Quantifying the contributions of atmospheric processes and meteorology to severe $PM_{2.5}$ pollution episodes during the COVID-19 lockdown in the Beijing-Tianjin-Hebei, China

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Key Points:

- Three severe $PM_{2.5}$ pollution episodes were identified in the Beijing-Tianjin-Hebei region during the COVID-19 lockdown.
- The $PM_{2.5}$ episodes were dominated by emissions and aerosol processes, and enhanced by unfavorable meteorological conditions.
- Designing more effective emissions control strategies with both chemistry and meteorology in thought could mitigate future $PM_{2.5}$ episodes.

Abstract

A major tool for curtailing the spread of COVID-19 pandemic in China was a nationwide lockdown, which led to significant reductions in anthropogenic emissions and fine particulate matter ($PM_{2.5}$). However, the lockdown measures did not prevent high $PM_{2.5}$ pollution episodes (EPs). Three severe EPs were identified in the Beijing-Tianjin-Hebei (BTH) region during the lockdown. The integrated process rate (IPR) analysis tool in the Community Multiscale Air Quality (CMAQ) model was employed to quantify the contributions of individual atmospheric processes to $PM_{2.5}$ formation during the lockdown in the BTH region. The IPR results showed that emissions and aerosol processes were the dominant sources of net surface $PM_{2.5}$ in Beijing and Tianjin, constituting a total of 86.2% and 92.9%, respectively, while emissions, horizontal transport, and aerosol processes dominated the net surface $PM_{2.5}$ in Shijiazhuang and Baoding. In addition, the EPs in Beijing and Tianjin were primarily driven by local emissions, while the EPs in Shijiazhuang and Baoding were attributed to combined local emissions and regional transport. The reductions in $PM_{2.5}$ in Case 2 relative to Case 1 were attributed to the weaker $PM_{2.5}$ formation from emissions and aerosol processes. However, the EPs were enhanced by low planetary boundary layer heights, low vertical export of $PM_{2.5}$ from the boundary layer to the free troposphere, and substantial horizontal import, especially in Shijiazhuang and Baoding. This study improves the understanding of buildup of $PM_{2.5}$ during the EPs, and the results provide insights for designing more effective emissions control strategies to mitigate future $PM_{2.5}$ episodes.

Keywords: Fine particulate matter; Pollution episodes; Process analysis; WRF-CMAQ; COVID-19 shutdown.

1. Introduction

For more than two decades, China has been suffering from severe haze pollution, attributed to its population growth, urbanization, fast industrialization, as well as economic advancement (Li et al., 2020; Shi et al., 2017; Zhao et al., 2021). The development of severe haze is caused by a combination of anthropogenic emissions (local and regional) (Sulaymon et al., 2020, 2021a) and adverse meteorological conditions (Chen et al., 2021; Hua et al., 2021; Hu et al., 2016; Shen, et al., 2021; Shi et al., 2020; Sulaymon et al., 2021a, 2021b). Severe air pollution causes reductions in visibility (Jiang et al., 2021; Li et al., 2019; Wang et al., 2018), changes in climate and ecosystem services (Jiang et al., 2021; Zhao et al., 2021), and adverse human health effects (Chen et al., 2017; Croft et al., 2019; Hopke et al., 2019; Shang et al., 2018; Shen et al., 2020; Yan et al., 2018). The global disease burden (GDB) has attributed about 2 million premature deaths per annum to severe air pollution exposure in China (Yin et al., 2020).

The issuance of the new ambient air quality standards (GB3095-2012) in 2012 and the subsequent implementation of the Air Pollution Prevention and Control Action Plan (APPCAP) in September 2013 by the Chinese authorities has led to reduction in the concentrations of fine particulate matter with aerodynamic diameters of ≤2.5 ($PM_{2.5}$) in Chinese cities (Fan et al., 2020; Sulaymon et al., 2021d; Wang et al., 2016). For instance, Xue et al. (2019) noted about 32.5% reduction in the national population-weighed $PM_{2.5}$ annual mean between 2013 and 2017. However, high $PM_{2.5}$ concentrations are still observed in most cities, with annual averages violating the annual Grade I (15 μg/m^3) and Grade II (35 μg/m^3) Chinese Ambient Air Quality Standards (CAAQS), and much higher than the WHO (5 μg/m^3) recommended limit or the USEPA (12 μg/m^3) standard.

The Beijing-Tianjin-Hebei (BTH) region that includes Beijing and Tianjin, and Hebei Province, is one of the most economically developed regions in China. The region has been suffering from severe $PM_{2.5}$ pollution over the past two decades (Chang et al., 2018; Dai et al., 2021a), particularly during the winter season. During past international events (e.g. 2008 Olympic Games, 2014 Asia-Pacific Economic Cooperation, and the 2015 Military Parade), Chinese authorities implemented major emissions reductions measures in the BTH region to improve air quality. The effectiveness and success of the emissions reduction policies have been assessed (Wang et al., 2016; Xu et al., 2017; Yang et al., 2016).

In December 2019, an outbreak of coronavirus disease (COVID-19) occurred in Wuhan (Zhu et al., 2020) and spread across China and many other countries within a short time. As one of the measures to curtail the spread of COVID-19 pandemic in China, a nationwide lockdown was implemented by the Chinese authorities, leading to significant reductions in anthropogenic emissions and $PM_{2.5}$ concentrations across China (Sulaymon et al., 2021a, 2021c; Wang et al., 2020; Zhao et al., 2020; Zhao et al., 2021). However, the BTH region still experienced high $PM_{2.5}$ pollution episodes during the lockdown (Sulaymon, et al., 2021a; Zhang et al., 2021). Compared to the past international events held in Beijing during the summer and autumn seasons (with no or few pollution episodes), the COVID-19 pandemic occurred in winter, a period with frequent severe pollution events especially in the BTH region. In addition, the COVID-19 lockdown had a longer period with very strict measures than the duration of the past three events.

Previous studies have assessed the impacts of COVID-19 lockdown on air quality as well as the relationships between air quality and meteorological conditions during lockdown in BTH region (Cui et al., 2020; Dai et al., 2020; 2021b; Sulaymon et al., 2021a;

Zhang et al., 2021; Zhao et al., 2021), other regions in China (Gao et al., 2021; Liu et al., 2020; Shen et al., 2021a, 2021b; Sulaymon, et al., 2021c; Wang et al., 2020; Wu et al., 2021; Xing et al., 2020), and outside mainland China (Bashir et al., 2020; Chauhan and Singh, 2020, 2021; Mishra et al., 2021; Muhammad et al., 2020; Orak and Ozdemir, 2021; Querol et al., 2021; Sharma et al., 2020; Singh and Chauhan, 2020; Srivastava, 2021; Ye et al., 2022). A few studies have also been performed on the regional source apportionment of $PM_{2.5}$ during the lockdown (Li et al., 2020; Ma et al., 2021). Li et al. (2020) reported that industry (32.2-61.1%) and residential (2.1-28.5%) were the two highest sources contributing to $PM_{2.5}$ in the Yangtze River Delta (YRD) region, while about 14.0-28.6% contribution was due to long-range transport from northern China. In the BTH region, a few studies have also investigated the source apportionment of $PM_{2.5}$ during lockdown (Cui et al., 2020; Dai et al., 2020). For example, Dai et al. (2020) used Positive Matrix Factorization (PMF) to investigate the sources of $PM_{2.5}$ in Tianjin. Their results showed that secondary inorganic aerosols (SIA) (50.5%), fireworks and residential burning (32.0%), and primary coal combustion emissions (13.3%) were the three dominant sources contributing to $PM_{2.5}$ during the lockdown. Overall, previous studies have reported persistent haze episodes in the BTH region during lockdown despite the emission reductions, and have generally attributed them to unfavorable meteorological conditions (Cui et al., 2020; Dai et al., 2020; 2021b; Sulaymon et al., 2021a; Zhang et al., 2021; Zhao et al., 2021). However, the formation of air pollutants involves various physical processes (such as emissions, condensation, advection, diffusion, deposition, etc.) as well as oxidative chemical process (Huang et al., 2005; Wang et al., 2014; Ye et al., 2022).

The process analysis (PA) tool in the Community Multiscale Air Quality (CMAQ) chemical transport model can provide quantitative analysis of the individual contributions of various physical and chemical processes to the observed air pollution (Liu et al., 2010; Liu and Zhang, 2011; Xing et al., 2011; Ye et al., 2022). Liu and Zhang (2011) employed the PA tool to analyze a regional $PM_{2.5}$ pollution episode in the U.S. They found that emissions and aerosol processes such as homogeneous nucleation, heterogeneous nucleation, and condensation were the dominant contributors to increased $PM_{2.5}$ concentrations, while horizontal and vertical transport and dry deposition were the primary loss mechanisms. Liu et al. (2010) utilized the PA to explore the contributions of various atmospheric processes on ozone and PM_{10} concentrations in China during four seasons. The results showed that emissions and aerosol processes were the main contributors to PM_{10} concentrations, while horizontal transport was the major removal pathway. Xing et al. (2011) used CMAQ-PA tool to quantify the air quality benefits from emissions reductions and meteorological variations during the 2008 Beijing Olympics. The results indicated that aerosol and emission processes acted as the major $PM_{2.5}$ pathways, while vertical transport was the major $PM_{2.5}$ sink at the surface.

Therefore, analyzing the air quality during the unique lockdown period to provide additional understanding of the underlying causes of high pollution episodes even during periods of substantially reduced anthropogenic activity is important for providing approaches to future air quality management strategies. The present study is the first that elucidated the contributions of various atmospheric processes to $PM_{2.5}$ pollution episodes during the lockdown in the BTH region. The results provide new insights into $PM_{2.5}$ formation of the three pollution episodes during lockdown. Thus, it provides a valuable

example of how to use opportunities like the lockdown period to better understand the causal factors of episodes in other areas of the world, which can then be applied to develop more effective control strategies that would reduce the magnitude of these episodes and better protect public health.

