Science of the Total Environment 779 (2021) 146619

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Quantifying the impacts of inter-city transport on air quality in the Yangtze River Delta urban agglomeration, China: Implications for regional cooperative controls of $PM_{2.5}$ and O_3

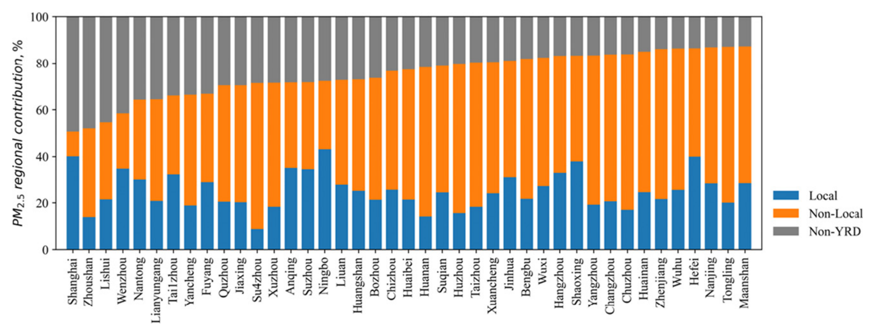
Kangjia Gong [a], Lin Li [a], Jingyi Li [a], Momei Qin [a], Xueying Wang [a], Qi Ying [b], Hong Liao [a], Song Guo [c], Min Hu [c], Yuanhang Zhang [c,d], Jianlin Hu [a,*]

[a] *Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control, Collaborative Innovation Center of Atmospheric Environment and Equipment Technology, Nanjing University of Information Science & Technology, Nanjing 210044, China*
[b] *Zachry Department of Civil Engineering, Texas A&M University, College Station, TX 77843, USA*
[c] *State Key Joint Laboratory of Environmental Simulation and Pollution Control, College of Environmental Sciences and Engineering, Peking University, Beijing 100871, China*
[d] *CAS Center for Excellence in Regional Atmospheric Environment, Chinese Academy of Science, Xiamen 361021, China*

HIGHLIGHTS

- Inter-city transport among 41 YRD cities is conducted for O_3 and $PM_{2.5}$ during the EXPLORE-YRD campaign.
- Inter-city transport has the largest contribution to $PM_{2.5}$ and O_3 in most cities.
- Transport from nearby cities becomes more important on pollution days.
- The cities within a distance of 184 km and 94 km contribute 60% of the $PM_{2.5}$ and O_3, respectively.
- Reducing secondary inorganic $PM_{2.5}$ in YRD requires cooperative emission controls on a much larger regional scale.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 23 December 2020
Received in revised form 14 March 2021
Accepted 15 March 2021
Available online 19 March 2021

Editor: Pingqing Fu

Keywords:
Yangtze River Delta
Inter-city transport
EXPLORE-YRD campaign
Secondary $PM_{2.5}$
Source-oriented model

ABSTRACT

The Yangtze River Delta (YRD) urban agglomeration is one of the most developed regions in China. During recent decades, this region has experienced severe regional haze and photochemical smog pollution problems. In this study, we used a source-oriented chemical transport model to quantitatively estimate the effects of inter-city transport on fine particulate matter ($PM_{2.5}$) and ozone (O_3) among the 41 cities in the YRD urban agglomeration during the EXPLORE-YRD (EXPeriment on the eLucidation of the atmospheric Oxidation capacity and aerosol foRmation, and their Effects in the Yangtze River Delta) campaign (May 17 to June 17, 2018). The results show that inter-city transport is very significant in the YRD region. On average, the emissions from the local city, the other YRD cities, and the regions outside of the YRD contribute 25.3%, 49.9%, and 24.8% to the $PM_{2.5}$, respectively, and they contribute 33.7%, 46.8%, and 19.5% of the non-background O_3, respectively. On $PM_{2.5}$ or O_3 pollution days, the transport contribution from the non-local YRD cities becomes much more important, while the local emissions and the transport from non-YRD emissions become less important. The results also suggest that the cities within a distance of 184 km and 94 km contribute 60% of the $PM_{2.5}$ and O_3, respectively. Therefore, we recommend that regional cooperative control programs in the YRD consider emission controls over cities within these ranges. The range for primary $PM_{2.5}$ (92 km) is very different from that for secondary $PM_{2.5}$ (515 km). Cooperative emission controls of SO_2 and NO_x on a much larger regional scale are required to reduce the secondary $PM_{2.5}$ in the YRD.

* Corresponding author.
E-mail address: jianlinhu@nuist.edu.cn (J. Hu).

https://doi.org/10.1016/j.scitotenv.2021.146619
0048-9697/

1. Introduction

The Yangtze River Delta (YRD) region is one of the most populous and most developed urban agglomerations in China. With rapid economic development and fast industrialization and urbanization, the air pollution in this region has become very serious, and it is characterized by high concentrations of fine particulate matter ($PM_{2.5}$) and ozone (O_3) (Hu et al., 2014; Ma et al., 2019; Tie et al., 2006; Wang et al., 2001; Wang et al., 2014b; Xiao et al., 2011). Wang et al. (2014b) reported that the annual average $PM_{2.5}$ values were 56, 64, 75, and 86 μg/m^3 in Shanghai, Hanghzou, Nanjing, and Hefei in 2013, respectively. The annual average $PM_{2.5}$ concentrations in the YRD significantly decreased by about 30%, while the 90th percentile maximum daily average (MDA8) O_3 concentrations increased by 22% from 2013 to 2017 (Wang et al., 2020b).

A number of studies (Chang et al., 2019; Li et al., 2017; Wang et al., 2017) have pointed out that regional transport plays a crucial role in the formation of $PM_{2.5}$ pollution, accounting for about 30–80% of the total $PM_{2.5}$ concentrations. Streets (Streets et al., 2007) used the Community Multi-scale Air Quality (CMAQ) model to simulate the contributions of the surrounding provinces and cities to Beijing's air pollution during the 2008 Beijing Olympic Games. The results showed that under the effect of a stable southerly wind, the pollution emissions from Hebei Province greatly affected the air quality in Beijing, and transport contributed 50–70% and 20–30% to the concentrations of $PM_{2.5}$ and O_3, respectively. Jie and Li (2014) used the Granger Causality test to explore the characteristics of air quality spillovers among the cities in the Pearl River Delta, and they showed that the emissions in Guangzhou and Foshan had significant impacts on the air quality in Shenzhen and Zhuhai. Due to the proximity of cities, the intensive emissions, and the relatively flat terrain in the YRD region, the mutual transport of air pollution among cities is significant (Cheng et al., 2011). Hu et al. (2018) revealed that the transport of O_3 and its precursors from Wuxi, Suzhou, and Shanghai plays an important role in the high downwind O_3 pollution in Nanjing and the western part of the YRD region. Wang et al. (2020a) found that the local, Zhejiang and Jiangsu emissions account for 53%, 19% and 14% of the non-background MDA8 O_3 in Shanghai in August 2013.

It is necessary to understand the impacts of transport on the local air quality so that regional cooperative prevention and control measures can be designed to effectively reduce $PM_{2.5}$ and O_3 pollution. Several methods have been developed and used to estimate the impacts of regional transport on local air quality. The Air Resources Laboratory's HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT), the Potential Source Contribution Function (PSCF), and the Concentration Weighted Trajectory (CWT) method have been widely used to identify major transport trajectories and high emission regions (Dimitriou et al., 2015; Wang et al., 2010; Zhang et al., 2018; Zong et al., 2018). However, these methods only take the atmospheric dynamics into account and do not involve any chemical reactions. Compared with the above models, the Chemical Transport Model (CTM) is more suitable for the quantitative estimation of the transport contributions because it contains a full description of the physical and chemical atmospheric processes (Li et al., 2014; Wang et al., 2014a).

This study aims to quantitatively estimate the transport among the cities in the YRD region. A source-oriented CMAQ model was used to quantify the inter-city transport of $PM_{2.5}$ and O_3 among 41 cities in the YRD region and to provide insights for effective regional cooperative emission control strategies.

2. Methods

2.1. Model description

We used source-oriented CMAQ model v5.0.2 to track the emissions of the precursors of $PM_{2.5}$ and O_3 from different regions. The source-oriented CMAQ model has been continuously developed in our previous studies (Hu et al., 2017b; Hu et al., 2015; Shi et al., 2017; Wang et al., 2019b; Wang et al., 2018; Zhang et al., 2012), and the details of the algorithms are documented in these studies. Therefore only a brief description is given here. To develop a source-oriented treatment in the CMAQ model, the vanilla version of the CMAQ model was modified by adding additional tagged species and by expanding the chemical reactions. To track the primary particles emitted from different regions, artificial non-reactive tracers were added to represent the total primary mass from each region (Hu et al., 2015), and the tracers were tracked separately through all of the atmospheric processes, including emission, transport, diffusion, and dry and wet deposition. To estimate the contributions of the emissions from different regions to the secondary inorganic $PM_{2.5}$ (i.e., sulfate (SO_4^{2-}), nitrate (NO_3^-), and ammonium (NH_4^+)), the emissions of their precursors (i.e., SO_2, NOx, and NH_3, respectively) in the different regions were tagged, reactive tracers were added, and the chemical mechanism that involved in SO_4^{2-}, NO_3^-, NH_4^+ formation was expanded to track their formation during the chemical transformation (Shi et al., 2017; Zhang et al., 2014). A similar treatment was created for regional source apportionment of the secondary organic aerosols (SOA) in the $PM_{2.5}$ by tracking the emissions of volatile organic compounds (VOCs), including long alkanes, high-yield aromatics, low-yield aromatics, benzene, isoprene, monoterpenes, and sesquiterpenes, from the different regions through all of the atmospheric processes as they reacted to form low-volatility SOA products (Hu et al., 2017a; Hu et al., 2017b; Wang et al., 2018). The contributions of the emissions from different regions to the O_3 level were estimated based on an improved O_3 source apportionment method, which attributes the O_3 formation in each time step to NOx and VOCs based on NOx-limited, VOC-limited, and transition regimes (Wang et al., 2019a). The contributions from the different regions to the NOx and VOCs were estimated using reactive region-tagged tracers, so that the regional contributions to the O_3 levels could be determined (Wang et al., 2019b).

2.2. Model application

The modeling domain covered the entire YRD region. There are 41 cities in the YRD, and they were individually tracked in our study. The cities include Shanghai, 13 cities in Jiangsu Province (i.e., Nanjing, Wuxi, Xuzhou, Changzhou, Suzhou, Nantong, Lianyungang, Huaian, Yancheng, Yangzhou, Zhenjiang, Taizhou, and Suqian), 11 cities in Zhejiang Province (i.e., Hangzhou, Ningbo, Wenzhou, Jiaxing, Huzhou, Shaoxing, Jinhua, Quzhou, Zhoushan, Tai1zhou, and Lishui), and 16 cities in Anhui Province (i.e., Hefei, Wuhu, Bengbu, Huainan, Maanshan, Huaibei, Tongling, Anqing, Huangshan, Chuzhou, Fuyang, Su4zhou, Liuan, Bozhou, Chizhou, and Xuancheng). The modeling domain and the locations of the cities are shown in Fig. 1. The region outside of the YRD in the modeling domain is grouped as Non-YRD in the study. In this study, the central point for the model domain was set at the coordinate (32 N, 118E), and bidirectional nested technology was employed, producing two layers of grids with a horizontal resolution of 36 km and 12 km, respectively. The first layer of the grids has a 36 km resolution with 107*137 grids, covering most areas of Eastern China other countries (including Japan, South Korea, North Korea). The inner domain has a 12-km resolution with 127*202 grids, covering the North China Plain and YRD region. The 12 km domain results were presented here. The anthropogenic emissions are based on two widely-used regional inventories: the Multi-resolution Emission Inventory for China (MEIC, http://www.meicmodel.org) for emissions within China and the Regional Emission inventory in Asia v2.1 (Kurokawa et al., 2013) for emissions outside of China. Biogenic emissions were estimated by Model of Emissions of Gases and Aerosols from Nature (MEGAN) v2.1. For more details, see (Qiao et al., 2015). Biomass burning emissions including both gases and aerosols from the 2018 the Fire INventory from NCAR (FINN), which is based on satellite observations (Wiedinmyer et al., 2011). The WRF model provided the meteorological field for the

