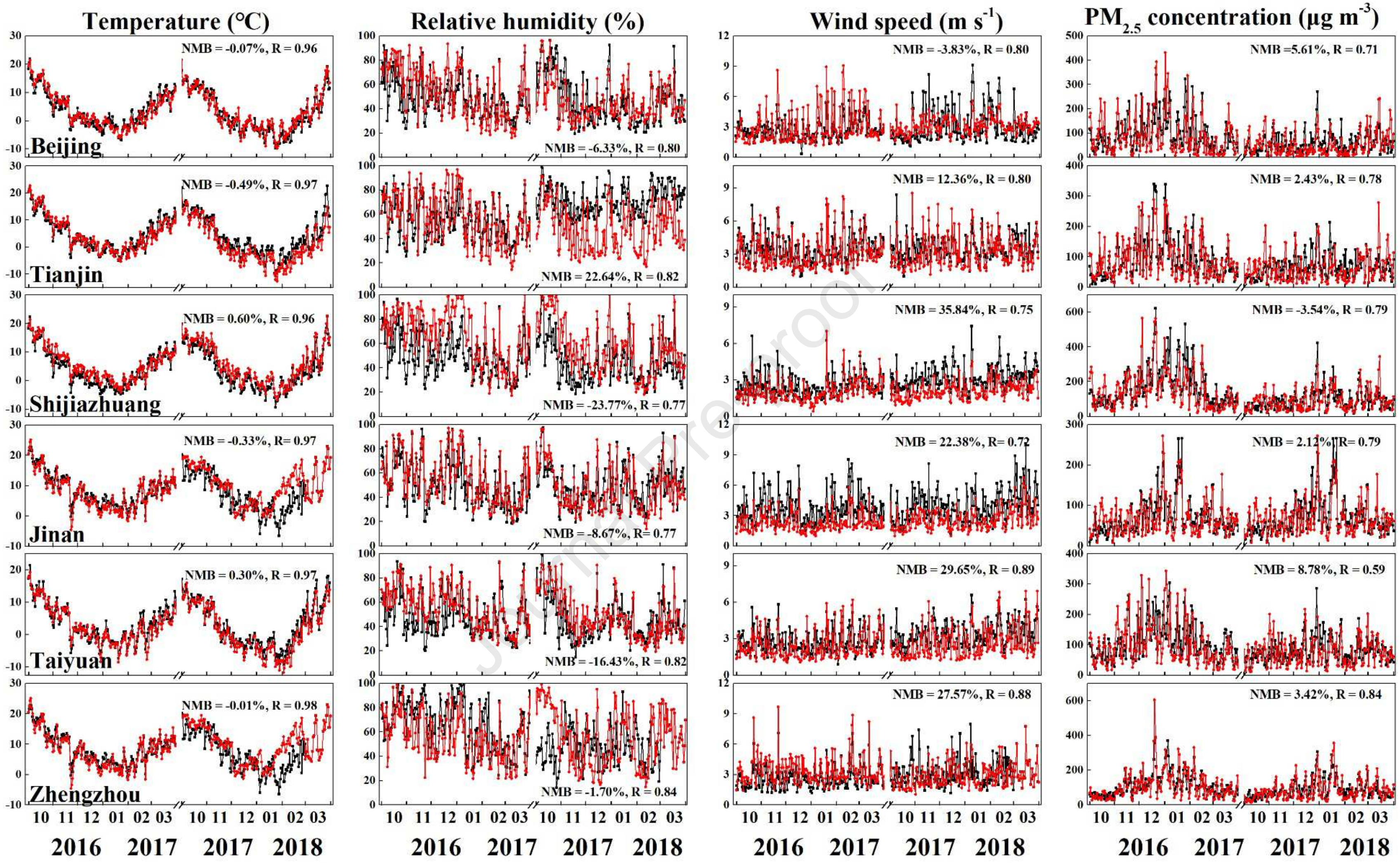
Temperature (°C)
Relative humidity (%)
Wind speed ($m\ s^{-1}$)
$PM_{2.5}$ concentration ($\mu g\ m^{-3}$)
Beijing
NMB = -0.07%, R = 0.96
NMB = -6.33%, R = 0.80
NMB = -3.83%, R = 0.80
NMB =5.61%, R = 0.71
Tianjin
NMB = -0.49%, R = 0.97
NMB = 22.64%, R = 0.82
NMB = 12.36%, R = 0.80
NMB = 2.43%, R = 0.78
Shijiazhuang
NMB = 0.60%, R = 0.96
NMB = -23.77%, R = 0.77
NMB = 35.84%, R = 0.75
NMB = -3.54%, R = 0.79
Jinan
NMB = -0.33%, R= 0.97
NMB = -8.67%, R= 0.77
NMB = 22.38%, R= 0.72
NMB = 2.12%, R= 0.79
Taiyuan
NMB = 0.30%, R = 0.97
NMB = -16.43%, R = 0.82
NMB = 29.65%, R = 0.89
NMB = 8.78%, R = 0.59
Zhengzhou
NMB = -0.01%, R = 0.98
NMB = -1.70%, R = 0.84
NMB = 27.57%, R = 0.88
NMB = 3.42%, R = 0.84
30
20
10
0
-10
100
80
60
40
20
0
12
9
6
3
0
500
400
300
200
100
600
10 11 12 01 02 03 10 11 12 01 02 03
2016 2017 2017 2018