2. Methodology

2.1. Model set-up and configurations

The Community Multiscale Air Quality model version 5.2 (CMAQv5.2) was applied to simulate the air quality in the BTH region during the COVID-19 lockdown period (January 24-February 29, 2020). The photochemical mechanism and the aerosol module used in configuring the model were the State-wide Air Pollution Research Center version 07 (SAPRC07tic) and AERO6i, respectively (Liu et al., 2020; Sulaymon et al., 2021a, 2021b). Two nested domains with horizontal resolutions of 36 and 12 km were used (Fig. S1). The outer domain (36 km) covers China and the surrounding regions (137 x 107 grids), and the inner domain (12 km) covers the study area, the BTH region (127 x 202 grids). Each of the two domains had 18 vertical layers, emanating from the surface to a height of about 20 km above the ground level. The initial and boundary conditions (IC/BC) used in the 36 km domain were based on the default profiles provided by the CMAQ model, while the IC/BC used for the 12 km domain were generated from the results of the 36 km simulations. As a way of reducing the impact of initial conditions on $PM_{2.5}$ predictions, the simulations began on January 19, and the results of the first 5 days (January 19-23, 2020) were excluded from the model analysis, thus serving as a spin-up of the model. The meteorological inputs were simulated by the Weather Research and Forecasting (WRF v4.0) model with the FNL reanalysis data serving as the IC/BC. The detailed settings and

configurations, including the major physics schemes used in this study are listed in Table S1, while other settings could be found in previous studies where the WRF model was applied (Hu et al., 2015, 2016; Wang et al., 2021).

In this study, the Multi-resolution Emission Inventory for China (MEIC) of year 2016 (http://www.meicmodel.org) served as the anthropogenic emissions from China. In addition, the anthropogenic emissions from adjacent countries and regions were processed based on the Regional Emission inventory in ASia version 2 (REAS2) (Kurokawa et al., 2013). Biogenic emissions were estimated with the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1. Open burning emissions were generated based on the data obtained from the Fire INventory from NCAR (FINN) (Wiedinmyer et al., 2011). Sea salt and windblown emissions were generated inline (Sulaymon et al., 2021a, 2021b). Further details regarding the emission processing can be found in Hu et al. (2016) and Qiao et al. (2015).

To evaluate the impacts of the emissions reductions on air quality, two scenarios (referred to as Cases 1 and 2) were simulated as presented in Table S2. The first scenario (Case 1) used the original MEIC16 emission inventory. In the second scenario (Case 2), emissions from transportation, industry, and power sectors were reduced (Table S2) during the lockdown period, while those of residential and agriculture were similar to Case 1. The basis for adopting the emission reduction factors has been previously presented (Sulaymon et al., 2021a; Wang et al., 2020), and has also been detailed in the Supplementary Material (Text S1). The differences between the results of Cases 1 and 2 represent the impact of emissions reductions on air quality during the lockdown.

2.2. Process analysis

The process analysis (PA) tool embedded in the CMAQ model has been described as a versatile analytical tool for quantifying the contributions of individual atmospheric processes and chemical reactions to a pollutant (Fu et al., 2020; Ye et al., 2022). PA is comprised of two components; the integrated process rate (IPR) and integrated reaction rate (IRR) analysis. The IPR involves the changes in the hourly concentrations of pollutants due to individual atmospheric processes such as gas-phase chemistry, emissions, aerosol processes, dry deposition, cloud processes, and vertical and horizontal transport at each grid cell in the model domain. The IPR analysis has been extensively used in quantifying the contributions individual atmospheric processes to air pollutants (Fan et al., 2015; Fu et al., 2020; Li et al., 2012; Wang et al., 2010; Xing et al., 2011; Ye et al., 2022), hence, detailed information about IPR can be found in these referenced studies.

In this study, the IPR module in CMAQv5.2 was employed to resolve both physical and chemical processes involved in the formation of $PM_{2.5}$ during the lockdown period in the BTH region. The IPR results were subsequently used to analyze the individual processes involved in $PM_{2.5}$ formation in the surface layer and full planetary boundary layer (PBL), respectively. For this purpose, the processes considered were the chemistry (gas-phase), emissions, aerosol processes (SOA formation, nucleation, condensation, coagulation, heterogeneous chemistry, mode merging, and aerosol thermodynamics), cloud processes, dry deposition, vertical transport (sum of vertical advection and diffusion), and horizontal transport (sum of horizontal advection and diffusion). Based on their contributions to $PM_{2.5}$ concentrations, atmospheric processes can be grouped into two; source process (concentration increases) and sink process (concentration decreases). Dry

deposition and emission belong to the sink and source process, respectively. The IPR of other processes can either be source (positive) or sink (negative). The contributions of individual atmospheric processes to the formation of $PM_{2.5}$ were estimated using the approach of Ye et al. (2022):

$$SOURCE_p = \frac{\sum_t IPR_{p,t}}{\sum_p \sum_t IPR_{p,t}} \times 100\% \quad (IPR_{p,t} > 0) \tag{1}$$

$$SINK_p = \frac{\sum_t IPR_{p,t}}{\sum_p \sum_t IPR_{p,t}} \times 100\% \quad (IPR_{p,t} < 0) \tag{2}$$

where p is the atmospheric process, and t is the time (in hour). $SOURCE_p$ and $SINK_p$ are the proportions of the atmospheric process p in all source and sink processes, respectively. Both source and sink categories are used to reveal how important an atmospheric process is in influencing the changes in $PM_{2.5}$ concentrations.

3. Results and discussion

3.1. WRF model performance

Meteorological parameters play an important role in the formation and transportation of air pollution (Hu et al., 2016; Sulaymon et al., 2021a, 2021b; Wang et al., 2021). In addition, the influences of meteorological parameters on the air quality simulations using chemical transport model have also been established (Hu et al., 2016; Sulaymon et al., 2021a; Wang et al., 2021). To evaluate the WRF model performance, the predicted temperature (T2) and relative humidity (RH) at 2 m above ground level, and wind speeds (WS) and wind directions (WD) at 10 m above surface were compared to the observational data downloaded from the official website of the Chinese Meteorological Agency (http://data.cma.cn/en, last access: January 2023). Table S3 shows the summary

240 statistics including the mean observation (OBS), mean prediction (PRE), mean bias (MB),
241 mean error (ME), and the root mean square error (RMSE). In addition to the BTH region
242 as a whole, four representative cities including Beijing (BJ), Tianjin (TJ), Shijiazhuang
243 (SJZ), and Baoding (BD) were evaluated. Generally, T2 (Table S3) was slightly over-
244 predicted in the BTH and the four representative cities during the lockdown. The MB and
245 ME of T2 in BTH were 0.4 and 1.7, respectively, which fell below the suggested
246 benchmarks (MB≤±0.5; and ME≤2.0) (Emery et al., 2001). These are consistent with a
247 previous study over BTH region (Chang et al., 2019). Except in Tianjin (MB:0.5), the MB
248 values in other three cities (Beijing:2.2; Shijiazhuang:0.6; and Baoding:1.3) exceeded the
249 benchmark. Except in Beijing (ME:2.3), the ME values in all the cities were within the
250 benchmark range. Although there were no suggested benchmarks for the MB and ME
251 indices of RH, however, RH (Table S3) was underpredicted in BTH region and the four
252 representative cities (Ma et al., 2021). Similar results had been reported by previous studies
253 over BTH region (Chang et al., 2018; Li et al., 2021b; Sulaymon et al., 2021a; Zhao et al.,
254 2021) and China as a whole (Hu et al., 2016; Sulaymon et al., 2021b; Wang et al., 2021).
255 Bhati and Mohan (2018) obtained a similar result and attributed it to the influence of the
256 boundary layer parameterization on the weather prediction. The mean observed WS across
257 the cities and BTH ranged from 1.8 to 2.3 m/s, an illustration of relatively calm conditions
258 during the lockdown. Generally, WS (Table S3) was over-predicted (Ma et al., 2021).
259 However, based on the ME, MB, and RMSE indices, the predictions reasonably captured
260 the observations across the four cities and BTH (Li et al., 2021b; Sulaymon et al., 2021a;
261 Zhao et al., 2021). The over-predictions of WS might be due to unresolved topography
262 within the WRF model (Li et al., 2014). The MB values met the suggested benchmark

(≤±0.5) in BTH and three cities except Shijiazhuang (0.7). During the lockdown, the ME and RMSE values ranged between 0.6-0.9 and 0.7-1.2, respectively, with both indices falling below the recommended benchmarks (≤2.0). WD (Table S3) was generally under-predicted except in Shijiazhuang where the PRE was slightly higher than the OBS. Overall, the MB values were above the suggested criterion range (≤±10) except in Baoding (MB: -0.8), Shijiazhuang (MB:2.3), and BTH (MB: -9.6). Also, the ME values in the four cities and BTH region greatly exceeded the benchmark (≤±30), especially in Shijiazhuang (ME:101.5), Beijing (ME:78.4), and BTH region (ME:70.5). Similar model performance of WD had been reported (Hu et al., 2016; Sulaymon et al., 2021a, 2021b; Wang et al., 2021). Generally, in this study, the WRF model exhibited better performance when compared to previous studies in BTH region (Chang et al., 2019; Li et al., 2021b; Sulaymon et al., 2021a; Zhang et al., 2021; Zhao et al., 2021) and China as a country (Ma et al., 2021; Sulaymon et al., 2021b; Wang et al., 2021). Since the simulated meteorological parameters were robust, they were used in driving the air quality simulations.