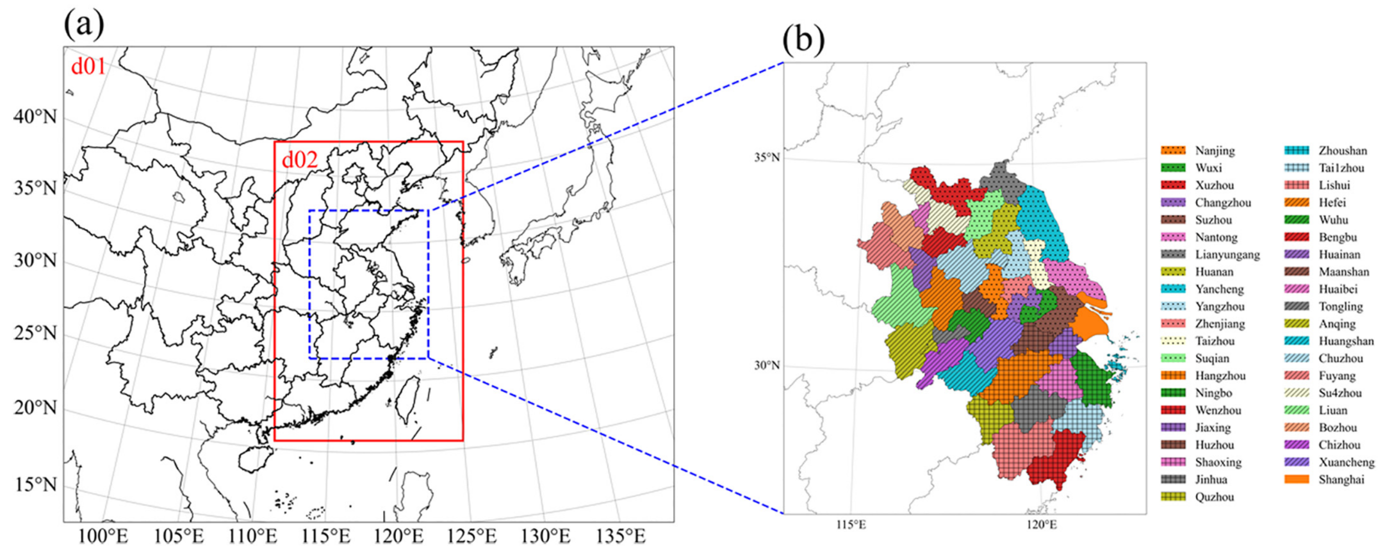


Fig. 1. Modeling domain (a) and the cities in the YRD region (b).

chemical transport model. The meteorological background field and boundary information with a spatial resolution of 1° × 1° and temporal resolution of 6 h were acquired from NCAR (National Center for Atmospheric Research, https://ncar.ucar.edu).

We conducted model simulations for the entire period of the EXPLORE-YRD (EXPeriment on the eLucidation of the atmospheric Oxidation capacity and aerosol foRmation, and their Effects in the Yangtze River Delta) field campaign, which took place from May 17 to June 17, 2018. In our previous studies, we used the same version of the CMAQ model to simulate the air quality (Wang et al., 2021) and conducted a source apportionment study of the $PM_{2.5}$ and O_3 (Li et al., 2021) during this period. The model configurations of the meteorological inputs and the initial and boundary conditions that were used in the previous studies were also used in this study. Model predictions of O_3, $PM_{2.5}$, and its major components were extensively evaluated against measured concentrations during the campaign, and good agreement was generally found between the predictions and measurements, which provides confidence in the transport analysis conducted in this study. Therefore, the details of the model evaluation are not repeated here (model predicted hourly NO_2, SO_2, and O_3 were illustrated in Figs. S1–S3 in the Supplement Materials), and we focus on the results of the inter-city transport of $PM_{2.5}$ and O_3. The contributions of 9 different cities to the $PM_{2.5}$ and O_3 can be solved in one set of simulations, and we conducted 6 sets of simulations to fully obtain the transport matrix for 41 cities in the YRD. The city settings for the 6 sets of simulations are showed in Table S1 in the Supplemental Materials.

3. Results and discussion

3.1. Inter-city transport matrix for $PM_{2.5}$ and O_3

Table 1 shows the transport matrix for $PM_{2.5}$ among the 41 cities. The emissions from the local city itself make the largest contribution in general, which is indicated by the diagonal in the table. The contributions from the other cities in the YRD normally do not exceed 10% individually, except for a few cases, such as Nanjing's contribution in Chuzhou, Hefei's contribution to Huainan, and Ningbo's contribution to Shaoxing. Fig. 2 further summarizes the results and groups the contributions into Local (emissions from the local city), Non-Local (emissions from the other cities within the YRD), and Non-YRD (emissions from the other regions outside of the YRD). During the study period, the emissions in the YRD region (Local + Non-Local) contributed the majority of the $PM_{2.5}$ in all of the cities. The Non-YRD emissions accounted for 24.8% of the $PM_{2.5}$, with a maximum contribution of 49.4% in Shanghai and a minimum contribution of 12.8% in Maanshan. The Non-YRD emissions contributed more than 40% of the $PM_{2.5}$ in four coastal cities, i.e., Shanghai (49.4%), Zhoushan (48.0%), Lishui (45.4%), and Wenzhou (41.5%), due to the dominant southeasterly winds during this period (Wang et al., 2021). The Non-YRD emissions contributed less than 15% in Maanshan (12.8%), Tongling (12.9%), Nanjing (13.2%), Hefei (13.5%), Wuhu (13.6%), Zhenjiang (13.9%), and Huainan (15.0%). These seven cities are all located in the middle of the YRD region.

Among the YRD emissions, the contributions of the Local emissions varied from 8.8% (Su4zhou) to 43.2% (Ningbo), with an average value of 25.3%; and the contributions of the Non-Local emissions varied from 10.5% (Shanghai) to 67.1% (Tongling), with an average of 49.9%. The Local contributions were only greater than the Non-Local contributions in three cities: Shanghai, Wenzhou, and Ningbo. These three cities are located at the east edge of the YRD region. During the study period (May–June 2018), the prevailing directions were southeast to northeast (illustrated in Fig. 4 for Taizhou as an example), resulting that the 'Non-local' emissions were mostly in the downwind areas of the three cities. The Non-Local YRD emissions contributed over 50% of the $PM_{2.5}$ in 23 out of 41 of the cities. These results indicate that transport among cities in the YRD is significant, and multi-city cooperative emission control programs are necessary to reduce the $PM_{2.5}$ in most of the cities in the YRD.

Table 2 shows the transport matrix for O_3 for the 41 cities, and Fig. 3 illustrates the contributions of the Local, Non-Local, and Non-YRD emissions to the O_3 formation in each city. The average Non-YRD contribution of the 41 cities was 19.5%. The Non-YRD contributions exceeded 30% in five cities, i.e., Zhoushan (48.2%), Shanghai (42.6%), Fuyang (34.6%), Wenzhou (33.7%), and Lishui (31.3%), and the Non-YRD contribution was is lower than 10% in seven cities, i.e., Wuxi (6.6%), Zhenjiang (6.8%), Nanjing (7.1%), Changzhou (7.6%), Shaoxing (8.3%), Jinhua (9.7%), and Maanshan (9.8%). The local contributions to O_3 were greater than 40% in Suzhou, Nanjing, Shaoxing, Shanghai, Ningbo, Wuxi, Hangzhou, Nantong, and Wenzhou; while the local contribution was only 9.7% in Su4zhou and 16.0% in Zhoushan. The O_3 values in the cities of Tongling, Maanshan, Su4zhou, Changzhou, and Wuxi were affected by the Non-Local YRD emissions the most, and the Non-Local emissions contributed 64.4%, 63.3%, 62.5%, 61.7%, and 61.1% to these cities, respectively. In total, there were 19 cities with Non-Local contributions of greater than 50%.

3.2. Inter-city contributions of $PM_{2.5}$ and O_3 in Taizhou

We further analyzed the inter-city contributions of $PM_{2.5}$ and O_3 in Taizhou where the EXPLORE-YRD campaign took place. Fig. 4 shows

Table 1
Transport matrix for $PM_{2.5}$ among cities in the Yangtze River Delta region. The numbers indicate the relative contributions (%).