Fig. 3. Time series of daily mean observed and simulated meteorological factors (temperature, relative humidity and wind speed) and $PM_{2.5}$ concentrations in six major cities (Beijing, Tianjin, Jinan, Shijiazhuang, Taiyuan and Zhengzhou) in HS1617 and HS1718. The NMB, and R values are also shown inside the figures.

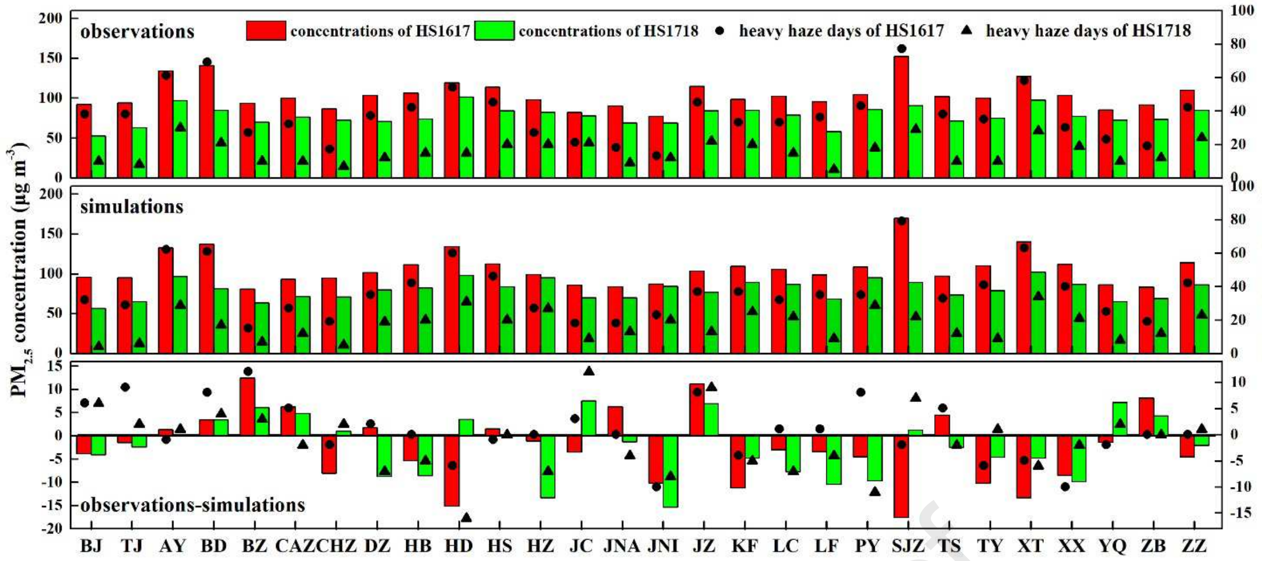

Fig. 4. Observed and simulated average $PM_{2.5}$ concentrations and the numbers of heavy haze (daily $PM_{2.5}$ ≥150 μg $m^{-3}$) days in the BTH2+26 cities in HS1617 and