3.2. CMAQ model performance

In evaluating the performance of CMAQ model in predicting $PM_{2.5}$, statistical indices, which include the mean observations (OBS), mean predictions (PRE), mean fractional bias (MFB), mean fractional error (MFE), mean normalized bias (MNB), and mean normalized error (MNE) were calculated. The performance of CMAQ model for $PM_{2.5}$ over the BTH and at four representative cities during the lockdown period based on the two cases are shown in Table S4. Generally, the simulated $PM_{2.5}$ concentrations exhibited good agreement with the observed data with the model performance indices

falling within the recommended benchmarks for $PM_{2.5}$ (MFB $\leq \pm 0.60$ and MFE ≤ 0.75) (Boylan and Russel, 2006) in BTH and the four cities for the two cases. For Case 1, $PM_{2.5}$ was over-estimated in BTH (0.10), Beijing (0.31), and Tianjin (0.41), while it was under-predicted in Shijiazhuang (-0.05) and Baoding (-0.19). Considering Case 2, all of the MFB values were negative except in Tianjin (0.32), an indication that CMAQ under-predicted the total $PM_{2.5}$ concentrations in BTH and the other three cities. Chang et al. (2019) had reported an under-estimation of $PM_{2.5}$ by CMAQ in Beijing and Shijiazhuang, which is consistent with this study for Case 2. Also, the model performances for Case 2 are in line with the findings of Sulaymon et al. (2021a) In addition, under-predictions of $PM_{2.5}$ in all of the prefectural-level cities of BTH region were reported by Jiang et al. (2021). The MFE values for the two cases ranged between 0.40-0.51, which were within the recommended benchmark (MFE$\leq$ 0.75). Overall, the CMAQ model has shown better performance in this study when compared to previous studies across the BTH region (Chang et al., 2019; Jiang et al., 2021; Li et al., 2021b; Sulaymon et al., 2021a; Zhang et al., 2021; Zhao et al., 2021). Thus, the model results were deemed acceptable for further analyses, including the IPR analysis.

3.3. IPR analysis of $PM_{2.5}$ formation at the surface layer

The hourly concentrations of $PM_{2.5}$ as well as the contributions of the individual atmospheric processes to the evolution of $PM_{2.5}$ at the surface layer in the BTH region and four representative cities for the two cases during the lockdown period are illustrated in Fig. 1. In the BTH region as a whole, the emissions (EMIS), horizontal transport (HTRA), and aerosol processes (AERO) were the major positive contributors (sources) to the net surface $PM_{2.5}$.

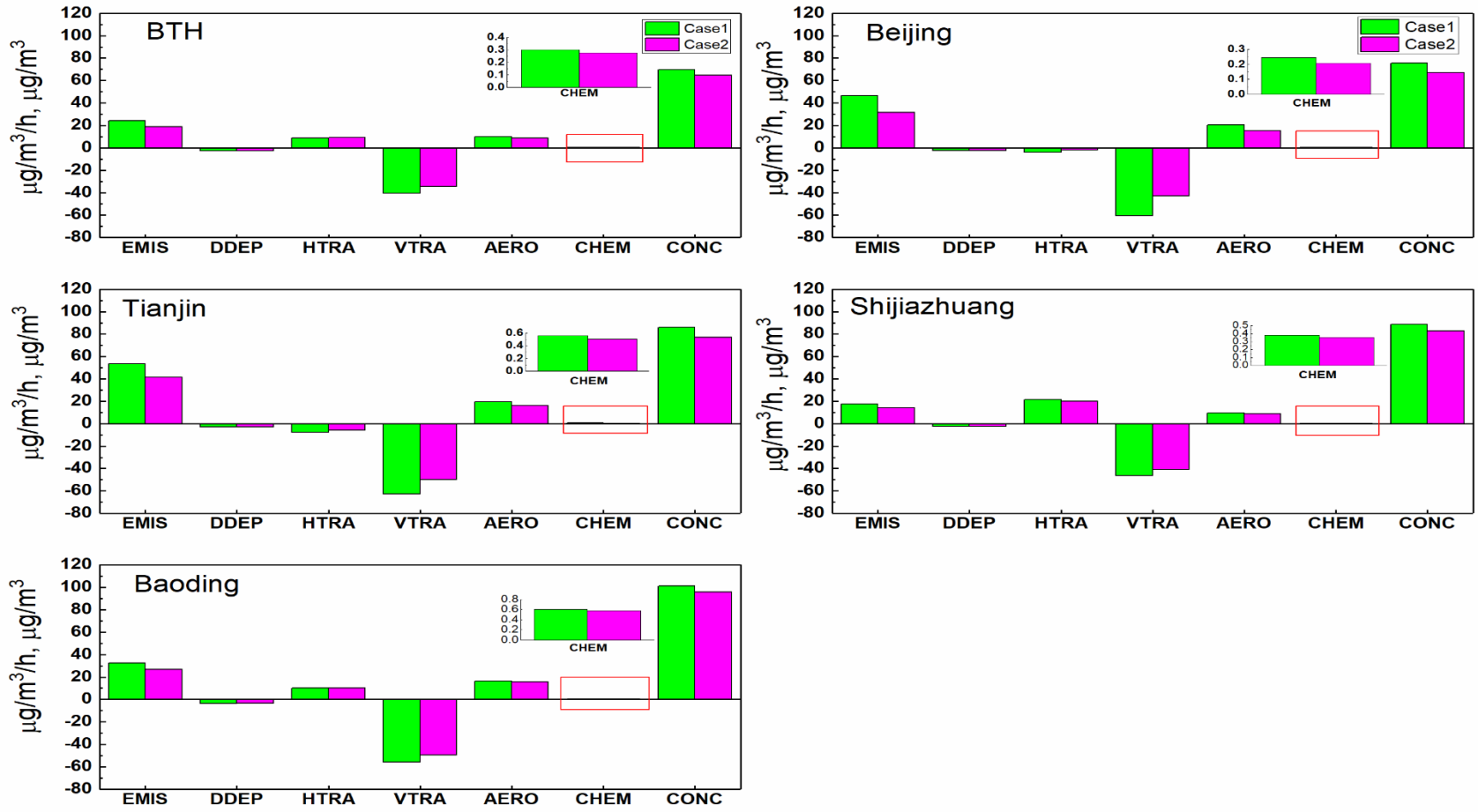


Fig. 1. Contributions of the individual processes to the concentrations of $PM_{2.5}$ at the surface layer in Cases 1 and 2 during the lockdown period. EMIS represents $PM_{2.5}$ input by emissions, DDEP represents $PM_{2.5}$ decrease by dry deposition; HTRA and VTRA represent $PM_{2.5}$ change by horizontal and vertical transport, respectively; AERO represents $PM_{2.5}$ change by the aerosol; and CHEM represents $PM_{2.5}$ change by gas-phase chemistry. The unit of the processes is $\mu g/m^3/h$. CONC is the hourly $PM_{2.5}$ concentrations in $\mu g/m^3$.

For Case 2, the contributions of EMIS, HTRA, and AERO to $PM_{2.5}$ formation were 42.4, 32.5, and 20.9% (Fig. 2b), respectively, while their contributions in the same order for Case 1 were 46.7, 29.4, and 20.3% (Fig. 2a), respectively. The reduction in the surface layer's $PM_{2.5}$ for the two cases was primarily attributed to the vertical transport (VTRA), while slight removal was also due to dry deposition (DDEP). In Beijing and Tianjin, EMIS and AERO were the predominant processes that contributed to the net surface $PM_{2.5}$ formation (Fig. 1) for both cases. The total contribution ratios of EMIS and AERO in Case 2 were 86.2% and 92.9% for Beijing and Tianjin (Fig. 2b), respectively. The reduction of surface $PM_{2.5}$ in Beijing and Tianjin for the two cases was associated with the VTRA, HTRA, and DDEP processes, with VTRA being the highest sink, with negative contributions of 79.0-81.3% (Beijing) and 80.2-81.0% (Tianjin). The results of the present study are consistent with those reported by Ye et al. (2022) in the coastal city of Kannur, India, where the EMIS, HTRA, and AERO were the dominant processes that positively contributed to $PM_{2.5}$ evolution, while VTRA and DDEP were responsible for surface $PM_{2.5}$ removal during the three periods considered in the study. Also, Fan et al. (2015) reported EMIS and VTRA as the two major processes that influenced $PM_{2.5}$ at the surface layer in the Pearl River Delta (PRD) region of China. Furthermore, Liu et al. (2010) and Xing et al. (2011) had earlier reported EMIS and AERO as the major $PM_{2.5}$ sources in both surface layer and the PBL in Beijing, while Xing et al. (2011) found VTRA as the major $PM_{2.5}$ sink in the surface layer. The $PM_{2.5}$ removal due to HTRA and DDEP in both cases were relatively the same in both Beijing and Tianjin. Considering Shijiazhuang and Baoding, similar trends were obtained regarding the contributions of individual processes to $PM_{2.5}$ formation.