	Nanjing	Wuxi	Xuzhou	Changzhou	Suzhou	Nantong	Lianyungang	Huanan	Yancheng	Yangzhou	Zhenjiang	Taizhou	Suqian	Hangzhou	Ningbo	Wenzhou	Jiaxing	Huzhou	Shaoxing	Jinhua	Quzhou
Nanjing	28.6	4.87	0.4	5.41	5.45	2.1	0.13	0.71	1.17	4.42	6.16	1.87	0.55	1.27	1.59	0.07	2.14	1.36	0.67	0.1	0.02
Wuxi	0.86	27.15	0.13	3.6	22.45	2.87	0.09	0.23	0.52	0.98	1.14	1.62	1.17	1.54	2.7	0.15	3.54	1.85	1.14	0.14	0.02
Xuzhou	1.77	1.32	18.33	0.75	2.41	1.44	2.84	2.21	2.08	1.56	0.91	1.04	0.19	0.44	0.4	0.03	0.42	0.26	0.28	0.03	0
Changzhou	2.39	14.57	0.24	20.66	12.29	2.7	0.12	0.33	0.75	1.91	3.2	2.67	1.15	1.95	2.22	0.13	3.51	2.01	1.18	0.13	0.02
Suzhou	0.45	3.6	0.1	0.99	34.59	3.72	0.08	0.17	0.44	0.55	0.61	0.87	0.89	1.09	3.1	0.2	3.75	1.18	0.83	0.15	0.02
Nantong	0.45	1.94	0.14	0.55	9.73	30.19	0.15	0.2	1.04	0.45	0.44	1.22	0.45	0.5	1.27	0.19	0.66	0.21	0.44	0.12	0.01
Lianyungang	1.77	1.86	1.12	1.22	3.92	4.01	20.84	4	9.27	2.27	1.52	1.82	0.13	0.28	0.42	0.05	0.37	0.16	0.22	0.06	0.01
Huanan	6.41	3.22	0.6	2.46	5.36	5.1	1.18	14.12	7.35	7.23	3.54	3.89	0.23	0.49	0.68	0.07	0.74	0.39	0.33	0.08	0.02
Yancheng	1.19	3.56	0.21	1.85	8.13	10.89	0.7	0.91	18.84	1.86	1.38	3.45	0.42	0.72	0.81	0.1	0.83	0.44	0.44	0.08	0.01
Yangzhou	3.42	5.54	0.24	4.99	8.06	7.04	0.38	1.75	3.96	19.15	7.92	7.96	0.45	0.99	0.66	0.07	1.07	0.68	0.47	0.08	0.01
Zhenjiang	3.9	8.64	0.26	11.53	9.39	3.77	0.15	0.49	1.14	4.19	21.58	4.89	0.66	1.33	1.26	0.08	2.1	1.01	0.71	0.1	0.01
Taizhou	1.01	8.23	0.15	4.37	13	12.42	0.26	0.46	4.1	2.15	3.04	18.33	0.72	0.82	0.96	0.09	1.2	0.53	0.68	0.08	0.01
Suqian	3.89	1.89	1.62	1.21	3.92	3.24	2.22	7.44	4.52	3.32	1.91	2.22	24.49	0.44	0.6	0.05	0.67	0.23	0.31	0.05	0.01
Hangzhou	0.78	0.99	0.21	0.47	2.15	0.61	0.06	0.19	0.22	0.48	0.43	0.29	12.13	33.03	6.15	0.42	2.2	2.12	10.91	2.14	0.41
Ningbo	0.8	1.26	0.17	0.87	2.51	0.82	0.1	0.22	0.3	0.68	0.77	0.47	2.77	1.32	43.15	2.12	2.12	0.32	2.34	0.6	0.07
Wenzhou	0.53	0.63	0.19	0.38	1.57	0.4	0.09	0.15	0.15	0.3	0.31	0.23	0.77	0.65	3.28	34.83	0.58	0.2	0.64	0.39	0.03
Jiaxing	0.84	1.52	0.12	1.17	5.21	0.96	0.07	0.2	0.33	0.81	0.97	0.59	4.72	4.97	12.73	0.5	20.24	1.55	3.49	0.29	0.04
Huzhou	1.53	3.24	0.21	1.37	7.97	1.35	0.08	0.21	0.4	1.09	1.13	0.66	3.99	11	6.66	0.28	10.11	15.58	3.77	0.44	0.07
Shaoxing	0.8	1.08	0.15	0.79	2.42	0.52	0.06	0.18	0.26	0.57	0.78	0.36	0.34	8.2	14.47	2.13	2.12	0.64	37.93	2.75	0.19
Jinhua	1.03	0.83	0.19	0.44	2.64	0.52	0.07	0.2	0.22	0.48	0.47	0.26	9.69	4.45	4.29	2.87	0.91	0.65	9.02	31.16	0.96
Quzhou	0.76	0.86	0.32	0.31	2.2	0.58	0.08	0.32	0.27	0.57	0.35	0.31	5.1	7.23	2.82	0.77	0.68	0.57	4.56	10.84	20.46
Zhoushan	0.82	1.87	0.22	0.78	5.17	1.75	0.21	0.31	0.42	0.99	0.84	0.86	1.11	0.85	7.82	1.63	2.1	0.29	0.98	0.29	0.03
Tai1zhou	0.87	0.94	0.19	0.59	2.08	0.57	0.09	0.19	0.21	0.45	0.53	0.33	1.93	0.98	7.44	6.17	0.84	0.33	1.67	0.92	0.11
Lishui	0.88	0.66	0.34	0.43	1.93	0.43	0.12	0.26	0.23	0.56	0.45	0.32	2.25	1.96	2.73	2.88	0.51	0.45	1.99	3.26	0.82
Hefei	3.06	1.61	0.46	1.15	2.42	0.87	0.1	0.99	0.73	0.87	0.86	0.69	0.36	1.19	0.72	0.03	0.76	0.92	0.48	0.07	0.02
Wuhu	4.17	2.2	0.35	1.53	3.72	1.23	0.07	0.5	0.64	0.79	1.14	0.83	0.74	2.41	1.31	0.04	1.27	1.83	0.89	0.12	0.03
Bengbu	3.98	1.49	1.46	1.07	2.5	0.95	0.49	2.17	1.57	1.77	1.32	0.93	0.29	0.77	0.73	0.04	0.76	0.57	0.46	0.06	0.01
Huainan	1.74	0.85	0.67	0.55	1.49	0.58	0.18	0.97	0.7	0.75	0.51	0.44	0.26	0.83	0.44	0.02	0.5	0.5	0.32	0.05	0.01
Maanshan	11.04	2.75	0.39	2.82	4.08	1.25	0.09	0.63	0.89	1.45	1.74	0.98	0.8	1.68	1.72	0.06	2.03	1.97	0.91	0.11	0.02
Huaibei	1.55	0.93	4.46	0.59	1.71	0.65	1.07	1.44	1.04	0.82	0.57	0.48	0.23	0.64	0.5	0.02	0.43	0.39	0.35	0.04	0
Tongling	1.64	1.85	0.31	0.96	2.79	0.9	0.07	0.44	0.42	0.39	0.72	0.57	0.34	1.36	0.39	0.02	0.41	0.75	0.46	0.1	0.04
Anqing	1.66	1.67	0.41	1.07	1.88	0.8	0.1	0.36	0.34	0.57	0.8	0.54	0.21	1.14	0.23	0.02	0.29	0.48	0.4	0.12	0.06
Huangshan	1.39	1.81	0.61	1.07	2.44	0.93	0.11	0.46	0.44	0.58	0.62	0.59	3.59	5.61	1.86	0.21	0.81	0.84	3.11	1.18	0.3
Chuzhou	11.71	2.82	0.68	2.31	3.88	2.15	0.27	2.39	1.76	4.53	3.21	1.92	0.43	1.02	1.13	0.06	1.39	0.87	0.62	0.09	0.02
Fuyang	0.79	1.02	1.13	0.52	1.56	0.53	0.44	0.63	0.47	0.38	0.33	0.31	0.13	0.55	0.21	0.02	0.3	0.32	0.22	0.03	0.01
Su4zhou	2.86	1.59	8.83	1	2.83	1.34	1.48	3	1.83	2.06	1.28	1.1	0.29	0.66	0.61	0.04	0.61	0.43	0.42	0.05	0.01
Liuan	2.23	1.59	0.59	1.02	2.06	0.89	0.24	0.89	0.65	0.92	0.86	0.68	0.21	0.86	0.28	0.03	0.37	0.47	0.32	0.06	0.02
Bozhou	1.05	0.93	2.45	0.52	1.64	0.52	0.67	0.84	0.63	0.52	0.41	0.35	0.19	0.64	0.33	0.02	0.37	0.39	0.28	0.04	0.01
Chizhou	1.71	1.99	0.51	1.42	3.03	1.24	0.12	0.58	0.51	0.5	0.82	0.78	0.97	2.21	0.59	0.07	0.51	0.74	1.04	0.31	0.09
Xuancheng	3.37	3.49	0.47	2.28	5.81	1.62	0.09	0.39	0.53	1.03	1.11	0.79	2.4	5.94	2.86	0.14	2.55	4.24	2.39	0.43	0.11
Shanghai	0.06	0.06	0.21	0.03	6.14	2.75	0.04	0.03	0.04	0.03	0.03	0.03	0.01	0.03	0.02	0	0.02	0.02	0.02	0	0

Zhoushan	Tai1zhou	Lishui	Hefei	Wuhu	Bengbu	Huainan	Maanshan	Huaibei	Tongling	Anqing	Huangshan	Chuzhou	Fuyang	Su4zhou	Liuan	Bozhou	Chizhou	Xuancheng	Shanghai	Non-YRD
0.12	0.07	0.01	0.73	1.87	0.11	0.14	5.46	0.06	0.47	0.54	0.06	1.86	0.06	0.4	0.1	0.06	0.49	2.24	2.89	13.16
0.23	0.15	0.01	0.19	0.26	0.04	0.05	0.23	0.03	0.09	0.1	0.06	0.17	0.03	0.13	0.04	0.03	0.11	0.56	6.05	17.57
0.02	0.03	0	2.31	0.32	1.64	1.34	0.56	1.66	0.13	0.24	0.01	2.09	0.7	18.33	0.38	0.97	0.12	0.21	1.73	28.48
0.19	0.12	0.02	0.29	0.7	0.05	0.07	0.59	0.03	0.22	0.17	0.07	0.32	0.04	0.24	0.05	0.03	0.24	1.72	4.56	16.18
0.3	0.19	0.02	0.14	0.12	0.03	0.05	0.14	0.03	0.05	0.07	0.04	0.11	0.03	0.09	0.03	0.03	0.06	0.21	12.69	28.2
0.19	0.18	0.02	0.17	0.14	0.04	0.06	0.16	0.03	0.05	0.07	0.02	0.12	0.03	0.13	0.03	0.03	0.06	0.15	12.32	35.66
0.04	0.05	0.01	0.94	0.23	0.37	0.37	0.36	0.2	0.07	0.2	0.01	1.01	0.14	1.12	0.13	0.15	0.07	0.18	3.45	35.56
0.06	0.07	0.01	2.32	0.55	0.24	0.31	1.05	0.1	0.14	0.33	0.02	4.31	0.12	0.6	0.22	0.11	0.12	0.37	3.74	21.74
0.09	0.1	0.01	0.42	0.2	0.11	0.12	0.29	0.07	0.07	0.12	0.01	0.45	0.06	0.2	0.06	0.06	0.07	0.21	6.86	33.57
0.06	0.07	0.01	0.43	0.48	0.08	0.1	0.79	0.05	0.12	0.2	0.02	1.15	0.04	0.24	0.06	0.04	0.12	0.38	4.1	16.57
0.11	0.08	0.01	0.27	0.83	0.05	0.07	1.16	0.04	0.24	0.22	0.03	0.48	0.03	0.26	0.05	0.03	0.21	0.88	3.86	13.89
0.1	0.08	0.01	0.22	0.23	0.05	0.07	0.29	0.04	0.08	0.1	0.01	0.23	0.03	0.15	0.04	0.03	0.07	0.26	5.65	19.64
0.04	0.05	0.01	2.28	0.46	0.83	0.67	0.81	0.15	0.1	0.26	0.01	3.39	0.15	1.62	0.29	0.15	0.08	0.26	2.95	21.17
0.31	0.42	0.17	0.38	0.49	0.1	0.13	0.51	0.07	0.09	0.09	0.38	0.3	0.07	0.21	0.07	0.06	0.09	0.74	2.15	16.81
1.32	2.11	0.27	0.15	0.12	0.04	0.06	0.19	0.04	0.04	0.06	0.06	0.2	0.05	0.16	0.04	0.04	0.05	0.17	3.4	27.67
0.21	6.62	1.13	0.18	0.11	0.06	0.08	0.18	0.07	0.03	0.05	0.02	0.16	0.06	0.18	0.05	0.06	0.03	0.16	2.77	41.49
0.84	0.49	0.07	0.15	0.15	0.03	0.05	0.27	0.03	0.04	0.08	0.04	0.2	0.03	0.11	0.04	0.03	0.06	0.18	6.3	29.53
0.45	0.27	0.05	0.24	0.36	0.07	0.07	0.65	0.04	0.09	0.15	0.14	0.36	0.04	0.21	0.06	0.04	0.12	1.3	3.95	20.22
0.43	2.13	0.31	0.14	0.12	0.05	0.06	0.22	0.04	0.03	0.06	0.06	0.2	0.05	0.15	0.04	0.04	0.04	0.22	2.2	16.66
0.14	2.87	2.27	0.31	0.27	0.1	0.13	0.47	0.07	0.05	0.07	0.11	0.29	0.1	0.18	0.08	0.08	0.05	0.39	1.67	18.91
0.08	0.77	1.4	1.27	0.74	0.25	0.43	0.47	0.13	0.26	0.13	0.71	0.49	0.18	0.32	0.14	0.16	0.15	0.75	1.66	29.55
13.94	1.61	0.21	0.17	0.16	0.03	0.05	0.29	0.04	0.05	0.06	0.03	0.21	0.03	0.21	0.03	0.03	0.05	0.18	5.3	47.98
0.58	32.43	0.73	0.18	0.1	0.06	0.08	0.18	0.06	0.03	0.06	0.05	0.2	0.06	0.18	0.05	0.06	0.04	0.17	3.33	33.92
0.13	2.88	21.42	0.64	0.4	0.2	0.3	0.49	0.16	0.11	0.15	0.12	0.41	0.19	0.34	0.16	0.18	0.12	0.47	2.29	45.36
0.04	0.03	0.01	39.93	3.79	0.7	1.91	4.57	0.17	1.34	2.91	0.07	3.37	0.37	0.46	3.74	0.25	0.78	1.91	1.69	13.54
0.09	0.04	0.01	2.6	25.53	0.27	0.47	8.07	0.17	4.18	1.99	0.27	1.27	0.15	0.35	0.3	0.16	2.87	9.21	2.56	13.62
0.04	0.04	0	7.78	0.72	21.7	5.41	1.9	0.98	0.24	0.49	0.03	11.33	0.88	1.46	1.16	2.01	0.16	0.51	1.67	18.09
0.02	0.02	0	24.57	1.68	3.86	21.51	2.24	0.36	0.4	0.77	0.04	3.8	3.29	0.67	5.48	1.26	0.3	0.85	1.57	14.97
0.12	0.06	0.01	1.51	6.58	0.24	0.26	28.67	0.15	1.07	1.27	0.08	2.34	0.12	0.39	0.2	0.1	0.99	3.24	2.43	12.75
0.03	0.02	0	6.07	0.6	4.55	5.34	1.02	21.41	0.21	0.41	0.02	3.32	2.32	4.46	1.27	6.29	0.21	0.36	1.52	22.67
0.02	0.02	0.01	4.12	8.34	0.31	0.77	1.85	0.12	20.07	15.76	0.21	0.89	0.2	0.31	1.01	0.17	12.53	3.71	1.74	12.88
0.01	0.02	0.01	3.04	2.44	0.41	0.83	1.43	0.2	2.3	35.15	0.18	0.72	0.44	0.41	1.9	0.25	5.91	1.59	1.31	28.3
0.07	0.21	0.1	3.63	1.4	0.48	0.85	0.74	0.28	1.15	0.46	25.09	0.85	0.22	0.61	0.26	0.25	1.61	4.42	1.71	27.04
0.08	0.06	0.01	9.44	1.23	1.32	1.21	3.76	0.22	0.29	0.77	0.04	17	0.3	0.68	0.76	0.21	0.2	0.8	2.25	16.13
0.01	0.02	0	4.17	0.91	1.79	4.41	0.67	1.29	0.33	0.55	0.03	0.83	29.1	1.13	5.5	4.56	0.2	0.45	1	33.16
0.04	0.04	0	4.6	0.57	5.52	2.78	1.08	4.01	0.19	0.36	0.02	4.42	1.24	8.83	0.82	2.03	0.15	0.34	2.07	28.57
0.02	0.02	0.01	8.06	2.06	1.49	4.66	1.5	0.36	0.9	2.47	0.05	1.72	2.02	0.59	27.87	0.63	0.56	1	1.53	27.25
0.02	0.02	0	7.03	0.96	2.84	5.76	0.99	4.25	0.28	0.39	0.03	1.22	9.15	2.45	2.18	21.33	0.18	0.5	1.23	26.38
0.03	0.06	0.03	4.66	4.38	0.48	1.14	1.31	0.21	3.9	4.72	1.78	1.01	0.3	0.51	0.95	0.26	25.61	3.8	1.76	23.36
0.18	0.14	0.05	1.19	2.34	0.26	0.27	1.99	0.17	0.54	0.3	1.32	0.86	0.08	0.47	0.11	0.09	0.58	24.04	3.47	19.48
0	0	0	0.12	0.04	0.04	0.05	0.04	0.09	0.02	0.05	0	0.05	0.07	0.21	0.04	0.07	0.02	0.03	40.14	49.35

