Simulation-based Design of Regional Emission Control Experiments with Simultaneous Pollution of O₃ and PM_{2.5} in Jinan, China

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ABSTRACT

High O₃ and PM_{2.5} concentrations were frequently observed in Jinan during June 2015 and simultaneously occurred on 8 days, with a maximum 8-hour-averaged O₃ concentration of 255 µg m⁻³ and a maximum daily averaged PM_{2.5} concentration of 111 µg m⁻³. In order to investigate simultaneously controlling these two air pollutants, two simulation-based regional emission control experiments were designed using a nested air quality prediction model system (NAQPMS). One emission control scenario (“Conventional Control”) implemented the strictest control measures in Jinan and surrounding areas and resulted in a 15.7% reduction of O₃ and a 21.3% reduction of PM_{2.5} on days polluted by O₃ and PM_{2.5}, respectively. The other emission control scenario (“Source-tagging Control”), by contrast, used online source-tagging modeling results from NAQPMS to select emission reduction regions based on their source contributions to the O₃ and the PM_{2.5} in Jinan and resulted in a 16.2% reduction of O₃ and a 22.8% reduction of PM_{2.5} on days polluted by O₃ and PM_{2.5}, respectively. Compared to Conventional Control, this scheme produced smaller reductions in emissions from areas with low contributions to the O₃ and PM_{2.5} concentrations in Jinan as well as in the total emissions of primary pollutants (the reduced emissions was only 61% of that needed by Conventional Control), and the area and the population affected by these reductions decreased by 12% and 31%, respectively. However, this study demonstrates that Source-tagging Control is more efficient than Conventional Control in reducing simultaneous pollution by O₃ and PM_{2.5} through regional measures.

Keywords: Emission control; Source-tagging method; Simultaneous pollution; O₃; PM_{2.5}.

INTRODUCTION

Air pollution has become one of the top environmental concerns in China. It has an impact on climate change and strongly affects human health. The annual premature deaths attributable to outdoor air pollution in China ranged from 350,000 to 520,000 between 2004 and 2013 (Ma *et al.*, 2016). Among many pollutants that contribute to air pollution, fine particulate matter (PM_{2.5}) and ozone (O₃) are suggested to be the two major pollutants and main harmful contaminants endangering human health (Guo *et al.*, 2016). The increases of

10 µg m⁻³ in PM_{2.5} have been associated with 0.71% increases in daily non-accidental mortality rates (Zhang *et al.*, 2017). However, these values were considered to be underestimated because their generation excluded the impact of O₃. Anenberg *et al.* (2010) suggested that both anthropogenic PM_{2.5} and O₃ contribute substantially to global premature mortality. Chen *et al.* (2018) pointed out that there is a statistically significant relationship between lung cancer incidence and PM_{2.5} and O₃ pollution.

The co-occurrence of PM_{2.5} and O₃ extremes has become a common problem, especially in summer. Shao *et al.* (2017) found that industrial areas in the Yangtze River Delta presented severe combined pollution consisting of high concentrations of O₃ and PM_{2.5} in summer. Zhang *et al.* (2015) have found that when the concentrations of PM_{2.5} precursors (SO₂, NO_x etc.) were high, NO_x could destroy O₃ at night, causing a negative correlation between O₃ and PM_{2.5}. However, Shi *et*

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al. (2015) reported a positive correlation ($R \approx 0.59$) between $PM_{2.5}$ and O_3 concentrations under the condition of O_3 pollution. Hence, as representative pollutants of complex air pollution, $PM_{2.5}$ and O_3 cannot be considered as separate problems. There is an important chemical coupling relationship between O_3 and $PM_{2.5}$, which is of great significance for understanding the chemical process of controlling the concentration of both (Meng *et al.*, 1997).

Due to the worsening of combined air pollution and its grave harm to human health, the Chinese government has attached great importance to prevention and control of air pollution. Under China's National Action Plan on Air Pollution (NAPAP), the $PM_{2.5}$ concentration has decreased significantly from 2013 to 2017 (Bi *et al.*, 2019). However, the simultaneous control of $PM_{2.5}$ and O_3 pollution is particularly challenging due to their strongly coupled relationship. Research indicates that the decline in $PM_{2.5}$ is believed to be the main reason for the increase in surface O_3 concentrations in the North China Plain (Li *et al.*, 2019). Liao *et al.* (2008) have pointed out that the interdependencies of $PM_{2.5}$ and O_3 responses to emission changes complicate the choice of optimum strategies to solve the air pollution problems. Furthermore, without the effective control of nitrogen oxides (NO_x) and VOCs in industrial areas, unilateral particulate emission reduction will aggravate regional O_3 pollution (Shao *et al.*, 2017).