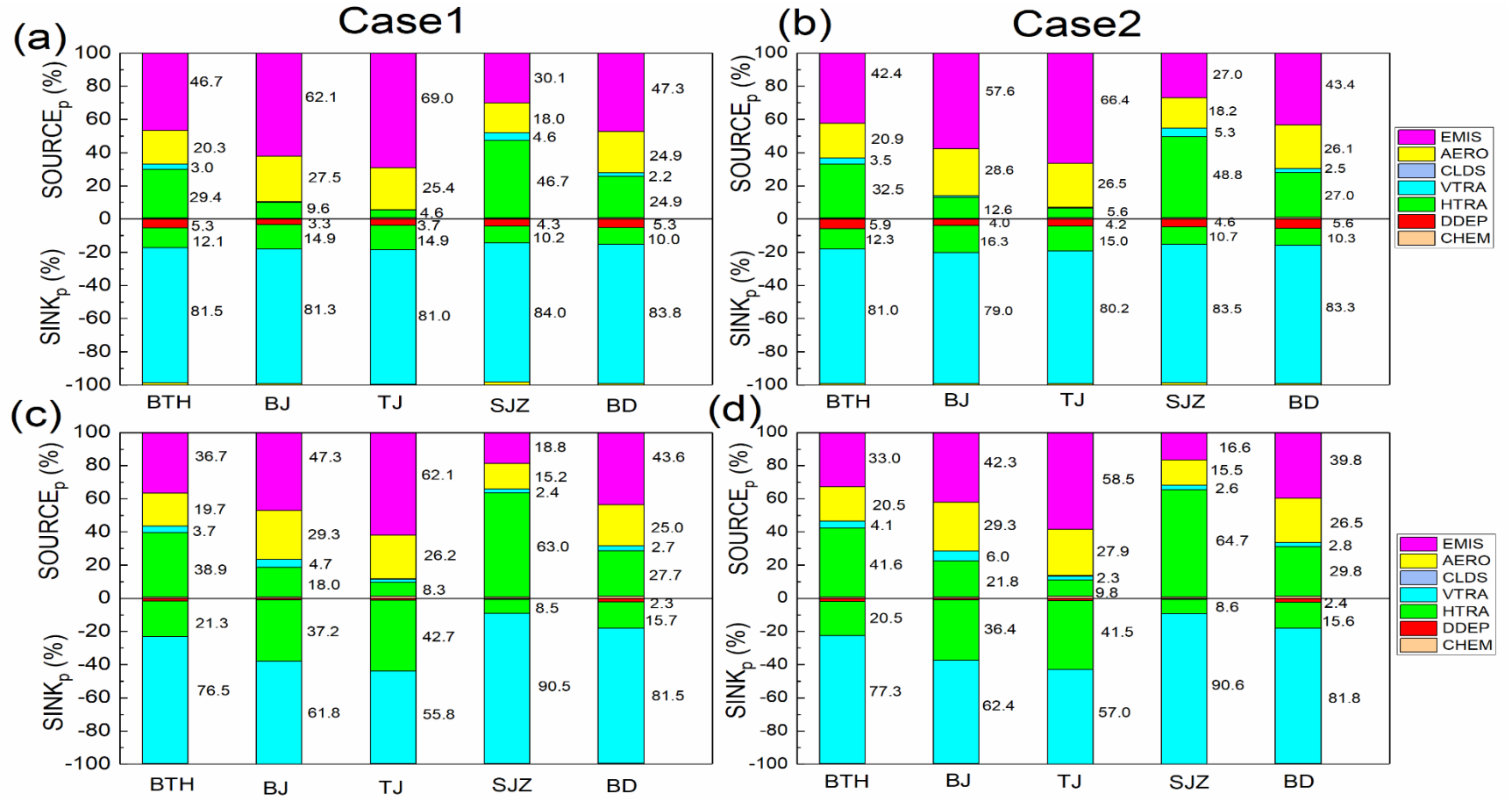


Fig. 2. Positive and negative contribution ratios of the individual processes to $PM_{2.5}$ concentrations (a,b) at the surface layer and (c,d) in the planetary boundary layer in Cases 1 and 2 during the lockdown period. EMIS, AERO, CLDS, VTRA, HTRA, DDEP, and CHEM represent the contributions of the emissions, aerosol, clouds, vertical transport, horizontal transport, dry deposition, and gas-phase chemistry, respectively to $PM_{2.5}$ formation.

The EMIS, AERO, and HTRA processes dominated the positive contributions to the net surface $PM_{2.5}$ in the two cities (Fig. 1), accounting for a total of 94.0 and 96.5% in Shijiazhuang and Baoding, respectively in Case 2 (Fig. 2b), while similar contributions were obtained in Case 1 (Fig. 2a). In addition to EMIS and AERO processes, the horizontal import of $PM_{2.5}$ to the surface layer via HTRA contributed to the elevated $PM_{2.5}$ concentrations in both Shijiazhuang and Baoding relative to Beijing and Tianjin. VTRA dominated the removal of $PM_{2.5}$ from the surface layer to upper layers in both cities, with higher rates in Case 1 relative to Case 2. DDEP process also had negative effects on $PM_{2.5}$ formation in the two cities, with very low contributions, especially in Shijiazhuang. The photochemistry (CHEM) process had positive net impacts on $PM_{2.5}$ evolution for the two cases across the BTH region and the four representative cities, however, the contributions were extremely low and negligible (Fan et al., 2015). This is contrary to what was reported in Kannur (Ye et al., 2022), as CHEM had negative effects on $PM_{2.5}$ in the city. Due to the negligible contributions of cloud (CLDS) processes to $PM_{2.5}$, it was not discussed in this study. It should be noted that the $PM_{2.5}$ concentrations in BTH region and the four cities in Case 2 were relatively low compared to Case 1. This could be attributed to low $PM_{2.5}$ formation from EMIS in Case 2. However, there was no substantive decrease in $PM_{2.5}$ concentrations in Case 2 despite reductions in anthropogenic emissions. This could be explained by the reduced $PM_{2.5}$ export from the surface layer to the upper layers due to low VTRA rates in Case 2 (compared to Case 1), leading to the accumulation of $PM_{2.5}$ in the surface layer, which subsequently led to high $PM_{2.5}$ pollution during the lockdown period.

3.4. IPR analysis of $PM_{2.5}$ formation in the PBL

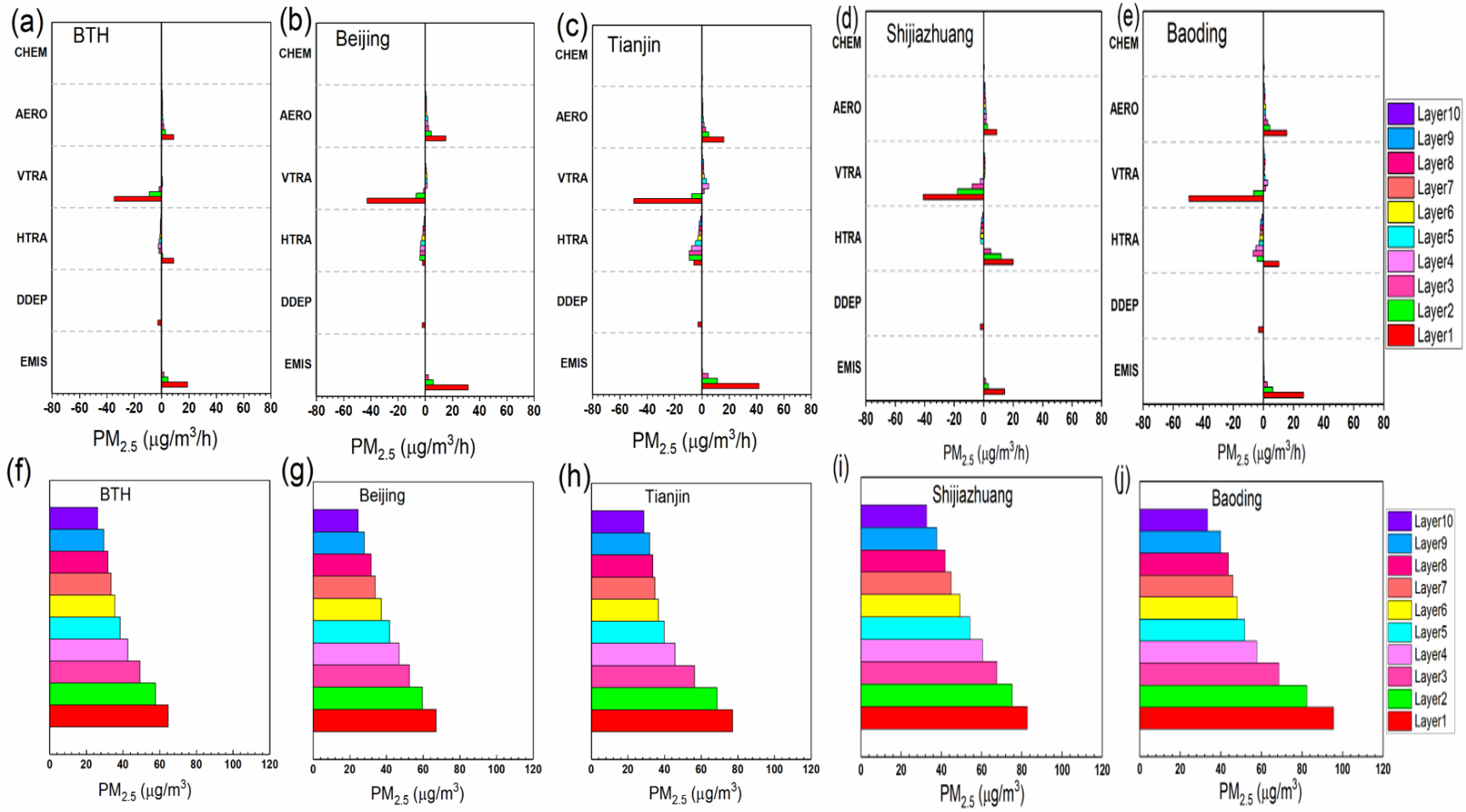
Fig. S2 shows the mean hourly change rates attributed to individual atmospheric processes to $PM_{2.5}$ production and the concentrations of $PM_{2.5}$ in the PBL during the lockdown. The contributions of the various processes to $PM_{2.5}$ formation within the PBL in BTH and the four cities followed the similar trends as found in the surface layer. However, the contributions of the individual processes to $PM_{2.5}$ were smaller in the PBL compared to the surface layer, as $PM_{2.5}$ concentrations decrease as vertical layers increase (Fan et al., 2015). Generally, the contributions of EMIS and VTRA to the net $PM_{2.5}$ (Case 2) were low in the PBL compared to the surface layer. For instance, relative to what was obtained to the surface layer, the rates due to EMIS and VTRA in the PBL decreased by half in BTH region and all of the representative cities. Similar to the surface layer, the EMIS, HTRA, and AERO were the predominant contributors to the net $PM_{2.5}$ in the whole BTH region and Baoding, while only EMIS and AERO processes contributed substantially to the net $PM_{2.5}$ in Beijing and Tianjin. In Shijiazhuang, however, HTRA was the dominant contributor to $PM_{2.5}$ formation. Compared to other processes, the contributions of CHEM process were extremely low and negligible (Fan et al., 2015; Ye et al., 2022). In addition to VTRA as the major process responsible for the removal of $PM_{2.5}$ across the study areas, $PM_{2.5}$ removal in Beijing and Tianjin was also associated with HTRA. In all of the study areas, slight removal of $PM_{2.5}$ was also attributed to DDEP process. It could be noted that the VTRA and HTRA effects within the PBL were opposite to those to the surface layer. As illustrated in Fig. 2(d), the negative contributions (sinks) due to VTRA in the entire PBL substantially reduced in all of the study areas except Shijiazhuang (increased) when compared to the surface layer. Contrary to the surface layer, the sinks ($PM_{2.5}$ removal) due

to HTRA within the PBL increased in all of the study areas except Shijiazhuang (decreased). It is worth noting that despite the decreases in EMIS rates by half (which might have adversely influenced $PM_{2.5}$ formation within the PBL) in comparison to the surface layer, the absolute difference in $PM_{2.5}$ concentrations between the surface layer and PBL was not significant, and ranged between 5.7-9.1 $\mu g/m^3$ across the study areas. This could be attributed to reduced $PM_{2.5}$ export due to low VTRA rates, leading to the accumulation of $PM_{2.5}$ within the PBL, and subsequently resulted to high $PM_{2.5}$ concentrations. Overall, in the PBL, EMIS and VTRA served as the two dominant processes that impacted $PM_{2.5}$ in BTH, Beijing, Tianjin, and Baoding, while VTRA and HTRA were the two major processes that influenced $PM_{2.5}$ formation in Shijiazhuang.