HS1718. BJ: Beijing; TJ: Tianjin; AY: Anyang; BD: Baoding; BZ: Binzhou; CAZ: Cangzhou; CHZ: Chizhi; DZ: Dezhou; HB: Hebi; HD: Handan; HS: Hengshui; HZ: Heze; JC: Jincheng; JNA: Jinan; JNI: Jining; JZ: Jiaozuo; KF: Kaifeng; LC: Liaocheng; LF: Langfang; PY: Puyang; SJZ: Shijiazhuang; TS: Tangshang; TY: Taiyuan; XT: Xingtai; XX: Xinxiang; YQ: Yangquan; ZB: Zibo; ZZ: Zhengzhou.

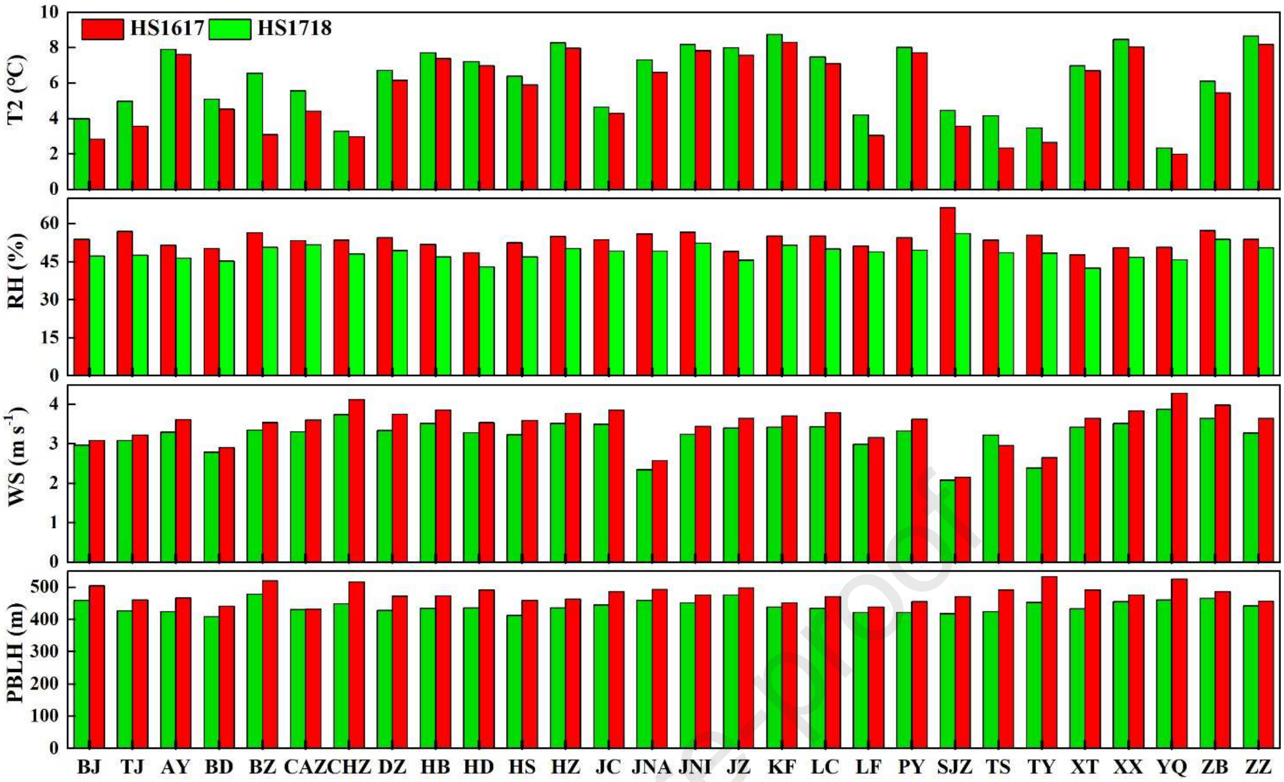

Fig. 5. Simulated mean values of meteorological parameters (T2, RH, WS and PBLH) in the BTH2+26 cities in HS1617 and HS1718.