Many major international events such as the 2008 Olympic Games, the 2014 Asia-Pacific Economic Cooperation summit (APEC) in China, the Shanghai World Expo, the 2015 Grand Military Parade and the 2016 G-20 summit have been held, during which emission control measures were implemented to achieve good air quality (Xing *et al.*, 2011; Liu *et al.*, 2016; Wen *et al.*, 2016; Li *et al.*, 2017b). To reduce air pollution during these events, the conventional emission control measures is focusing on the emission reduction of the host city and its surrounding cities and provinces. And during the Shanghai World Expo, a key emission reduction region with a radius of 300 kilometers was designed with the Expo Park as the center. However, the emission control regions in these control measures are relatively fixed and does not change flexibly according to different pollution characteristics. Here, we investigate the combined pollution episodes in Jinan, a large city in eastern China, in June 2015. As one of the Beijing-Tianjin-Hebei air pollution transmission channel cities, Jinan is experiencing serious combined air pollution (Sui *et al.*, 2019). In this study, based on quantifiable results for $PM_{2.5}$ and O_3 by source-tagging modeling, we investigate the emission reduction schemes for effectively controlling O_3 and $PM_{2.5}$ under combined pollution. Accordingly, effective suggestions for emission control under urban $PM_{2.5}$ and O_3 pollutions are proposed.

METHODS

Model Description and Setup

The nested air quality prediction model system (NAQPMS) is a three-dimensional multiscale chemical transport model developed by the Institute of Atmospheric Physics, Chinese Academy of Sciences. This model is widely used in the

simulation of O_3 and $PM_{2.5}$ pollution and has achieved good results (Li *et al.*, 2008; Wang *et al.*, 2014; Wang *et al.*, 2016). It is an online access model and has shown good performance for simulating various pollutants and aerosol species in MICS-ASIA III project (Chen *et al.*, 2018). In China, the model has been successfully applied to the air quality assurance of Beijing Olympic Games, Shanghai World Expo, 2014 Asia-Pacific Economic Cooperation (APEC) summit and other major events. The NAQPMS simulates the physical and chemical processes of air pollutants by solving the mass balance equation with the terrain-following coordinates. Carbon bond mechanism Z (CBM-Z) (Zaveri and Peters, 1999), which consists of 133 reactions for 53 species, is applied in NAQPMS to calculate gas chemistry processes. It includes a detailed description of tropospheric O_3 - NO_x -hydrocarbon chemistry. For aerosol chemistry, an aerosol thermodynamic model (ISORROPIA 1.7) is used to calculate the composition of an inorganic aerosol system (Nenes *et al.*, 1998). Based on Odum *et al.* (1997) and Pandis *et al.* (1991), the reaction rates and methods for 2 anthropogenic precursors and 4 biogenic precursors was calculated to form 6 secondary organic aerosols (SOAs). The RADM mechanism (Chang *et al.*, 1987) is used in wet deposition processes and aqueous-phase chemistry modules. The advection scheme employs a simplified but accurate mass-conserving, peak-preserving, mixing ratio-bounded advection algorithm (Walcek and Aleksic, 1998). The dry deposition module is derived from the scheme of Wesely (1989).

The model is configured with three nested domains (shown in Fig. 1). The first domain covers most of China with a $27\text{ km} \times 27\text{ km}$ horizontal resolution on 106×106 grids. The second domain includes eastern and northern China with a $9\text{ km} \times 9\text{ km}$ horizontal resolution on 163×151 grids. The third domain covers Shandong Province and its surrounding provinces with a $3\text{ km} \times 3\text{ km}$ horizontal resolution on 271×241 grids. The simulation period of air pollutants occurs from May 25, 2015, to June 30, 2015, with a spin-up period during the first 7 days.

The Weather Research and Forecasting Model (WRF) Version 3.6 is used to provide hourly meteorological data for air pollutant simulations of NAQPMS. The model domains and horizontal resolutions are the same as in NAQPMS. For WRF modeling, the piecewise integration was carried out in the long-term simulation to improve the simulation accuracy, that is, each simulation lasted for 48 hours and the first 24 hours of simulation was considered as the spin-up period. The simulated data of the second day were used as the input for NAQPMS. Then all the 1-day's meteorological inputs were connected to derive a continuous simulation of NAQPMS. The input meteorological data used for WRF is provided by the Final Operational Global Analysis Data (FNL) with a resolution of $1^\circ \times 1^\circ$ from the National Centers for Environmental Prediction (NCEP).