3.5. Vertical profiles of the atmospheric processes contributing to $PM_{2.5}$

The mean hourly $PM_{2.5}$ change rates attributed to individual atmospheric processes for the first ten layers (layers 1-10), as well as the vertical profiles of $PM_{2.5}$ evolution for Case 2 during the lockdown period are illustrated in Fig. 3, while that of Case 1 are shown in Fig. S3. As earlier stated in section 3.4, the characteristics of $PM_{2.5}$ concentrations at upper layers (layer 4 and above) were different from near-surface layers, hence, the contributions of emissions sources at upper layers were negligible (Fan et al., 2015). Across the study areas, the contributions from EMIS sources were only found within layers 1-3 (Fig. 3a-e) (Fan et al., 2015; Ye et al., 2022), and this was associated with the height of the emissions sources (Fan et al., 2015; Ye et al., 2022). The highest and lowest contributions of EMIS were found in Tianjin and Shijiazhuang, respectively, and the contribution decreased as the vertical layer increased. Within the first three layers, AERO process was another major source of $PM_{2.5}$ in all of the study areas, and the formation rate of $PM_{2.5}$

through the AERO process decreased as the vertical layer increased. Furthermore, VTRA contributed negatively and served as the predominant sink for removing the near-surface $PM_{2.5}$ at the lower layers in Beijing (layers 1-3), Shijiazhuang (layers 1-4), Tianjin and Baoding (layers 1-2), while it slightly contributed positively (acted as source) at the upper layers. This is consistent with the results of Fan et al. (2015), in which VTRA was reported as a sink in the near-surface layers and a source in the upper layers (layer 4 and above). In Beijing (Fig. 3b) and Tianjin (Fig. 3c), HTRA served as another sink for $PM_{2.5}$ across the vertical layers, and the rate initially increased between layers 1 and 2, but continuously decreased as the vertical layer increased. In Shijiazhuang (Fig. 3d), HTRA contributed positively at the lower layers (layers 1-3) and negatively at the upper layers. Considering Baoding (Fig. 3e), HTRA only acted as the source at the first layer, while it behaved as the sink from the second layer upward. It could be deduced that there were vertical and horizontal exports of $PM_{2.5}$ in Beijing and Tianjin at the surface layer, while Shijiazhuang, Baoding, and the whole BTH region witnessed vertical export and horizontal import of $PM_{2.5}$ in the surface layer. DDEP acted as another sink of $PM_{2.5}$, and only existed at the first layer across the study areas (Fan et al., 2015; Ye et al., 2022). DDEP contributions were only found in the first layer because dry deposition was treated as a bivariate variable by the CMAQ model, and integrated it over the whole atmospheric column (Fan et al., 2015). As shown in Fig. 3(f-j), the highest $PM_{2.5}$ concentration was found in the surface layer, and decreased with increases in vertical layer height (Fan et al., 2015). This result could be attributed to the contributions of EMIS and AERO processes, as well as the decreasing trends of the two processes as the vertical layer increased.



435

436 **Fig. 3.** Hourly $PM_{2.5}$ change rates due to individual atmospheric processes for layers 1-10 (a-e) and evolution of hourly $PM_{2.5}$ vertical
437 profiles (f-j) in Case 2 during the lockdown period. Abbreviations used in this figure are the same as in Fig. 1.

438

3.6 IPR analysis of $PM_{2.5}$ pollution episodes during lockdown

During the lockdown period, the predicted $PM_{2.5}$ concentrations (in Case 2) (Fig.4; Tables S5-S6) indicated that persistent $PM_{2.5}$ pollution episodes could not be avoided in the BTH region despite reductions in anthropogenic emissions (Sulaymon et al., 2021a). Fig. S6 illustrates the spatial distributions of $PM_{2.5}$ in the BTH region during the lockdown for the two cases. Fig. S6(b) shows that the $PM_{2.5}$ concentrations during the lockdown were higher ($PM_{2.5}\geq 75$ μg/m^3, level II of Chinese air quality standard) in Tianjin and southern Hebei Province, while the northern Hebei was characterized with low concentrations ($PM_{2.5}\leq 50$ μg/m^3). In the prefectural-level cities of BTH region, three severe $PM_{2.5}$ pollution episodes (EPs) (Fig. 4) occurred during the lockdown (Case 2). They are represented as EP1 (January 24-31, 2020), EP2 (February 7-13), and EP3 (February 19-21). It should be noted that EP1 and EP2 coincided with the 2020 Spring (January 25) and Lantern (February 8) festivals, respectively. The statistics for all the EPs in each city are enumerated in Tables S5-S6. For the purpose of process analysis of $PM_{2.5}$ during the EPs, Beijing, Tianjin, Shijiazhuang, and Baoding were selected as the representative cities. As illustrated in Fig. 5, Beijing and Tianjin had the highest $PM_{2.5}$ concentrations during EP2, while Shijiazhuang and Baoding recorded their highest $PM_{2.5}$ concentrations during EP1. Also, during EP2 and EP3, Baoding experienced severe pollution with elevated $PM_{2.5}$ concentrations. This indicates that the region suffered severe pollution episodes during the lockdown. Dai et al. (2021) and Sulaymon et al. (2021a) had previously reported severe haze episodes in the BTH region during the lockdown. Therefore, it becomes pertinent to elucidate the major atmospheric processes responsible for the formation of the EPs.

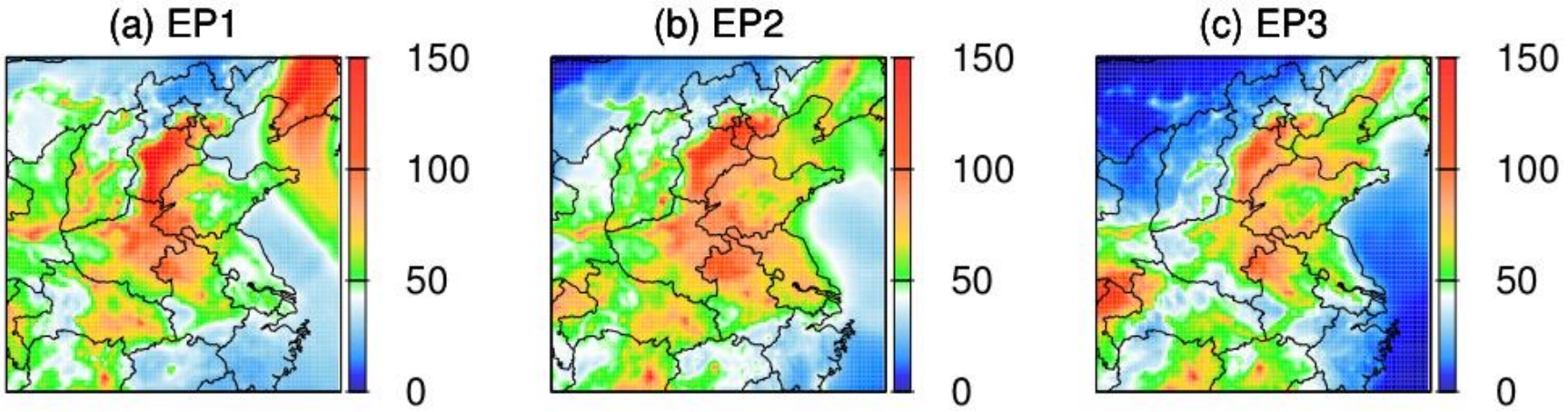


Fig. 4. Spatial distributions of predicted $PM_{2.5}$ (Case 2) during (a) EP1, (b) EP2, and (c) EP3 in the BTH region. Units are $\mu g/m^3$.