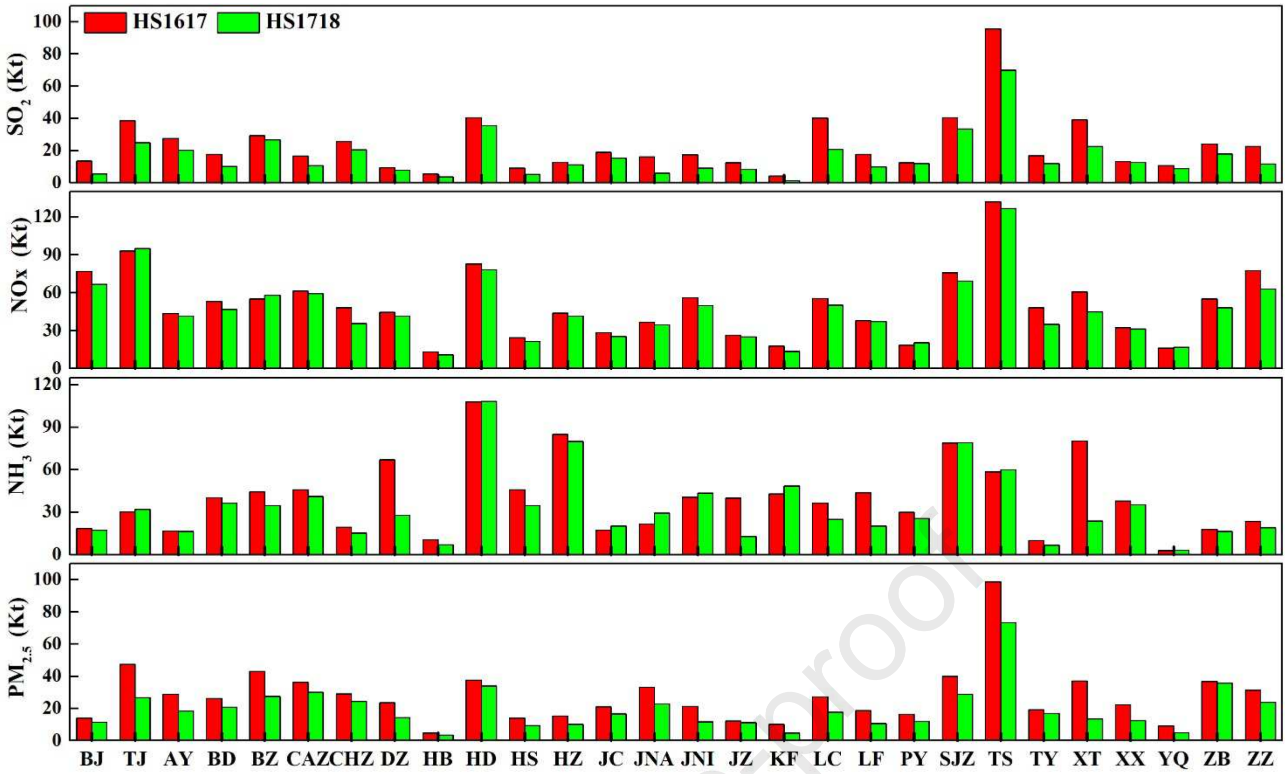

Fig. 6. Anthropogenic emissions of $SO_2$, $NO_x$, $NH_3$ and primary $PM_{2.5}$ in the BTH2+26 cities in HS1617 and HS1718.