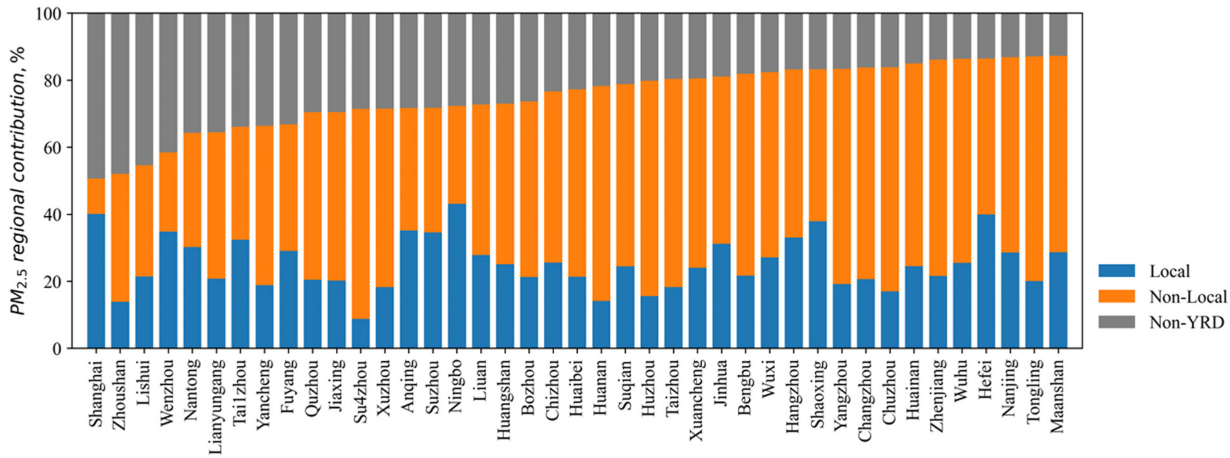


Fig. 2. Contributions of emissions from the local city ('Local'), the other cities in YRD ('Non-Local'), and the other regions outside YRD ('Non-YRD') to $PM_{2.5}$ in 41 cities during the EXPLORE-YRD campaign.

the time series of the inter-city transport contributions to the $PM_{2.5}$ in Taizhou during the study period. On the five days with high $PM_{2.5}$ levels (May 24th, May 30th, June 1st, June 6th, and June 13th), the contributions of Suzhou and Shanghai were relatively large (~20%). Wuxi also significantly contributed on May 30th and Nantong was important on June 1st, contributing about 30%. These cities are located to the southeast of Taizhou. On the pollution days, the wind from the southeast blows pollution from the upwind cities.

Fig. 5 shows the transport characteristics at different times. Throughout the entire study period, the cities in the YRD that made important contributions to the $PM_{2.5}$ in Taizhou were as follows: Taizhou (18%), Suzhou (12%), Nantong (12%), Wuxi (8%), Shanghai (5%), Changzhou (4%), Yancheng (4%), Zhenjiang (3%), and Yangzhou (2%). All of the other cities contributed less than 1% individually. By comparing the transport contributions during the daytime (i.e., 6:00 to 18:00) to those during the nighttime (i.e., 18:00 to 6:00), it was determined that the local contribution was 5% lower during the nighttime. For example, Nantong's contribution was 6% higher and Suzhou's contribution was 4% higher, indicating stronger local contributions of primary emissions during the daytime and more regional contributions during the nighttime. We also analyzed the contributions on $PM_{2.5}$ pollution days (indicated by 'PM25_EPS' in Fig. 5, defined as days with a daily average $PM_{2.5}$ concentration of greater than 75 μg m^{-3}). Suzhou, Wuxi, and Changzhou contributed 3–9% more to the $PM_{2.5}$ in Taizhou on pollution days, while Nantong and Yancheng become much less important compared to their contributions on non-pollution days. Geographically Suzhou, Wuxi, and Changzhou are closer to Taizhou than Nantong and Yancheng. On the pollution days, the meteorological conditions were more stagnant, and therefore, the contributions from these cities became more important.

The contributions to the different $PM_{2.5}$ components during the study period are shown in Fig. 6. The regional contributions to the different components of the $PM_{2.5}$ in Taizhou are substantially different. The Local emissions were the largest source of primary $PM_{2.5}$ in Taizhou, accounting for 26.3%. The largest source of secondary $PM_{2.5}$ was emissions from outside of the YRD region, accounting for 30.4%, with local sources only contributing 7.9%, which is even smaller than those of Nantong and Suzhou. Among the secondary $PM_{2.5}$, the non-YRD emissions contribute about 54.7% of the SO_4^{2-} and 25.5% of the NO_3^-, but only 7.9% of the SOA. The local VOC emissions were the largest source of SOAs in Taizhou, contributing 40.3%. These results indicate that VOC emission controls in local and nearby cities could effectively reduce the SOA concentrations, but reducing the amount of SO_4^{2-} would require SO_2 emission controls over much larger areas.

Li et al. (2021) demonstrated that during the EXPLORE-YRD campaign, the background O_3 (O_3 that directly enters the domain through initial and boundary conditions) contributed more than half of the total O_3 on low O_3 days, and the non-background O_3 (O_3 produced by photochemical reactions) contributed significant amounts on high O_3 days. Fig. 7 shows the time series of the regional contributions to the non-background O_3. The local sources for Taizhou contribute more when the non-background O_3 concentration is higher. In addition to local sources, Nantong and Yancheng also made significant contributions on these days. Similar to the $PM_{2.5}$, the transport contributions to the non-background O_3 were different during the day and at night and on O_3 pollution days (defined as a daily maximum 8 h average O_3 of greater than 160 μg/m^3) vs. non O_3 pollution days (Fig. 8). The local sources in Taizhou contributed more during the daytime, but their contributions remained the same on pollution days and non-pollution days. However, on O_3 pollution days, the contributions from Wuxi, Suzhou, Changzhou, Zhenjiang, and Yangzhou significantly increased. Their total contributions to the O_3 in Taizhou were 20% on non-O_3 pollution days and 39% on O_3 pollution days.

3.3. Transport range and implications for regional emission control

The above described analyses clearly show that multi-city cooperative emission controls are necessary in order to improve the air quality in any of the cities in the YRD. A key question is: What is an appropriate range for the regional emission controls? If the range is too small, then it will not be effective enough to reduce $PM_{2.5}$ or O_3 concentrations, but if the range is too large, it may cause unnecessary economic losses. To answer this question, we calculated the cumulative contributions to the $PM_{2.5}$ and O_3 pollution in a certain city while accounting for the YRD cities within different distances. We repeated the calculation for all 41 YRD cities and conducted fitting using the mean values for the different distances. The fitting uses the form of the Michaelis-Menten equation assuming the cumulative contribution (y) of regional transport within a distance range x as follows:

$$y = K_1 x / (K_2 + x) \quad (1)$$

where K_1 indicates the maximum contribution of regional transport, and K_2 is the distance where the regional transport contribution is half of the maximum contribution. The results are shown in Fig. 9. The fitting R^2 values are over 0.85 for $PM_{2.5}$, O_3, and primary and secondary $PM_{2.5}$, suggesting the assumption that the cumulative contribution of regional transport satisfies the Michaelis-Menten equation is reasonable. For

Table 2
Transport matrix for O_3 among cities in YRD. The numbers indicate the relative contributions (%).