Emission Inventory

The anthropogenic emission inventory for NAQPMS is provided by Hemispheric Transport of Air Pollution (HTAP) Version 2.2 (Janssens-Maenhout *et al.*, 2015) with $0.25^\circ \times 0.25^\circ$ resolution. The HTAP dataset is a bottom-up database

Fig. 1. Configuration of three model domains. The locations of 88 cities in tagged emission source regions are presented: 1) Beijing, 2) Tianjin, 3) Zhangjiakou, 4) Chengde, 5) Qinhuangdao, 6) Tangshan, 7) Langfang, 8) Baoding, 9) Cangzhou, 10) Shijiazhuang, 11) Hengshui, 12) Xingtai, 13) Handan, 14) Liaocheng, 15) Dezhou, 16) Jinan, 17) Laiwu, 18) Zibo, 19) Binzhou, 20) Dongying, 21) Yantai, 22) Weihai, 23) Qingdao, 24) Weifang, 25) Rizhao, 26) Linyi, 27) Taian, 28) Jining, 29) Zaozhuang, 30) Heze, 31) Datong, 32) Shuozhou, 33) Xinzhou, 34) Taiyuan, 35) Lvliang, 36) Yangquan, 37) Jinzhong, 38) Linfen, 39) Changzhi, 40) Yuncheng, 41) Jincheng, 42) Anyang, 43) Puyang, 44) Hebi, 45) Xinxiang, 46) Jiaozuo, 47) Jiyuan, 48) Sanmenxia, 49) Luoyang, 50) Zhengzhou, 51) Kaifeng, 52) Shangqiu, 53) Xuchang, 54) Pingdingshan, 55) Nanyang, 56) Luohe, 57) Zhoukou, 58) Zhumadian, 59) Xinyang, 60) Suzhou, 61) Huaibei, 62) Bozhou, 63) Fuyang, 64) Huainan, 65) Bengbu, 66) Chuzhou, 67) LuAn, 68) Hefei, 69) MaAnshan, 70) Wuhu, 71) Tongling, 72) Anqing, 73) Chizhou, 74) Huangshan, 75) Xuancheng, 76) Lianyun, 77) Xuzhou, 78) Suqian, 79) Yancheng, 80) HuaiAn, 81) Taizhou, 82) Yangzhou, 83) Zhenjiang, 84) Nanjing, 85) Changzhou, 86) Nantong, 87) Wuxi, and 88) Suzhou.

that includes 7 main categories of human activity emission (power, industry, residential, agriculture, ground transport, aviation and shipping) for the year of 2010. The anthropogenic emissions in China from HTAP is provided by MIX (Li *et al.*, 2017a). It has been widely adopted by various air quality models and achieved good simulation results (Yu *et al.*, 2017; Li *et al.*, 2018). The Global Fire Emissions Database (GFED) Version 4 (Guido *et al.*, 2014) is employed for emissions from biomass burning. The biogenic emissions are based on the dataset MEGAN-MACC created by the Model of Emissions of Gases and Aerosols from Nature (Sindelarova *et al.*, 2014). VOC emissions from the ocean are obtained from Precursors of Ozone and their Effects in the Troposphere (POET) database (Granier *et al.*, 2005). NO_x emissions from soils are based on Regional Emission Inventory in Asia (REAS) (Yan *et al.*, 2003). The Global Emissions Inventory Activity (GEIA) (Price *et al.*, 1997) is employed for lightning NO_x emissions.

In order to reduce the uncertainty induced by the changes of emission between 2010 and 2015, emission adjustments have been made in the simulations. We employed the inverse method developed by Tang *et al.* (2013) and surface observations to update the regional emission inventory for CO , SO_2 and NO_x . Table S1 shows the total emissions of CO , SO_2 and NO_x in June 2010 over Jinan in the original and inversed emission inventory.