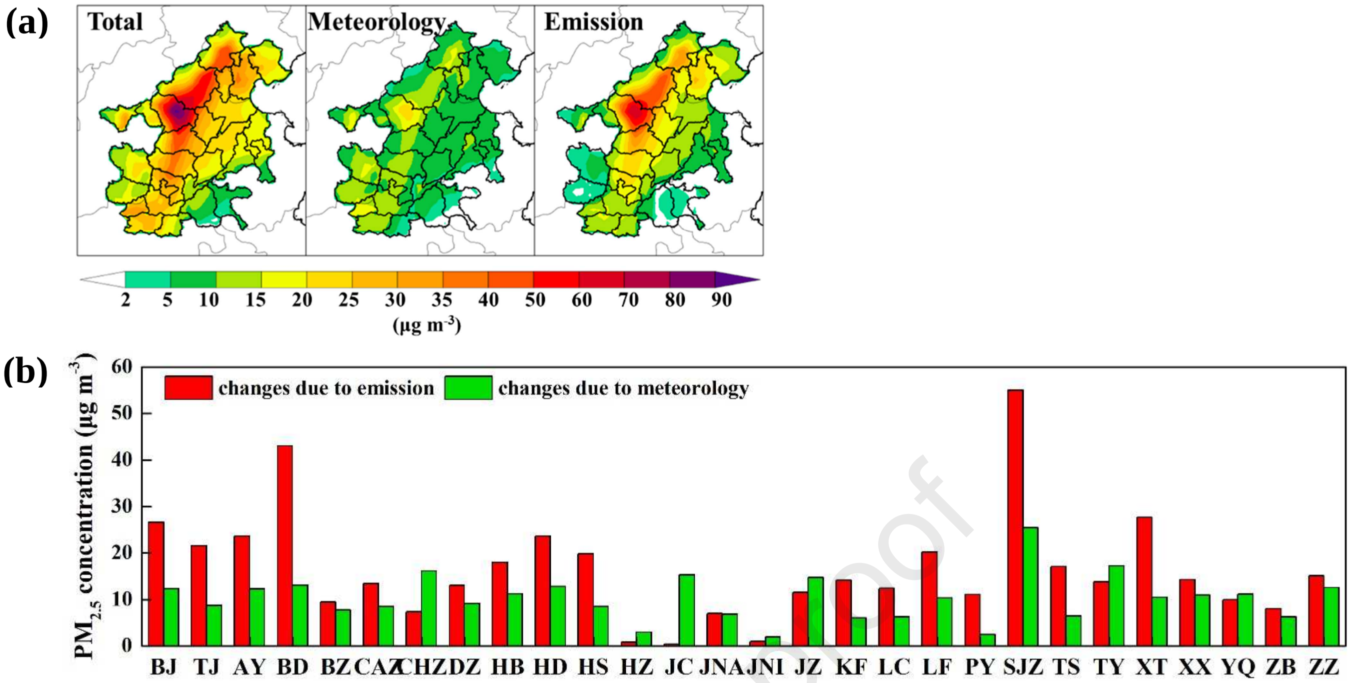

Fig. 7. (a) Spatial distributions of the total changes and the changes of $PM_{2.5}$ concentrations attributed to meteorology conditions and emission reductions over the BTH area from HS1617 to HS1718; (b) Predicted changes of $PM_{2.5}$ concentrations in each city attributed to meteorology and emissions in the BTH2+26 cities from HS1617 to HS1718.