	Nanjing	Wuxi	Xuzhou	Changzhou	Suzhou	Nantong	Lianyungang	Huanan	Yancheng	Yangzhou	Zhenjiang	Taizhou	Suqian	Hangzhou	Ningbo	Wenzhou	Jiaxing	Huzhou	Shaoxing	Jinhua	Quzhou
Nanjing	43.5	3.81	0.1	5.08	3.08	0.5	0.08	0.62	0.45	4.7	6.95	0.89	0.76	1.75	1.14	0.07	1.31	1.53	0.84	0.18	0.06
Wuxi	1.45	43.15	0.02	7.61	18.97	1.14	0.02	0.08	0.13	0.47	1.14	1.69	1.18	1.58	1.58	0.13	3.19	2.98	1.03	0.33	0.05
Xuzhou	1.67	1.15	27.68	0.72	1.58	0.71	1.95	1.83	1.27	1.43	0.87	0.8	0.11	0.22	0.18	0.01	0.27	0.19	0.13	0.02	0.01
Changzhou	5.11	15.27	0.04	30.7	8.53	0.96	0.05	0.15	0.29	1.11	4.81	2.83	1.15	2.23	1.82	0.14	2.78	3.11	1.29	0.37	0.07
Suzhou	0.47	5.89	0.02	1.48	48.48	3	0.02	0.04	0.11	0.3	0.56	0.78	0.93	0.94	1.94	0.25	4.87	1.97	0.76	0.24	0.05
Nantong	0.5	1.91	0.07	0.73	6.92	41.3	0.07	0.09	2.03	0.79	0.68	2.54	0.59	0.69	0.89	0.26	0.35	0.28	0.44	0.17	0.04
Lianyungang	1.43	1.58	3.42	0.88	2.87	2.43	35.13	3.54	7.43	1.29	0.96	1.12	0.13	0.22	0.26	0.07	0.25	0.12	0.16	0.07	0.02
Huanan	4.4	2.37	0.67	1.42	3.7	1.96	1.32	33.73	7.61	6.14	1.96	2.47	0.28	0.62	0.44	0.05	0.51	0.39	0.31	0.11	0.05
Yancheng	1.56	2.56	0.28	1.2	5.26	6.34	0.83	2.27	33.96	2.08	1.16	2.57	0.43	0.62	0.63	0.12	0.54	0.33	0.4	0.17	0.04
Yangzhou	4.62	2.66	0.21	2.04	3.63	3.13	0.23	4.44	6.51	36.26	6.02	9.24	0.47	0.63	0.45	0.06	0.52	0.4	0.39	0.21	0.04
Zhenjiang	8.36	6.65	0.06	10.81	4.79	1.2	0.06	0.23	0.56	4.66	38.78	3.83	0.64	1.14	1.04	0.07	1.16	0.97	0.69	0.24	0.04
Taizhou	1.91	3.97	0.13	2.39	6.74	10.54	0.14	0.89	11.01	5.74	4.43	31.56	0.65	0.62	0.45	0.07	0.47	0.29	0.49	0.2	0.03
Suqian	2.39	1.98	2.53	1.13	3.13	1.43	2.47	9.31	3.04	2.39	1.39	1.47	38.12	0.39	0.33	0.03	0.45	0.24	0.21	0.05	0.02
Hangzhou	0.76	0.74	0.15	0.39	1.21	0.3	0.04	0.16	0.13	0.29	0.28	0.2	9.77	41.97	3.27	0.57	1.5	2.84	8.57	4.24	1.63
Ningbo	0.42	0.76	0.11	0.39	1.63	0.51	0.05	0.06	0.12	0.24	0.27	0.2	5.3	2.81	43.19	4.5	1.94	0.58	4.88	1.52	0.18
Wenzhou	0.52	0.44	0.15	0.26	0.87	0.21	0.06	0.08	0.09	0.18	0.19	0.13	1.26	1.42	3.66	40.93	0.46	0.33	1.17	1.83	0.22
Jiaxing	0.55	1.48	0.03	0.89	5.68	0.54	0.02	0.04	0.14	0.3	0.49	0.25	4.09	7.2	7.69	0.7	32.82	3.19	2.41	0.32	0.06
Huzhou	1.64	3.18	0.07	1.31	8.01	0.52	0.03	0.11	0.17	0.36	0.53	0.39	2.81	12.02	3.32	0.29	8.32	31.65	2.87	1.09	0.26
Shaoxing	0.38	0.7	0.06	0.32	1.13	0.23	0.02	0.05	0.09	0.15	0.21	0.14	0.2	9.71	13.31	4.89	1.21	0.81	43.48	5.69	0.66
Jinhua	0.56	0.51	0.07	0.29	0.91	0.16	0.03	0.07	0.07	0.22	0.22	0.14	10.34	5.87	4.55	3.85	0.52	0.6	9.85	38.81	2.43
Quzhou	0.56	0.42	0.18	0.26	0.79	0.21	0.05	0.2	0.12	0.36	0.29	0.15	5.58	10.63	2.59	0.79	0.47	0.7	5.38	10.81	27.97
Zhoushan	0.42	0.99	0.17	0.39	3.5	1.67	0.12	0.12	0.27	0.41	0.37	0.44	1.77	1.67	7.11	2.7	2.04	0.32	1.58	0.55	0.06
Tai1zhou	0.64	0.67	0.15	0.38	1.12	0.25	0.05	0.08	0.11	0.24	0.28	0.18	3.61	1.89	9.85	5.39	0.73	0.5	3.53	4.39	0.52
Lishui	0.64	0.57	0.16	0.44	0.87	0.19	0.06	0.11	0.1	0.36	0.37	0.23	2.95	3.27	3.28	3.27	0.43	0.54	2.74	8.75	3.23
Hefei	3.29	1.53	0.35	1.03	1.93	0.43	0.16	0.95	0.56	0.6	0.48	0.44	0.48	1.86	0.63	0.04	0.9	1.5	0.55	0.14	0.08
Wuhu	4.74	1.83	0.12	1.53	2.65	0.54	0.07	0.27	0.26	0.77	0.93	0.4	1.07	3.29	0.97	0.06	1.13	3.07	1.09	0.32	0.14
Bengbu	4.21	1.78	1.53	1.22	2.4	0.62	0.45	3.29	1.5	2.16	1.39	0.9	0.29	0.87	0.51	0.03	0.94	1	0.34	0.07	0.01
Huainan	1.89	0.84	0.53	0.54	1.23	0.26	0.2	0.95	0.45	0.44	0.37	0.3	0.25	0.99	0.3	0.02	0.56	0.78	0.28	0.06	0.02
Maanshan	19.13	2.33	0.17	2.86	2.26	0.53	0.12	0.52	0.47	1.47	1.92	0.56	1.14	2.24	1.28	0.06	1.67	2.02	1.06	0.2	0.07
Huaibei	2.26	1.09	3.37	0.78	1.42	0.52	0.52	2.02	0.87	1.54	0.84	0.69	0.2	0.52	0.27	0.01	0.4	0.56	0.21	0.04	0.01
Tongling	1.45	1.5	0.17	0.7	1.92	0.48	0.05	0.23	0.19	0.32	0.37	0.34	0.5	2.37	0.31	0.03	0.36	1.65	0.56	0.2	0.13
Anqing	1.36	1.28	0.34	0.75	1.24	0.46	0.07	0.29	0.22	0.48	0.56	0.37	0.25	1.23	0.19	0.02	0.23	0.62	0.32	0.18	0.15
Huangshan	1.38	1.22	0.28	0.78	1.56	0.45	0.07	0.21	0.22	0.35	0.48	0.32	4.33	10.66	1.72	0.28	0.82	1.48	4.19	2.33	0.93
Chuzhou	12.14	2	0.54	1.45	2.37	0.61	0.3	3.64	1.43	4.31	1.97	1.08	0.54	1.52	0.84	0.05	1.11	1.02	0.63	0.11	0.03
Fuyang	1.25	0.8	0.73	0.58	0.99	0.29	0.2	0.45	0.22	0.37	0.39	0.27	0.12	0.46	0.14	0.01	0.27	0.35	0.14	0.04	0.02
Su4zhou	3.8	2.03	9.67	1.33	2.81	0.95	1.01	3.83	1.84	2.55	1.56	1.3	0.25	0.58	0.41	0.03	0.68	0.66	0.28	0.06	0.01
Liuan	1.72	1.17	0.51	0.78	1.28	0.41	0.14	0.42	0.25	0.5	0.55	0.39	0.15	0.67	0.17	0.02	0.28	0.51	0.2	0.08	0.07
Bozhou	1.68	0.8	1.45	0.58	1.08	0.36	0.32	1.02	0.47	0.77	0.58	0.4	0.15	0.49	0.19	0.01	0.31	0.4	0.17	0.04	0.01
Chizhou	1.54	1.4	0.26	0.99	1.64	0.61	0.07	0.2	0.23	0.39	0.57	0.44	1.65	4.54	0.63	0.09	0.56	1.16	1.63	0.83	0.35
Xuancheng	2.65	2.89	0.17	2.17	3.54	0.58	0.06	0.19	0.24	0.54	0.87	0.47	2.85	9.27	2	0.22	1.96	6.85	2.95	1.31	0.46
Shanghai	0.02	0.01	0.14	0.01	6.4	2.71	0.02	0.02	0.02	0.01	0.01	0.01	0.01	0.01	3.4	0.21	0	0.01	0	0.72	0

Zhoushan	Tai1zhou	Lishui	Hefei	Wuhu	Bengbu	Huainan	Maanshan	Huaibei	Tongling	Anqing	Huangshan	Chuzhou	Fuyang	Su4zhou	Liuan	Bozhou	Chizhou	Xuancheng	Shanghai	Non-YRD
0.03	0.07	0.04	0.68	1.63	0.06	0.13	3.96	0.03	0.34	0.54	0.13	3.09	0.06	0.1	0.19	0.06	0.48	2.33	1.58	7.05
0.09	0.13	0.05	0.18	0.31	0.01	0.03	0.3	0.01	0.08	0.15	0.12	0.11	0.02	0.02	0.09	0.01	0.13	1.09	2.59	6.58
0.01	0.01	0	1.05	0.11	0.82	0.72	0.2	1.4	0.03	0.2	0.01	1.29	0.61	27.68	0.37	0.87	0.03	0.1	0.99	20.69
0.07	0.14	0.05	0.36	1.17	0.02	0.06	1.02	0.01	0.28	0.27	0.18	0.23	0.03	0.04	0.13	0.02	0.39	2.79	2.28	7.63
0.18	0.25	0.05	0.1	0.11	0.01	0.02	0.12	0.01	0.05	0.09	0.07	0.06	0.02	0.02	0.06	0.01	0.08	0.45	9.15	16
0.19	0.26	0.04	0.13	0.13	0.04	0.05	0.13	0.02	0.04	0.05	0.04	0.1	0.02	0.07	0.05	0.02	0.05	0.19	7.95	29.08
0.02	0.07	0.03	0.89	0.12	0.46	0.42	0.16	0.36	0.03	0.27	0.01	1.01	0.2	3.42	0.17	0.25	0.04	0.12	2.59	25.97
0.02	0.05	0.04	2.04	0.38	0.72	0.58	0.65	0.14	0.08	0.63	0.03	5.5	0.21	0.67	0.25	0.19	0.12	0.35	2.78	14.06
0.08	0.12	0.03	0.43	0.14	0.19	0.15	0.21	0.08	0.04	0.15	0.03	0.64	0.08	0.28	0.07	0.08	0.06	0.18	5.38	28.21
0.02	0.06	0.03	0.57	0.2	0.27	0.23	0.38	0.06	0.06	0.24	0.05	2.4	0.08	0.21	0.11	0.07	0.07	0.23	2.31	10.18
0.03	0.07	0.04	0.18	0.99	0.03	0.04	1.63	0.02	0.3	0.29	0.09	0.37	0.03	0.06	0.12	0.02	0.27	0.76	1.87	6.83
0.03	0.07	0.02	0.27	0.17	0.14	0.13	0.24	0.04	0.05	0.12	0.04	0.63	0.05	0.13	0.08	0.04	0.06	0.21	3.08	11.71
0.02	0.03	0.03	1.9	0.28	1.24	0.64	0.35	0.25	0.06	0.42	0.02	2.8	0.26	2.53	0.29	0.29	0.07	0.26	2.09	13.68
0.06	0.57	0.53	0.3	0.79	0.08	0.12	0.47	0.06	0.14	0.23	1.39	0.27	0.11	0.15	0.15	0.09	0.23	2.55	1.34	11.38
1.03	4.5	0.39	0.1	0.09	0.02	0.04	0.09	0.03	0.03	0.09	0.14	0.08	0.03	0.11	0.06	0.04	0.06	0.27	2.26	20.87
0.22	5.9	2.74	0.18	0.13	0.05	0.08	0.15	0.06	0.04	0.11	0.07	0.14	0.06	0.15	0.08	0.07	0.07	0.28	1.24	33.72
0.63	0.7	0.11	0.11	0.12	0.02	0.03	0.13	0.02	0.04	0.11	0.09	0.08	0.03	0.03	0.07	0.02	0.07	0.4	6.07	22.25
0.11	0.29	0.16	0.27	0.63	0.05	0.06	0.75	0.03	0.18	0.37	0.41	0.24	0.07	0.07	0.16	0.04	0.32	4.48	2.34	10.03
0.17	4.89	0.55	0.09	0.07	0.02	0.04	0.09	0.03	0.02	0.09	0.2	0.07	0.04	0.06	0.07	0.04	0.05	0.35	1.39	8.26
0.09	3.85	3.22	0.19	0.2	0.04	0.08	0.24	0.04	0.05	0.12	0.28	0.13	0.07	0.07	0.1	0.07	0.08	0.57	0.77	9.67
0.04	0.79	2.28	0.65	0.7	0.11	0.21	0.25	0.07	0.3	0.25	1.36	0.26	0.16	0.18	0.19	0.13	0.3	1.25	0.68	21.33
15.98	2.7	0.2	0.11	0.08	0.03	0.04	0.1	0.03	0.02	0.05	0.06	0.1	0.02	0.17	0.03	0.03	0.04	0.18	5.09	48.23
0.56	37.4	2.01	0.17	0.12	0.04	0.07	0.12	0.05	0.04	0.13	0.14	0.12	0.06	0.15	0.11	0.06	0.07	0.29	1.62	22.12
0.11	3.27	28.1	0.45	0.33	0.11	0.19	0.26	0.1	0.13	0.28	0.28	0.24	0.14	0.16	0.18	0.16	0.2	0.64	0.85	31.27
0.03	0.04	0.02	37.06	2.61	0.65	2.35	2.78	0.1	0.96	5.86	0.27	5.46	0.37	0.35	3.87	0.22	1.02	1.93	1.12	15.03
0.03	0.06	0.05	3.12	27.27	0.13	0.33	4.92	0.04	4.71	3.76	0.56	0.78	0.11	0.12	0.52	0.07	4.2	11.06	1.26	11.65
0.02	0.03	0.01	3.57	0.32	28.18	4.85	0.82	1.23	0.1	0.56	0.04	11.24	1.12	1.53	1.12	2.09	0.09	0.43	1.61	15.48
0.01	0.02	0.01	19.24	0.72	2.71	29.65	1.07	0.33	0.29	1.18	0.07	4.02	2.9	0.53	5.95	1.7	0.22	0.86	0.8	16.44
0.04	0.06	0.05	2.87	4.59	0.21	0.25	26.92	0.06	0.75	2.08	0.23	4.19	0.11	0.17	0.6	0.09	0.98	2.6	1.27	9.79
0.01	0.01	0.01	1.77	0.37	3.82	2.1	0.62	33.43	0.07	0.26	0.03	2.81	2.04	3.37	0.95	8.29	0.05	0.36	1.01	20.46
0.01	0.03	0.03	3.53	6.24	0.16	0.61	1.09	0.04	18.16	23.08	0.4	0.65	0.12	0.17	0.9	0.08	13.35	4.02	0.97	12.51
0.01	0.02	0.03	3.41	1.76	0.24	0.77	0.59	0.17	1.65	39.66	0.35	0.81	0.59	0.34	4.16	0.27	4.19	2.14	0.8	27.44
0.04	0.28	0.28	2.01	1.46	0.23	0.39	0.52	0.11	1.11	0.75	24.15	0.48	0.22	0.28	0.35	0.15	2.78	6.27	1.44	22.67
0.04	0.05	0.03	6.71	0.7	1.92	1.72	1.68	0.22	0.18	0.94	0.09	30.74	0.35	0.54	1	0.2	0.18	0.75	1.62	12.63
0.01	0.01	0.01	3.28	0.72	1.24	3.64	0.66	0.7	0.22	0.61	0.04	1	34.55	0.73	4.09	4.58	0.13	0.43	0.65	34.61
0.02	0.03	0.01	2.33	0.32	5.35	1.92	0.58	5.06	0.08	0.43	0.03	3.7	1.28	9.67	0.98	2.47	0.07	0.31	1.88	27.88
0.01	0.02	0.02	6.5	1.57	0.61	2.93	1.02	0.23	0.83	5.54	0.1	1.2	2	0.51	34.48	0.51	0.74	1.13	0.78	29
0.01	0.01	0.01	2.69	0.62	2.94	3.55	0.67	3.29	0.17	0.34	0.03	1.48	8.55	1.45	1.3	32.69	0.07	0.41	0.83	27.61
0.02	0.09	0.1	2.54	3.4	0.22	0.64	0.96	0.09	2.51	4.35	3.55	0.49	0.25	0.26	0.92	0.14	28.59	5.4	1.06	24.65
0.06	0.22	0.17	0.71	2.56	0.14	0.12	1.11	0.07	0.68	0.61	2.39	0.44	0.09	0.17	0.23	0.06	1.07	31.96	1.96	12.94
0	0	0	0.02	0.01	0.01	0.02	0	0.04	0	0.01	0	0.01	0.02	0.14	0.01	0.03	0	0.01	43.3	42.63