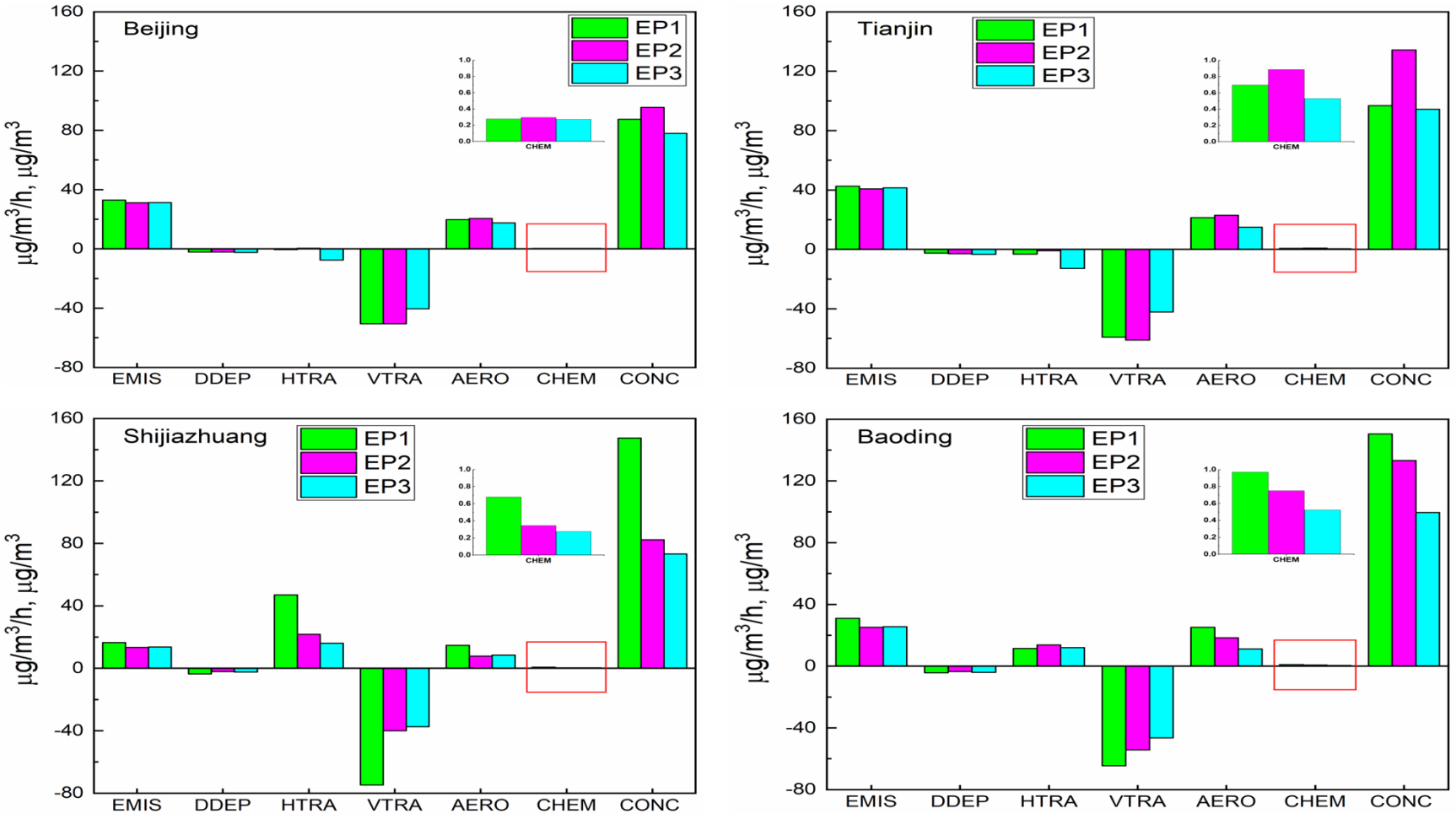


Fig. 5. Contributions of the individual processes to the concentrations of $PM_{2.5}$ (Case 2) at the surface layer during the three pollution episodes in the four representative cities. Abbreviations used in this figure are the same as in Fig. 1.

The contributions of different atmospheric processes to $PM_{2.5}$ formation (in Case 2) during the three EPs in the four representative cities were analyzed. Table S7 shows the average planetary boundary layer height (PBLH) and wind speed for the four cities during the three EPs. The PBLH during the third pollution episode (EP3; PBLH>400 m) was higher than the PBLH during the first two pollution episodes (EP1 and EP2; PBLH<370 m) across the study areas. A thinner boundary layer is more conducive for the accumulation of locally-emitted particles, leading to increased $PM_{2.5}$ concentrations and results in haze events (Fan et al., 2015). As illustrated in Fig. 5, the three pollution episodes in Beijing and Tianjin were principally caused by local emissions (EMIS), while the pollution events in Shijiazhuang and Baoding could be attributed to both local emissions (EMIS) and regional transport (HTRA).

Fig. 5 shows that emissions sources and aerosols were the major positive contributors to $PM_{2.5}$ pollution in Beijing, Tianjin, and Baoding, while horizontal transport was the most significant positive contributor to pollution level in Shijiazhuang, followed by emissions and aerosols. In Baoding, however, HTRA also contributed positively towards $PM_{2.5}$ concentrations throughout the three episodes. There were no significant differences between emissions sources among the three episodes in all of the study areas, and the average total emissions ranged between 13.3-42.6 $\mu g/m^3/h$ across the EPs in the cities. During the three episodes across the four cities, $PM_{2.5}$ was released into the atmosphere through EMIS and AERO processes, and fell back to the surface layer via DDEP process (Fan et al., 2015), with very low rates of dry deposition (ranged between -2.1 $\mu g/m^3/h$ and -4.3 $\mu g/m^3/h$). Also, $PM_{2.5}$ was transported and diffused through VTRA and HTRA processes. In Beijing and Tianjin, there were negligible differences between

the contributions from VTRA during the first two episodes, and the total rates were approximately -51 $\mu g/m^3/h$ and -60 $\mu g/m^3/h$ in Beijing and Tianjin, respectively. During EP3 in the two cities, the contributions by VTRA (-40 $\mu g/m^3/h$ in Beijing; and -42 $\mu g/m^3/h$ in Tianjin) were low relative to the first two episodes. The VTRA contributions in Shijiazhuang (-75 $\mu g/m^3/h$) and Baoding (-65 $\mu g/m^3/h$) during EP1 were greater than those contributed in Beijing and Tianjin during the same period, and this was due to lower PBLH values (Table S7) in Shijiazhuang and Baoding relative to Beijing and Tianjin. The difference in the VTRA rates between the first two episodes and the third episode in Beijing and Tianjin could be explained by the accumulation of particulates on near-surface layers due to the nature of boundary layer (thinner) (Fan et al., 2015) being exhibited during the first two pollution episodes. Therefore, VTRA had a greater clearing impact for $PM_{2.5}$ during the first two episodes in the two cities. Contrarily, during the third episode in the two cities, a more uniform vertical mixing of $PM_{2.5}$ was achieved, and this was due to the thicker PBLHs during EP3 (Table S7). Hence, the clearing effect of VTRA during EP3 was low compared to EP1 and EP2. In addition, due to higher wind speed during EP3 (Table S7), the negative contributions due to HTRA were higher in Beijing (-8 $\mu g/m^3/h$) and Tianjin (-13 $\mu g/m^3/h$) during EP3 compared to EP1 and EP2, and this subsequently reduced the contributions from VTRA during EP3. In Shijiazhuang, VTRA also exhibited a very greater clearing effect during EP1 than EP2 and EP3, and similar scenario also occurred in Baoding. In Shijiazhuang, HTRA was the dominant positive contributor to $PM_{2.5}$ throughout the episodes, with the highest contribution rate during EP1. In Baoding, however, there was negligible difference between the contributions from HTRA to $PM_{2.5}$ pollution during the three episodes. Due to availability of several emissions sources in

Beijing and Tianjin, which result to a large quantity of local emissions, $PM_{2.5}$ concentrations were generally higher in the two cities. Hence, the effects of both VTRA and HTRA on pollution levels were negative during the three pollution episodes, and both mainly provided dilution and clearing effects in the two cities (Fan et al., 2015). On the other hand, in both Shijiazhuang and Baoding, horizontal transport contributed positively and significantly increased $PM_{2.5}$ concentrations during the three episodes. It is also worthy to mention that $PM_{2.5}$ concentrations during the three episodes in the four cities were greatly and positively influenced by the planetary boundary layer height, as the episode with the lowest PBLH had the highest $PM_{2.5}$ concentration in a city. The results of the present study are consistent with those reported by Fan et al. (2015) during three pollution episodes over the PRD region. Fan et al. (2015) had reported surface emissions, aerosol processes, and horizontal transport as the major contributors to air pollution episodes over the PRD. In the PBL (Fig. S7), EMIS and AERO were also the dominant contributors to $PM_{2.5}$ concentrations in Beijing and Tianjin, HTRA was the most important source in Shijiazhuang, and EMIS, AERO, and HTRA actively contributed to $PM_{2.5}$ formation in Baoding during the EPs. VTRA was the major removal pathway. However, the contribution and removal rates were low in the PBL relative to the surface layer.

To better understand the roles of the atmospheric processes towards the pollution episodes, $PM_{2.5}$ formation and removal within the PBL (layers 1-10) were analyzed in Tianjin (Fig. 6), while those of other three cities are illustrated in Figs S8-S10. Considering the three EPs in Tianjin, the positive contributions of EMIS and AERO processes to the hourly $PM_{2.5}$ significantly occurred within layers 1-3 (Fig. 6a-c), while they both contributed less at upper layers. Also, there were vertical imports of $PM_{2.5}$ (although very

548 low) at upper layers (layers 3-10 for EP1 and EP2; and layers 3-6 for EP3). Conversely,
549 VTRA (layers 1-2) and HTRA (layers 1-10) served as the predominant sinks and $PM_{2.5}$
550 removal pathways. DDEP process also acted as another sink for $PM_{2.5}$ during the EPs, and
551 only existed at the first layer (Fan et al., 2015). The contributions by CHEM process were
552 negligible across the vertical layers (Fan et al., 2015; Ye et al., 2022). As shown in Fig.
553 6(d-f), EP2 was characterized with the highest $PM_{2.5}$ concentrations, followed by EP1 and
554 EP3. Furthermore, the $PM_{2.5}$ formation processes in the surface layer during the EPs were
555 compared. Fig. 7 illustrates the percentage contributions of the atmospheric processes to
556 $PM_{2.5}$ formation/removal during the EPs in the four cities. The total contributions of EMIS
557 and AERO (EMIS+AERO) during the EPs ranged between 80-89% and 88-97% in Beijing
558 (Fig. 7a) and Tianjin (Fig. 7b), respectively. In Shijiazhuang (Fig. 7c), the contributions
559 due to HTRA during the episodes ranged between 44-61%, making it the major $PM_{2.5}$
560 source. As earlier revealed in Fig. 5, $PM_{2.5}$ formation during the episodes in Baoding (Fig.
561 7d) was attributed to the contributions of EMIS, AERO, and HTRA, with total
562 contributions of 93-98% during the EPs. In the four cities, $PM_{2.5}$ removal was dominantly
563 influenced by VTRA during the EPs.

564 Furthermore, the diel variations of the contributions of various atmospheric
565 processes to the formation of $PM_{2.5}$ as well as the hourly variations of $PM_{2.5}$ concentrations
566 at the surface layer during the three episodes are illustrated in Fig. 8. In Beijing, EMIS and
567 AERO processes were the major $PM_{2.5}$ sources, and showed two peaks (07:00 LT and
568 20:00 LT) during the three episodes.