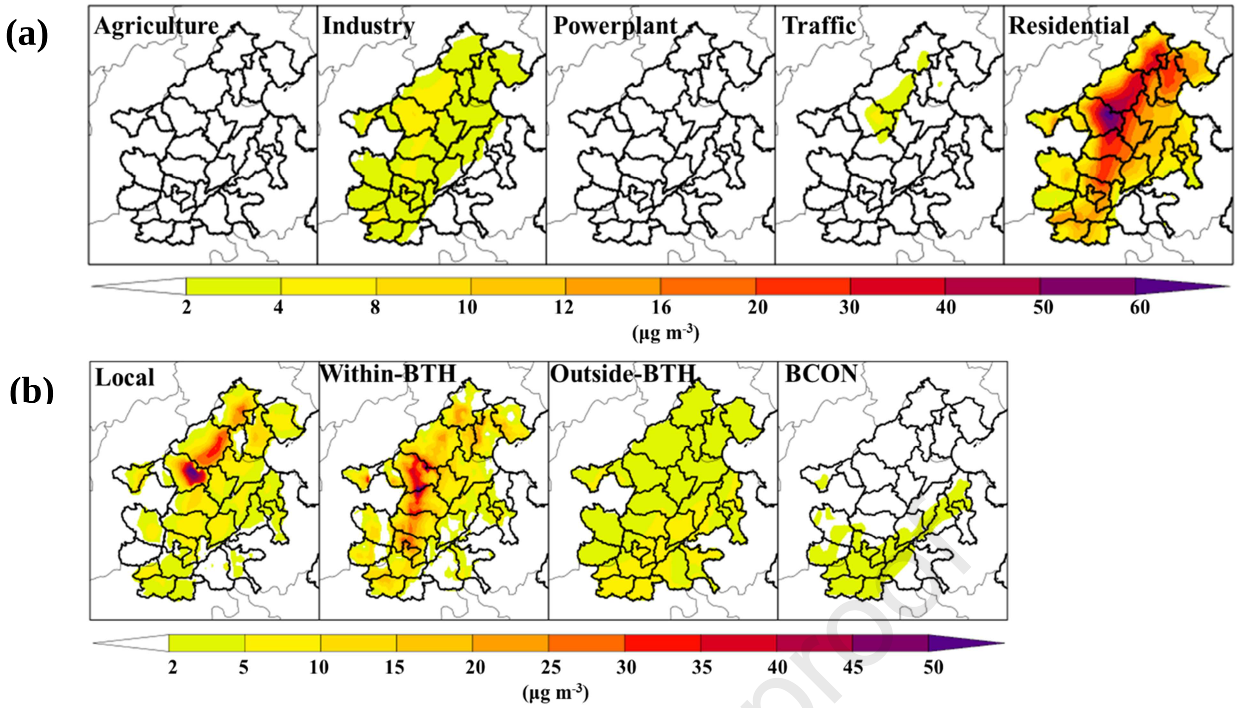

Fig. 8. Spatial distributions of the differences in contributions of the different sectoral emissions (agriculture, industry, power plant, traffic and residential) (a) and regional sources (local, within-BTH, outside-BTH and BCON) (b) to the $PM_{2.5}$ concentration decreases from HS1617 to HS1718 over the BTH area.

Table 1. Contributions of local, within-BTH and outside-BTH sources to $PM_{2.5}$ concentrations in the BTH2+26 cities in HS1617 and HS1718 and their differences.