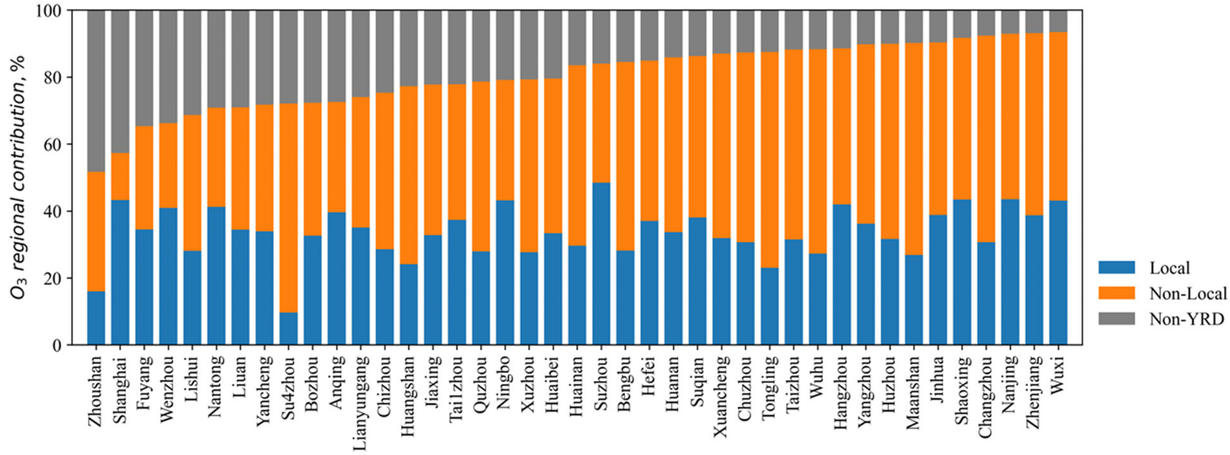


Fig. 3. Contributions of emissions from the local city ('Local'), the other cities in YRD ('Non-Local'), and the other regions outside YRD ('Non-YRD') to non-background O_3 in 41 cities during the EXPLORE-YRD campaign.

$PM_{2.5}$, cumulative contributions of 50%, 60%, and 70% correspond to emissions within distances of 92, 184, and 662 km, respectively. In other words, the regional cooperative emission controls for cities within a distance of at least 184 km are typically needed since these cities dominate the $PM_{2.5}$ contributions. However, the corresponding distances for primary and secondary $PM_{2.5}$ are significantly different. As is shown in Fig. 9(c) and (d), for a cumulative contribution of 50%, the distance is 50 km for primary $PM_{2.5}$ and is 231 km for secondary $PM_{2.5}$. For a cumulative contribution of 60%, the distance becomes 92 km for primary $PM_{2.5}$ and 515 km for secondary $PM_{2.5}$. Therefore, reducing emissions of primary $PM_{2.5}$ in local and the cities within a distance of about 100 km is effective enough to reduce the primary $PM_{2.5}$ concentrations, but reducing the secondary $PM_{2.5}$ in the YRD requires joint emission controls over much larger areas. A cumulative contribution of 60% to

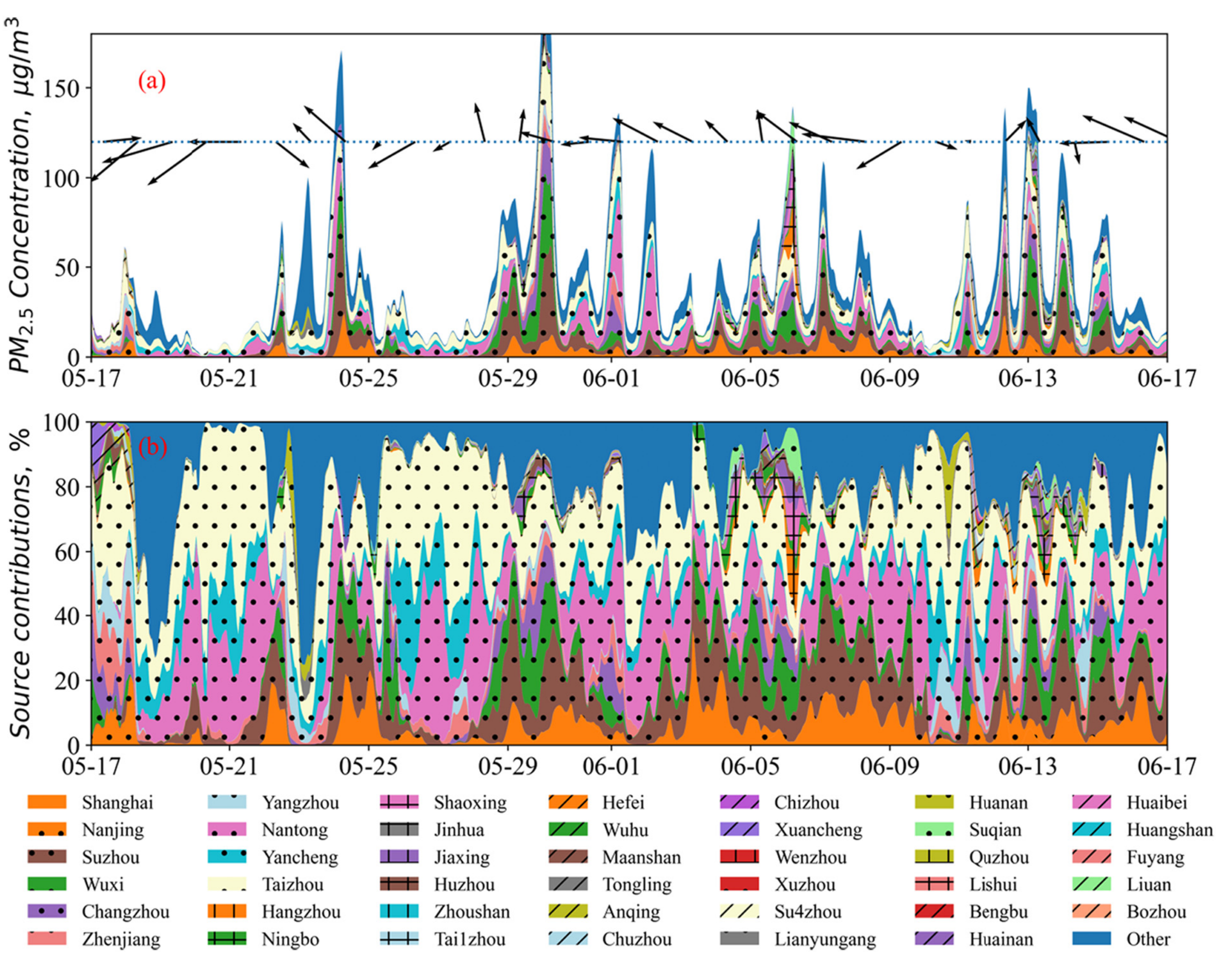


Fig. 4. Hourly contributions of transport from individual cities to $PM_{2.5}$ in Taizhou during the EXPLORE-YRD campaign.

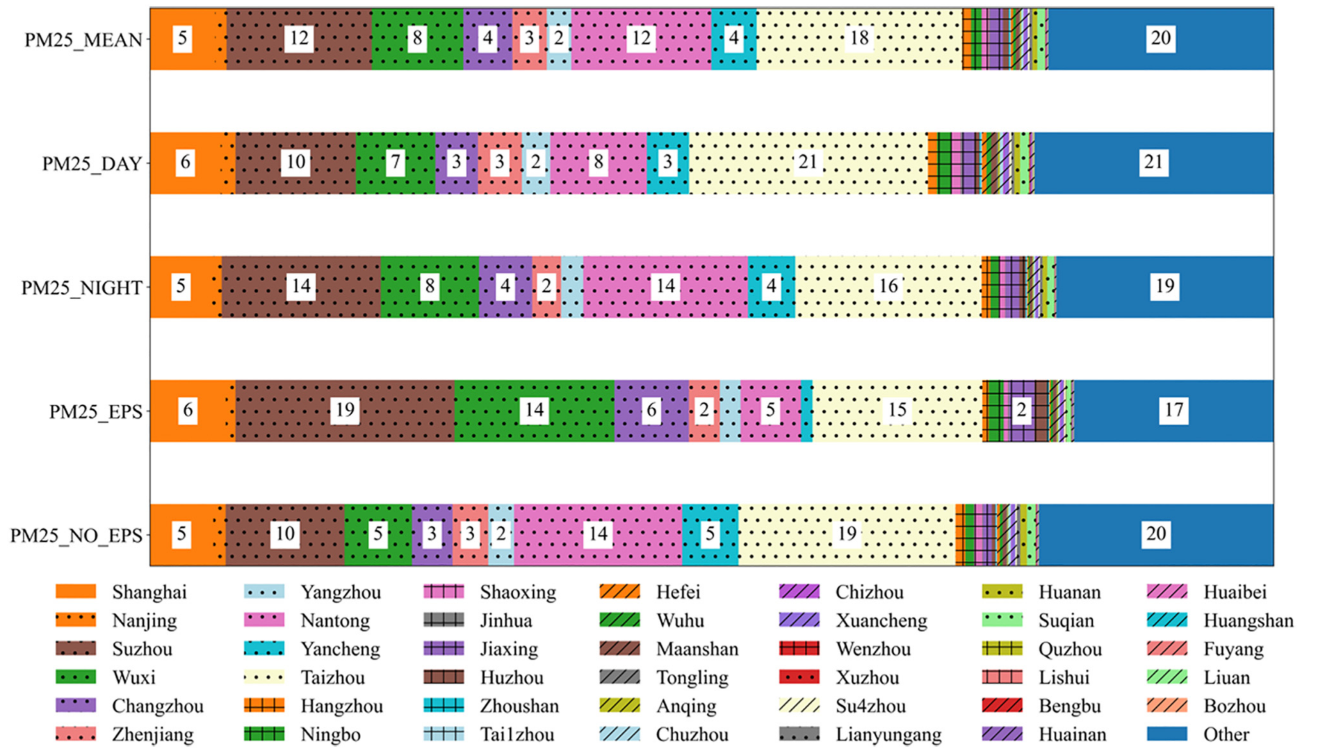


Fig. 5. Average relative contributions of inter-city transport to $PM_{2.5}$ in Taizhou during the entire periods ('PM25_MEAN'), daytime ('PM25_DAY', 6:00 to 18:00), nighttime ('PM25_NIGHT', 18:00 to 6:00), $PM_{2.5}$ pollution days ('PM25_EPS', daily average $PM_{2.5}$ concentration greater than 75 μg m^{-3}), and $PM_{2.5}$ non pollution days ('PM25_NO_EPS').