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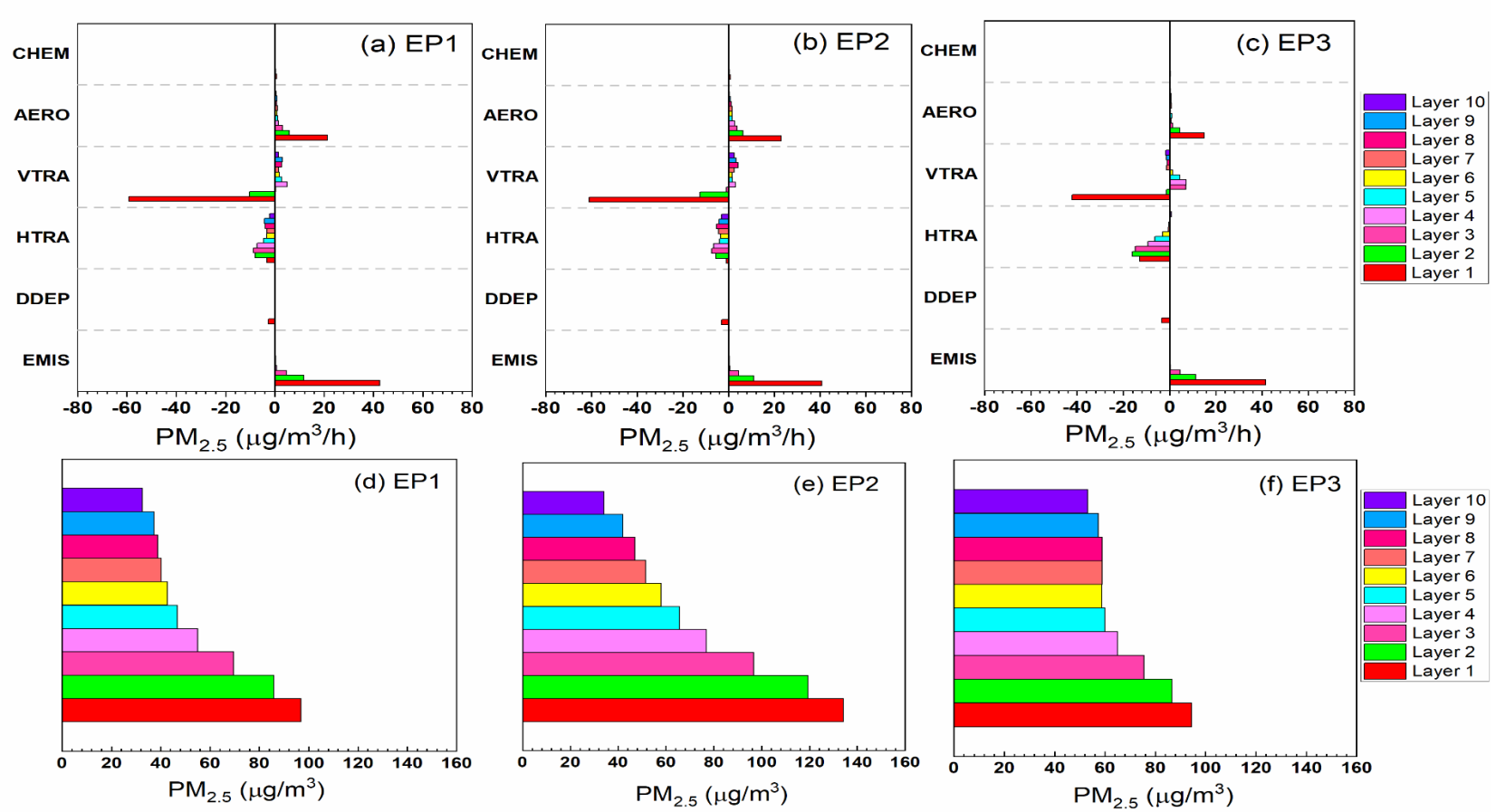

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572 **Fig. 6.** Hourly $PM_{2.5}$ change rates (Case 2) due to individual atmospheric processes for layers 1-10 (a-c) and evolution of hourly $PM_{2.5}$
573 vertical profiles (d-f) during the three pollution episodes in Tianjin. Abbreviations used in this figure are the same as in Fig. 1.
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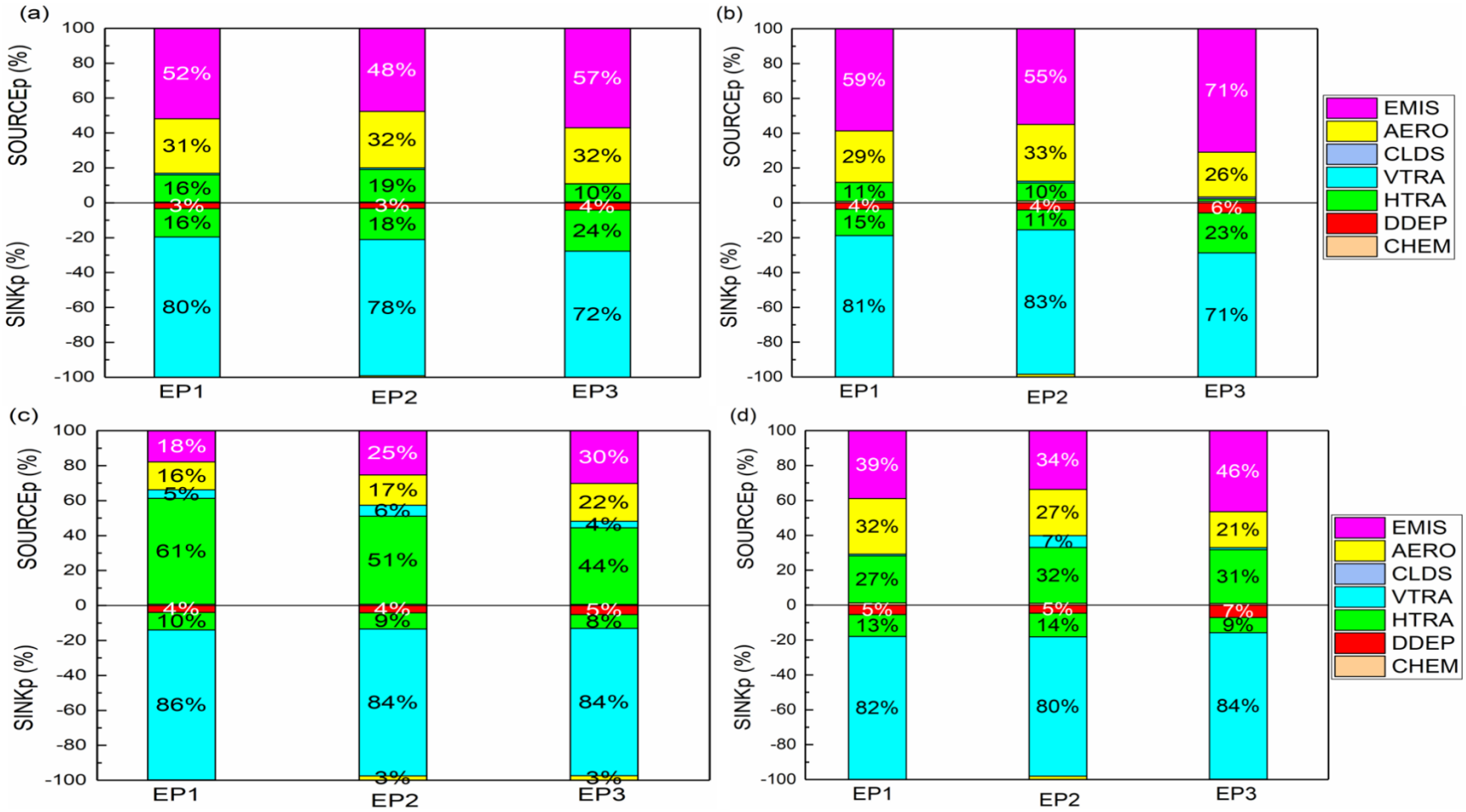
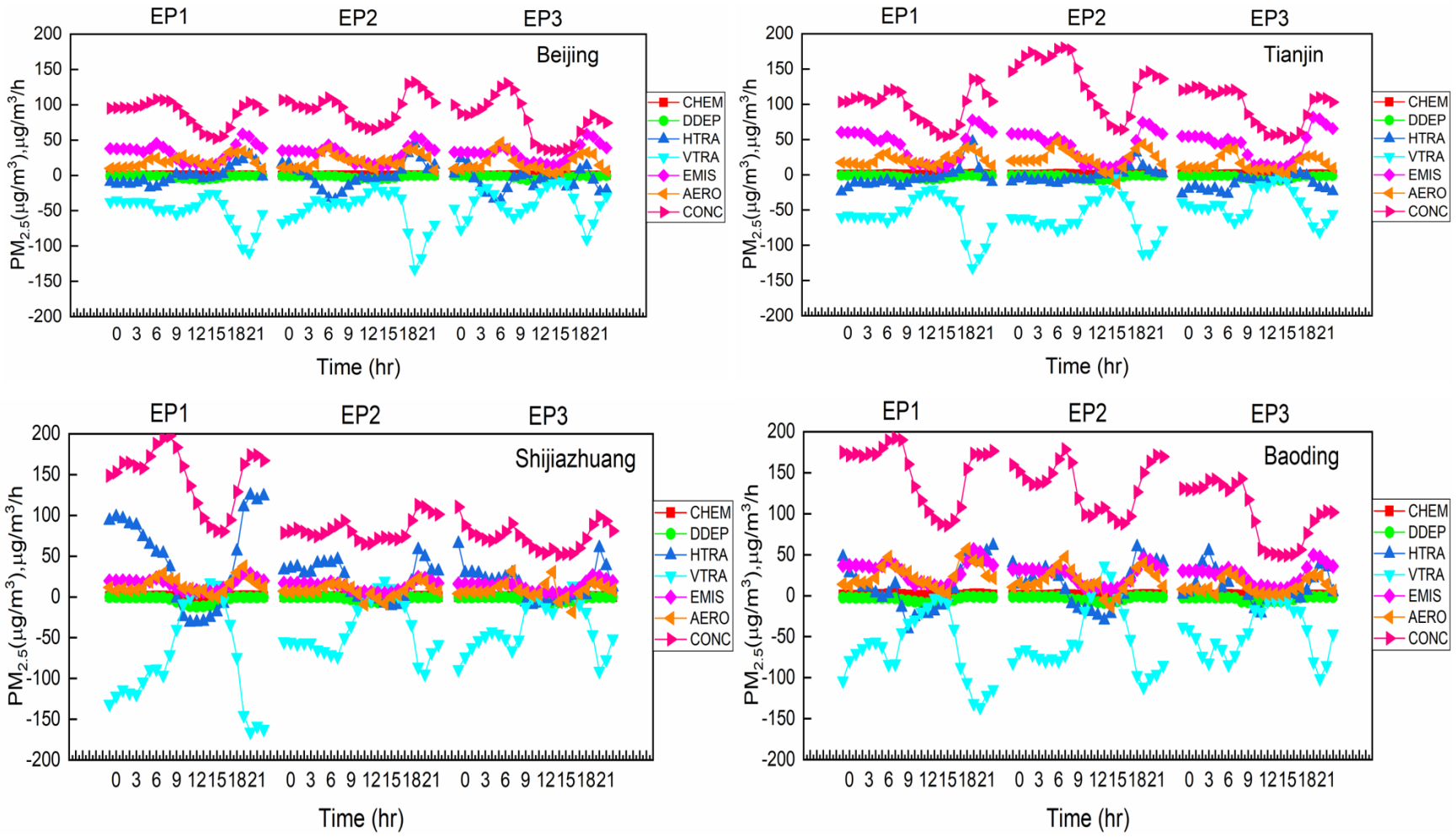