| | HS1617 ($\mu g\ m^{-3}$) | | | HS1718 ($\mu g\ m^{-3}$) | | | HS1617-HS1718 ($\mu g\ m^{-3}$) | | |
|---|---|---|---|---|---|---|---|---|---|
| | local | within-BTH | outside-BTH | local | within-BTH | outside-BTH | local | within-BTH | outside-BTH |
| BJ | 43.6 | 23.1 | 10.5 | 23.9 | 13.8 | 7.5 | 19.6 | 9.3 | 3.0 |
| TJ | 30.2 | 30.3 | 13.2 | 18.7 | 21.6 | 9.5 | 11.5 | 8.7 | 3.7 |
| AY | 32.3 | 53.9 | 17.8 | 22.1 | 40.1 | 12.6 | 10.2 | 13.8 | 5.2 |
| BD | 65.3 | 33.2 | 11.8 | 35.7 | 19.6 | 8.6 | 29.6 | 13.7 | 3.2 |
| BZ | 14.9 | 22.6 | 22.7 | 11.6 | 19.4 | 16.6 | 3.3 | 3.2 | 6.1 |
| CAZ | 25.9 | 31.3 | 14.8 | 20.4 | 23.6 | 10.7 | 5.4 | 7.7 | 4.0 |
| CHZ | 36.1 | 19.2 | 14.9 | 26.8 | 14.6 | 11.3 | 9.3 | 4.6 | 3.6 |
| DZ | 21.0 | 41.6 | 17.5 | 15.2 | 34.0 | 12.9 | 5.8 | 7.6 | 4.6 |
| HB | 10.2 | 55.3 | 18.4 | 7.8 | 40.4 | 13.0 | 2.4 | 14.9 | 5.4 |
| HD | 38.9 | 51.1 | 16.1 | 29.7 | 35.0 | 11.7 | 9.2 | 16.1 | 4.4 |
| HS | 24.7 | 49.5 | 15.4 | 18.2 | 36.6 | 11.3 | 6.5 | 12.9 | 4.1 |
| HZ | 23.6 | 28.8 | 25.1 | 23.3 | 24.8 | 24.3 | 0.3 | 0.8 | 4.0 |
| JC | 18.1 | 17.6 | 23.3 | 17.3 | 13.9 | 17.1 | 0.7 | 6.2 | 4.6 |
| JNA | 15.8 | 20.5 | 22.9 | 12.4 | 19.3 | 18.4 | 3.4 | 1.2 | 4.6 |
| JNI | 13.0 | 15.9 | 37.8 | 11.8 | 15.6 | 30.6 | 1.2 | 0.3 | 7.2 |
| JZ | 21.6 | 31.6 | 19.4 | 16.3 | 23.9 | 14.4 | 5.3 | 7.6 | 5.0 |
| KF | 16.4 | 32.4 | 31.7 | 12.6 | 28.5 | 24.3 | 3.8 | 3.8 | 7.3 |
| LC | 25.0 | 32.2 | 23.4 | 19.3 | 29.4 | 17.7 | 5.7 | 2.8 | 5.7 |
| LF | 12.9 | 56.0 | 11.1 | 12.8 | 33.9 | 8.5 | 0.1 | 22.1 | 2.6 |
| PY | 16.2 | 43.5 | 23.0 | 15.2 | 41.3 | 17.2 | 1.0 | 2.3 | 5.8 |
| SJZ | 81.8 | 40.3 | 13.2 | 34.6 | 25.1 | 9.3 | 47.2 | 15.2 | 3.8 |
| TS | 36.5 | 22.5 | 13.2 | 29.1 | 14.7 | 10.2 | 7.3 | 7.8 | 3.0 |
| TY | 52.1 | 9.1 | 20.4 | 38.8 | 6.6 | 13.5 | 13.3 | 2.6 | 6.9 |
| XT | 50.5 | 46.1 | 14.3 | 39.2 | 30.2 | 10.5 | 11.3 | 15.9 | 3.8 |
| XX | 19.8 | 40.5 | 21.3 | 15.9 | 32.0 | 15.5 | 3.9 | 8.5 | 5.9 |

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| YQ | 22.1 | 21.6 | 18.1 | 18.6 | 14.9 | 13.1 | 3.4 | 6.7 | 5.0 |
| ZB | 18.5 | 17.6 | 23.9 | 14.0 | 16.6 | 19.3 | 4.6 | 1.0 | 4.5 |
| ZZ | 20.5 | 34.7 | 27.0 | 15.9 | 26.3 | 19.8 | 4.6 | 8.4 | 7.2 |

# City-level Air quality improvement in the Beijing-Tianjin-Hebei region from 2016/17 to 2017/18 heating seasons: attributions and process analysis

## Highlights

1) $PM_{2.5}$ pollution abatements were witnessed by observations and simulations.
2) Both emission reductions and favorable meteorological conditions help to reduce $PM_{2.5}$ concentrations.
3) Residential sources dominated the $PM_{2.5}$ decreases in the Beijing-Tian-Hebei (BTH) region.
4) Local and within-BTH sources emission reduction controls contributed the most to $PM_{2.5}$ concentration decreases.

# Inorganic water-soluble ions of $PM_{2.5}$ in metropolitan Hangzhou, China: Characteristics, sources, and process analysis

Conflict of interests

Authors declare no conflict of interests.