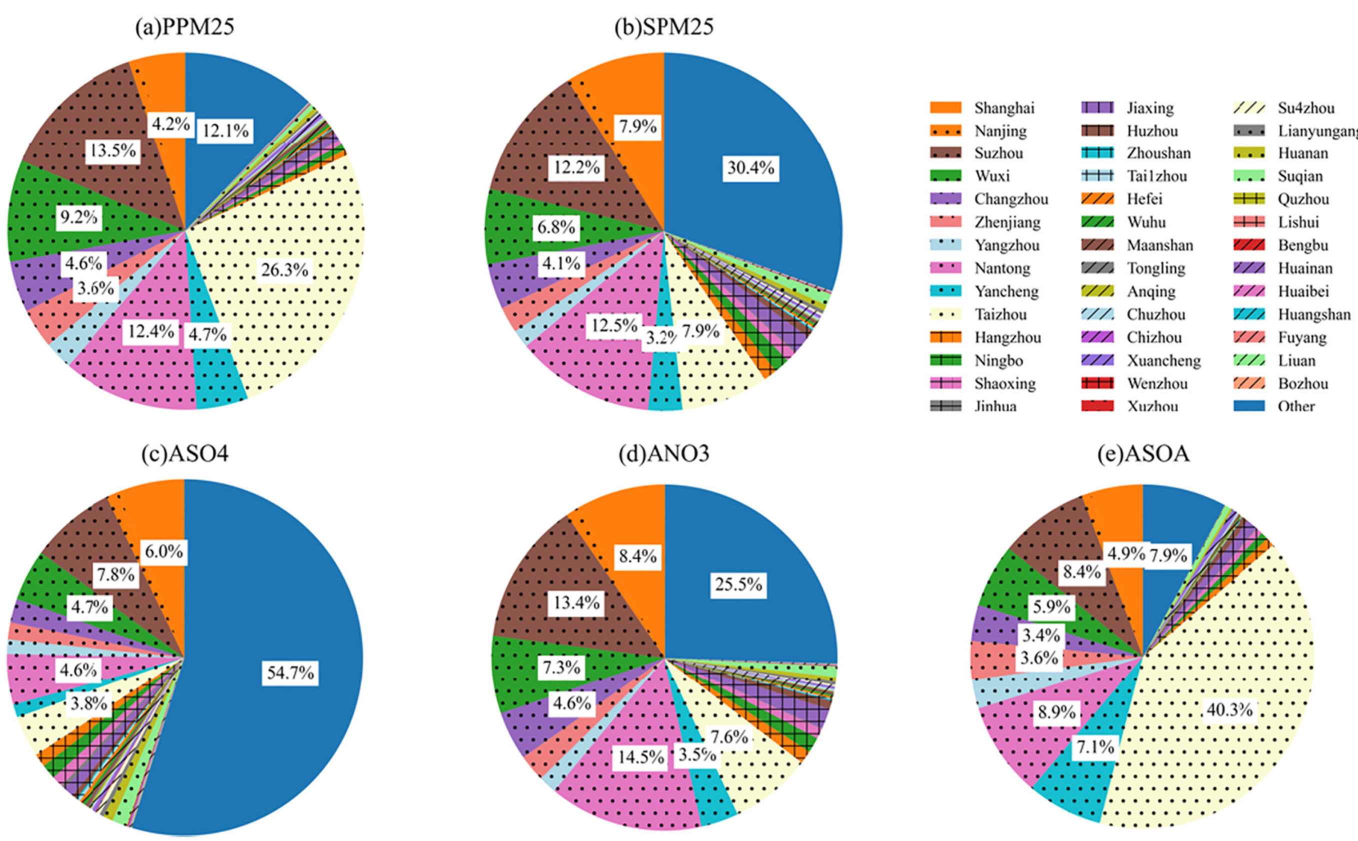


Fig. 6. Relative contributions of Inter-city transport to (a) primary $PM_{2.5}$, (b) secondary $PM_{2.5}$, (c) SO_4^{2-}, (d) nitrate (NO_3^-), and (e) SOA in Taizhou during the EXPLORE-YRD campaign.

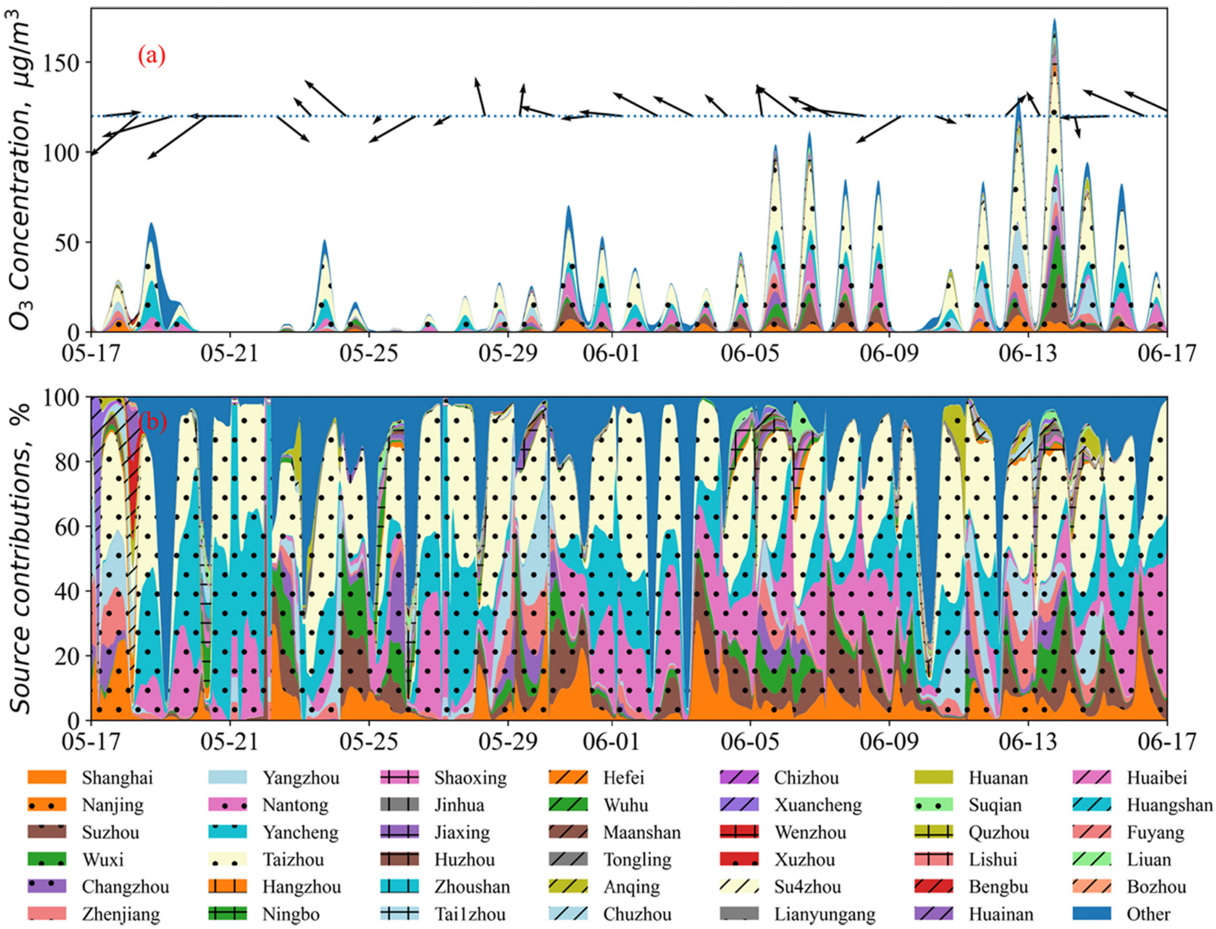


Fig. 7. Hourly contributions of transport from individual cities to non-background O_3 in Taizhou during the EXPLORE-YRD campaign.

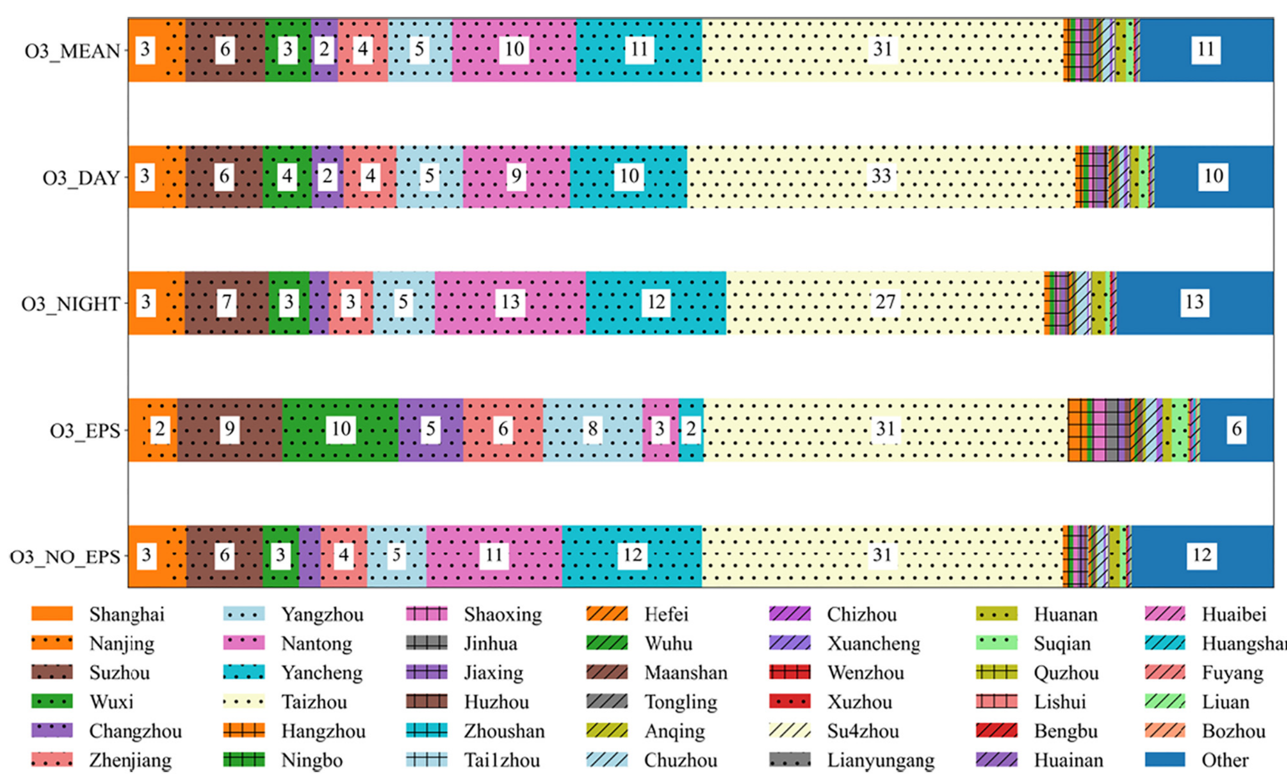


Fig. 8. Average relative contributions of inter-city transport to non-background O_3 in Taizhou during the entire periods ('O3_MEAN'), daytime ('O3_DAY', 6:00 to 18:00), nighttime ('O3_NIGHT', 18:00 to 6:00), O_3 pollution days ('O3_EPS', daily maximum 8 h average O_3 more than 160 $\mu g/m^3$), and O_3 non pollution days ('O3_NO_EPS').

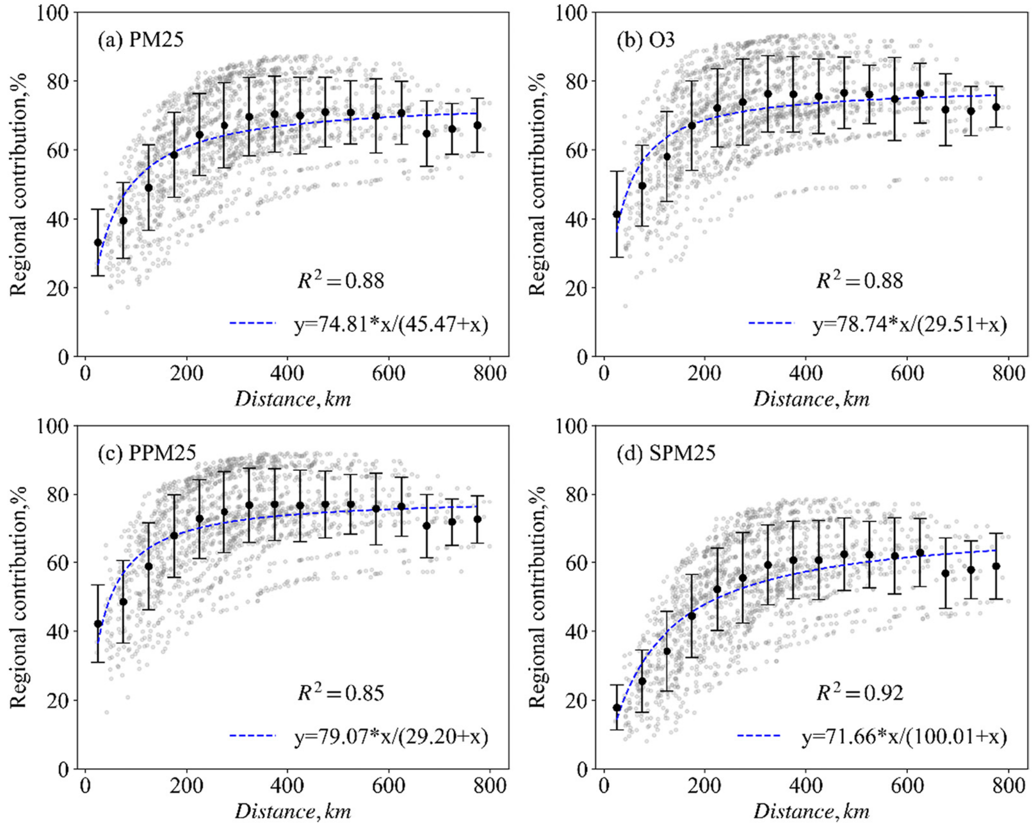


Fig. 9. The accumulative regional contributions as a function of distance in YRD for (a) $PM_{2.5}$, (b) non-background O_3, (c) primary $PM_{2.5}$, and (d) secondary $PM_{2.5}$. The gray dots represent the results of all cities, and the dark dots with error bars represent the mean and standard deviations. The blue dash lines represent the fitted curves.

the O_3 level corresponds to emissions within a distance of 94 km (Fig. 9 (b)). Undoubtedly, the distances estimated in this study are affected by the meteorological conditions and would be different during other time periods. In the future, we will investigate the impacts of inter-city transport on $PM_{2.5}$ and O_3 during different seasons in order to obtain a more general sense of the distance required for regional cooperative $PM_{2.5}$ and O_3 controls in the YRD.