Fig. 7. Positive and negative contribution ratios of the individual processes to $PM_{2.5}$ concentrations (Case 2) at the surface layer in (a) Beijing, (b) Tianjin, (c) Shijiazhuang, and (d) Baoding during the three pollution episodes. Abbreviations used in this figure are the same as in Fig. 2.



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581 **Fig. 8.** Diel variations of contributions of individual processes to $PM_{2.5}$ formation (Case 2) at the surface layer during the three pollution
582 episodes in the four representative cities. Abbreviations used in this figure are the same as in Fig. 1.

583 The highest rates of EMIS during the EPs ranged between 54.8-58.6 μg/m^3/h, all occurred
584 at 20:00 LT. The VTRA dominated the $PM_{2.5}$ removal, with the highest removal rates of -
585 109.3 μg/m^3/h (at 21:00 LT), -132.6 μg/m^3/h (at 20:00 LT), and -90.7 μg/m^3/h (at 20:00
586 LT) during EP1, EP2, and EP3, respectively. In addition, HTRA was another major $PM_{2.5}$
587 removal pathway during the EPs. However, HTRA later acted as another $PM_{2.5}$ source
588 during EP1 (17:00-22:00 LT; with maximum rate of 29.6 μg/m^3/h at 21:00 LT), EP2
589 (00:00-03:00 and 18:00-23:00 LT; with maximum rate of 44.7 μg/m^3/h at 20:00 LT), and
590 EP3 (00:00-02:00 and 19:00-20:00 LT; with highest rate of 24.0 μg/m^3/h at 01:00 LT),
591 leading to the horizontal import of $PM_{2.5}$ during the periods. Considering Tianjin, EMIS
592 process was the dominant $PM_{2.5}$ source and exhibited two distinct peaks (00:00 LT and
593 20:00 LT) during the three episodes. The highest contributions of EMIS during EP1, EP2,
594 and EP3 were 77.7 μg/m^3/h, 74.1 μg/m^3/h, and 81.7 μg/m^3/h, respectively, all occurred at
595 20:00 LT. AERO process was another $PM_{2.5}$ source with two peaks during EP1 (07:00 LT
596 and 19:00 LT), EP2 and EP3 (07:00 LT and 20:00 LT). The negative contributions of
597 VTRA made it the dominant $PM_{2.5}$ sink throughout the 24hrs period, with the maximum
598 removal rates of -132.1 μg/m^3/h (at 20:00 LT), -112.5 μg/m^3/h (at 20:00 LT), and -81.3
599 μg/m^3/h (at 21:00 LT) during EP1, EP2, and EP3, respectively. Besides VTRA, HTRA
600 served as the second major $PM_{2.5}$ sink during the three EPs. However, HTRA later became
601 another $PM_{2.5}$ source during EP1 (16:00-21:00 LT; with maximum rate of 47.5 μg/m^3/h),
602 EP2 (14:00-23:00; with maximum rate of 31.1 μg/m^3/h), and EP3 (17:00-18:00 LT; with
603 very low rates). In Shijiazhuang, HTRA was the major $PM_{2.5}$ contributor, and showed two
604 peaks during EP1 (98.3 μg/m^3/h at 01:00 LT and 124.8 μg/m^3/h at 21:00 LT), EP2 (45.8
605 μg/m^3/h at 08:00 LT and 57.8 μg/m^3/h at 20:00 LT), and EP3 (65.2 μg/m^3/h at 00:00 LT

and 60.3 μg/m^3/h at 21:00 LT). The VTRA was the dominant $PM_{2.5}$ removal pathway, with the highest rates of -165.6 μg/m^3/h, -94.5 μg/m^3/h, and -91.0 μg/m^3/h during EP1, EP2, and EP3, respectively, all at 21:00 LT. However, VTRA later became positive and served as another $PM_{2.5}$ source during EP1 (13:00-16:00 LT; with maximum rate of 17.4 μg/m^3/h at 15:00 LT), EP2 (14:00-17:00 with maximum rate of 19.4 μg/m^3/h at 15:00 LT), and EP3 (15:00-17:00 with highest rate of 13.6 μg/m^3/h at 17:00 LT), resulting to the vertical import of $PM_{2.5}$ during the periods. In Baoding, EMIS, AERO, and HTRA were the major $PM_{2.5}$ formation pathways during nighttime, while EMIS and AERO were the dominant sources during daytime. Being the major $PM_{2.5}$ removal pathway, VTRA had the highest rates of -136.3 μg/m^3/h (21:00 LT), -111.4 μg/m^3/h (20:00 LT), and -101.1 μg/m^3/h (21:00 LT) during EP1, EP2, and EP3, respectively. During EP2, VTRA shortly behaved as another $PM_{2.5}$ source (13:00-15:00 LT, with highest rate of 36.5 μg/m^3/h. With very low rates during the episodes, DDEP and CHEM processes served as $PM_{2.5}$ sink and source, respectively across the four cities. The $PM_{2.5}$ concentrations peaked in Beijing (EP1: 107.8 μg/m^3 at 07:00 LT; EP2: 132.1 μg/m^3 at 20:00 LT; EP3: 130.3 μg/m^3 at 08:00 LT), Tianjin (EP1: 135.7 μg/m^3 at 20:00 LT; EP2: 180.1 μg/m^3 at 08:00 LT; EP3: 124.6 μg/m^3 at 02:00 LT), Shijiazhuang (EP1: 197.6 μg/m^3 at 09:00 LT; EP2: 112.6 μg/m^3 at 20:00 LT; EP3: 110.3 μg/m^3 at 00:00 LT), and Baoding (EP1: 192.0 μg/m^3 at 08:00 LT; EP2: 178.2 μg/m^3 at 08:00 LT; EP3: 142.8 μg/m^3 at 09:00 LT).

4. Conclusions

This study employed the PA tool in the CMAQ model to identify and quantify the contributions of individual atmospheric processes and meteorology to the three $PM_{2.5}$ pollution episodes that occurred during the COVID-19 lockdown in the BTH region even

with the required reductions in human activities. Due to emission reductions, the total $PM_{2.5}$ concentrations across the BTH decreased by 6.2-11.0%. However, the region still experienced three $PM_{2.5}$ pollution episodes during the lockdown. The IPR results showed that the EMIS and AERO processes were the dominant positive contributors to the net surface $PM_{2.5}$ in Beijing and Tianjin, while the EMIS, HTRA, and AERO pathways dominated the net surface $PM_{2.5}$ formation in Shijiazhuang and Baoding. In Case 2, the decrease in surface $PM_{2.5}$ concentrations across the BTH was primarily attributed to the reduced EMIS and AERO processes, which shows the reduction in the primary source of $PM_{2.5}$ as well as decrease in the formation of secondary aerosol through gas-to-particle conversion. Both vertical and horizontal transport had significant impacts on the changes in surface $PM_{2.5}$. Elevated $PM_{2.5}$ concentrations (in Case 2) in the BTH region during the lockdown could be attributed to a low vertical transport rate of $PM_{2.5}$ from the surface layer to the upper layers. Furthermore, during the three pollution episodes, EMIS and AERO processes were the dominant sources of $PM_{2.5}$ formation in Beijing, Tianjin, and Baoding, while HTRA was the major source in Shijiazhuang. In all of the four cities, vertical transport served as the major $PM_{2.5}$ sink throughout the episodes, with differences in vertical rates between the episodes in each city. The pollution levels in the four cities were greatly and positively influenced by the PBLH, as the episode with the lowest PBLH had the highest $PM_{2.5}$ concentration in a city. This study reveals the various atmospheric processes and meteorological factors governing the $PM_{2.5}$ formation during the severe pollution episodes in the BTH region, as well as the changes in the individual atmospheric processes and $PM_{2.5}$ concentrations due to the lockdown measures, and shows that the existing emissions control strategies could not prevent pollution episodes in the region,

especially during the winter period. Since it is not possible to control the aerosol and transport processes, only further changes in emissions will reduce the severity of the episodes. Thus, better forecasting of the conditions that would foment such episodes combined with more effective emissions control strategies are urgently required to be able to mitigate such future severe pollution episodes in the BTH region.

Acknowledgements

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Conflict of Interest

The authors declare that they have no conflict of interest.

Open Research

The simulated and the observation data ($PM_{2.5}$ and meteorological variables) used in this study for model evaluation and postprocessing (Figures and Tables) can be found in Sulaymon et al. (2023).

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