4. Conclusions

The impacts of inter-city transport on $PM_{2.5}$ and O_3 pollution in all of the 41 cities in the YRD region were estimated using a source-oriented CMAQ model during the EXPLORE-YRD campaign. The results show that inter-city transport is very significant in the YRD region. On average, the emissions from the local city, the other YRD cities, and the regions outside of the YRD contributed 25.3% (min-max: 8.8–43.2%), 49.9% (min-max: 10.5–67.1%), and 24.8% (min-max: 12.8–49.4%) to the $PM_{2.5}$, respectively; and they contributed 33.7% (min-max: 9.7–48.5%), 46.8% (min-max: 14.1–64.4%), and 19.5% (min-max: 6.6–48.2%) to the non-background O_3, respectively. In Taizhou, the local contribution to the $PM_{2.5}$ during the daytime was 5% higher than during the nighttime, and the transport contributions from Nantong and Suzhou were 6% and 4% lower, respectively. On $PM_{2.5}$ pollution days, the transport contribution from the cities of Suzhou, Wuxi, and Changzhou became more important (3–9% higher), but the local contribution was 4% lower, and the transport contributions from the farther away cities of Nantong and Yancheng were 5–9% lower. Similar conclusions can be drawn for the O_3 in Taizhou. Therefore, emission controls in local and these nearby cities should be more strongly enforced on pollution days. Local emissions and inter-city transport from other YRD cities dominate the primary $PM_{2.5}$ and SOA, while transport from outside the YRD are important for the secondary inorganic components, e.g., contributing more than half of the SO_4^{2-}. On average, the cities within distances of 184 km and 94 km contributed to 60% of the $PM_{2.5}$ and O_3, respectively. Therefore, we suggest that regional cooperative control programs in the YRD should be considered for emission controls in the cities within these ranges. We also suggest that SO_2 and NO_x emission controls should be enforced on a much larger scale to reduce the secondary inorganic $PM_{2.5}$ in the YRD.

CRediT authorship contribution statement

KG, and JH designed research. KG, LL, and XW conducted the simulations, JL, MQ, QY and JH contributed to model development and configuration. KG, LL, XW, and JH analyzed the data. HL, SG, MH, YZ discussed the results. KG prepared the manuscript and all coauthors helped improve the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

This study was supported by the National Key R&D Program of China (2018YFC0213800), the National Natural Science Foundation of China (41975162, 41675125 and 41705102), and the Jiangsu Environmental Protection Research Project (2016015).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.146619.

References

Chang, X., Wang, S., Zhao, B., Xing, J., Liu, X., Wei, L., et al., 2019. Contributions of inter-city and regional transport to PM2. 5 concentrations in the Beijing-Tianjin-Hebei region and its implications on regional joint air pollution control. Sci. Total Environ. 660, 1191–1200.

Cheng, Z., Chen, C., Huang, C., Huang, H., Li, L., Wang, H., 2011. Trans-boundary primary air pollution between cities in the Yangtze River Delta. Acta Sci. Circumst. 31, 686–694.

Dimitriou, K., Remoundaki, E., Mantas, E., Kassomenos, P., 2015. Spatial distribution of source areas of PM2. 5 by concentration weighted trajectory (CWT) model applied in PM2. 5 concentration and composition data. Atmos. Environ. 116, 138–145.

Hu, J., Wang, Y., Ying, Q., Zhang, H., 2014. Spatial and temporal variability of PM2.5 and PM10 over the North China plain and the Yangtze River Delta, China. Atmos. Environ. 95, 598–609.

Hu, J., Wu, L., Zheng, B., Zhang, Q., He, K., Chang, Q., et al., 2015. Source contributions and regional transport of primary particulate matter in China. Environ. Pollut. 207, 31–42.

Hu, J., Jathar, S., Zhang, H., Ying, Q., Chen, S.H., Cappa, C.D., et al., 2017a. Long-term particulate matter modeling for health effect studies in California – part 2: concentrations and sources of ultrafine organic aerosols. Atmos. Chem. Phys. 17, 5379–5391.

Hu, J., Wang, P., Ying, Q., Zhang, H., Chen, J., Ge, X., et al., 2017b. Modeling biogenic and anthropogenic secondary organic aerosol in China. Atmos. Chem. Phys. 17, 77–92.

Hu, J., Li, Y., Zhao, T., Liu, J., Hu, X.M., Liu, D., et al., 2018. An important mechanism of regional O3 transport for summer smog over the Yangtze River Delta in eastern China. Atmos. Chem. Phys. 18, 16239–16251.

Jie, L., Li, T., 2014. The time-space characteristics and determinants of urban air quality in Pearl River Delta. Urban Insight 11.

Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T., et al., 2013. Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: regional emission inventory in ASia (REAS) version 2. Atmos. Chem. Phys. 13, 11019–11058.

Li, L., Huang, C., Huang, H., Wang, Y., Yan, R., Zhang, G., et al., 2014. An integrated process rate analysis of a regional fine particulate matter episode over Yangtze River Delta in 2010. Atmos. Environ. 91, 60–70.

Li, J., Du, H., Wang, Z., Sun, Y., Yang, W., Li, J., et al., 2017. Rapid formation of a severe regional winter haze episode over a mega-city cluster on the North China Plain. Environ. Pollut. 223, 605–615.

Li, L., Hu, J., Li, J., Gong, K., Wang, X., Ying, Q., et al., 2021. Modelling air quality during the EXPLORE-YRD campaign - Part II. Regional source apportionment of ozone and PM2.5. Atmos. Environ. 247, 118063.

Ma, T., Duan, F., He, K., Qin, Y., Tong, D., Geng, G., et al., 2019. Air pollution characteristics and their relationship with emissions and meteorology in the Yangtze River Delta region during 2014–2016. J. Environ. Sci. 83, 8–20.

Qiao, X., Tang, Y., Hu, J., Zhang, S., Li, J., Kota, S.H., et al., 2015. Modeling dry and wet deposition of sulfate, nitrate, and ammonium ions in Jiuzhaigou National Nature Reserve, China using a source-oriented CMAQ model: part I. Base case model results. Sci. Total Environ. 532, 831–839.

Shi, Z., Li, J., Huang, L., Wang, P., Wu, L., Ying, Q., et al., 2017. Source apportionment of fine particulate matter in China in 2013 using a source-oriented chemical transport model. Sci. Total Environ. 601, 1476–1487.

Streets, D.G., Fu, J.S., Jang, C.J., Hao, J., He, K., Tang, X., et al., 2007. Air quality during the 2008 Beijing Olympic games. Atmos. Environ. 41, 480–492.

Tie, X., Brasseur, G.P., Zhao, C., Granier, C., Massie, S., Qin, Y., et al., 2006. Chemical characterization of air pollution in eastern China and the eastern United States. Atmos. Environ. 40, 2607–2625.

Wang, T., Cheung, V.T., Anson, M., Li, Y., 2001. Ozone and related gaseous pollutants in the boundary layer of eastern China: overview of the recent measurements at a rural site. Geophys. Res. Lett. 28, 2373–2376.

Wang, F., Chen, D., Cheng, S., Li, J., Li, M., Ren, Z., 2010. Identification of regional atmospheric PM10 transport pathways using HYSPLIT, MM5-CMAQ and synoptic pressure pattern analysis. Environ. Model. Softw. 25, 927–934.

Wang, L., Wei, Z., Yang, J., Zhang, Y., Zhang, F., Su, J., et al., 2014a. The 2013 severe haze over southern Hebei, China: model evaluation, source apportionment, and policy implications. Atmos. Chem. Phys. 14, 3151–3173.

Wang, Y., Ying, Q., Hu, J., Zhang, H., 2014b. Spatial and temporal variations of six criteria air pollutants in 31 provincial capital cities in China during 2013–2014. Environ. Int. 73, 413–422.

Wang, Y., Bao, S., Wang, S., Hu, Y., Shi, X., Wang, J., et al., 2017. Local and regional contributions to fine particulate matter in Beijing during heavy haze episodes. Sci. Total Environ. 580, 283–296.

Wang, P., Wang, Q., Zhang, H.L., Hu, J.L., Lin, Y.C., Mao, H.J., 2018. Source apportionment of secondary organic aerosol in China using a regional source-oriented chemical transport model and two emission inventories. Environ. Pollut. 237, 756–766.

Wang, P., Chen, Y., Hu, J.L., Zhang, H.L., Ying, Q., 2019a. Attribution of tropospheric ozone to NOx and VOC emissions: considering ozone formation in the transition regime. Environ. Sci. Technol. 53, 1404–1412.

Wang, P., Chen, Y., Hu, J.L., Zhang, H.L., Ying, Q., 2019b. Source apportionment of summertime ozone in China using a source-oriented chemical transport model. Atmos. Environ. 211, 79–90.

Wang, P., Wang, T., Ying, Q., 2020a. Regional source apportionment of summertime ozone and its precursors in the megacities of Beijing and Shanghai using a source-oriented chemical transport model. Atmos. Environ. 224, 117337.

Wang, Y., Gao, W., Wang, S., Song, T., Gong, Z., Ji, D., et al., 2020b. Contrasting trends of PM2.5 and surface-ozone concentrations in China from 2013 to 2017. Natl. Sci. Rev. 7, 1331–1339.

Wang, X., Li, L., Gong, K., Mao, J., Hu, J., Li, J., et al., 2021. Modelling air quality during the EXPLORE-YRD campaign – part I. model performance evaluation and impacts of meteorological inputs and grid resolutions. Atmos. Environ. 118131.

Wiedinmyer, C., Akagi, S., Yokelson, R.J., Emmons, L., Al-Saadi, J., Orlando, J., et al., 2011. The fire INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning. Geosci. Model Dev. 4, 625–641.

Xiao, Z.M., Zhang, Y.F., Hong, S.M., Bi, X.H., Jiao, L., Feng, Y.C., et al., 2011. Estimation of the Main factors influencing haze, based on a long-term monitoring campaign in Hangzhou, China. Aerosol Air Qual. Res. 11, 873–882.

Zhang, H., Li, J., Ying, Q., Yu, J.Z., Wu, D., Cheng, Y., et al., 2012. Source apportionment of PM2. 5 nitrate and sulfate in China using a source-oriented chemical transport model. Atmos. Environ. 62, 228–242.

Zhang, H., Hu, J., Kleeman, M., Ying, Q., 2014. Source apportionment of sulfate and nitrate particulate matter in the eastern United States and effectiveness of emission control programs. Sci. Total Environ. 490, 171–181.

Zhang, H., Cheng, S., Wang, X., Yao, S., Zhu, F., 2018. Continuous monitoring, compositions analysis and the implication of regional transport for submicron and fine aerosols in Beijing, China. Atmos. Environ. 195, 30–45.

Zong, Z., Wang, X., Tian, C., Chen, Y., Fu, S., Qu, L., et al., 2018. PMF and PSCF based source apportionment of PM2. 5 at a regional background site in North China. Atmos. Res. 203, 207–215.