Measurement Data

The meteorological data provided by the China Meteorological Administration (including hourly surface temperature, relative humidity, wind direction and wind speed) at Jinan (located at 116.98°E and 36.68°N), Beijing (located at 116.28°E and 39.93°N), Tianjin (located at 117.07°E and 39.08°N) and Taiyuan (located at 112.55°E and 37.78°N) are used for WRF model validation and analysis of the meteorological conditions during pollution processes. The air pollutant observation data are provided by the China National Environmental Monitoring Center. The locations of the sites in Jinan, Beijing, Tianjin and Taiyuan are given in Table S2. The observation data used in this study in Jinan, Beijing, Tianjin and Taiyuan are the average of all sites in each city. The observations, including hourly $\text{PM}_{2.5}$, PM_{10} , O_3 , SO_2 , NO_2 and CO concentrations, are used for NAQPMS model validation and analyzing the characteristics of the pollution processes.

Source Apportionment Method

Studies have shown that regional transport has always been considered the major source of severe air pollutions, especially for $\text{PM}_{2.5}$ (Li *et al.*, 2015; Yang *et al.*, 2019). The online pollutant source-tagging module implemented in the NAQPMS modeling system (Li *et al.*, 2008; Wu *et al.*, 2017) is employed to quantify the contributions of the air

pollutant concentration from predefined pollution source regions. This method has been used in many previous studies to assess the impact of regional transport on PM_{2.5}, O₃ and other pollutants (Lu *et al.*, 2017; Shen *et al.*, 2017). Pollutants are tagged by their geographical sources, and the contribution of each defined source region is strictly positive in the module. The regional contribution of primary and secondary aerosols is calculated separately in emissions and physical (e.g., advection, diffusion and convection) and chemical processes. The method suffered a lower error compared to the traditional sensitivity method, with the turning on/off of the emissions of the source-tagged regions due to the high nonlinearity of the chemical transport model. In this method, different tagged species are assumed to be properly integrated in each grid, and each tagged species is assumed to share the same loss coefficients during the processes of outflow, dry and wet deposition and chemical destruction (Davis *et al.*, 2003; Kondo *et al.*, 2004). The secondary aerosols were simplified by connecting them directly to their corresponding precursors. Therefore, the fraction of each species (F_s) was calculated as follows:

$$\left(\frac{dF_s}{dt}\right)_i = (E_{r,i})_{\text{emis}^r} + (P_{r,i})_{\text{adv+conv+diff}} + (C_{r,i})_{\text{chem}} \quad (1)$$

where i is the index of the model cell and r is the tagged region, and $(E_{r,i})_{\text{emis}^r}$ and $(C_{r,i})_{\text{chem}}$ represent the production during the emission process and the chemical reactions of the tagged regions (r) in the i^{th} grid cell, respectively. If the i^{th} grid is not included in the tagged emission region, the emission is equal to zero. $(P_{r,i})_{\text{adv+conv+diff}}$ is the flow flux of the species from the tagged region (r) originating from advection, convention and diffusion and S_i represents the total concentrations of the species in the i^{th} grid. The detailed description of the algorithms can be referred to Wu *et al.* (2017).

According to the geographical location of Jinan, 90 predefined regional sources were tagged in this simulation study. As is shown in Fig. 1, the predefined regional sources include Beijing, Tianjin and all the cities in Shandong, Shanxi, Henan, Hebei, Jiangsu and Anhui Provinces. The other two predefined regional sources are other areas in China (“OIC”) and other areas outside China (“Others”).

Numerical Emission Control Experiments

A series of simulation-based regional emission control experiments was developed to investigate the effective emission control scheme under simultaneous pollutions of urban PM_{2.5} and O₃. The experiments were designed according to the following steps:

- (1) Based on the source-tagging modeling results, the average contribution ratio of each tagged city to O₃/PM_{2.5} concentrations on O₃/PM_{2.5} pollution days of Jinan were calculated respectively. Cities whose average contribution ratio to O₃ or PM_{2.5} in Jinan was more than 1% were chosen as the emission control region in the source-tagging emission control scenario (named “Source-tagging Control”). According to the average contribution ratios to

O₃ or PM_{2.5}, the cities in Source-tagging Control are divided into Response Levels I, II and III. The criteria are shown in Table 1.

- (2) Following the conventional regional emission control measures, with Jinan as the center, the region that has the corresponding area size of the Source-tagging Control region is chosen as the emission control region in the conventional emission control scenario (named “Conventional Control”). And all the cities in Conventional Control are considered as Response Level I.
- (3) The emission reductions were mainly carried out for the primary pollutant BC, OC, PM_{2.5}, NO_x, SO₂ and VOCs. Response Level I corresponds to the strictest emission reduction, Response Level II corresponds to the smaller reduction percentages, and Response Level III corresponds to the smallest reduction percentages.

In addition, compared with the pollutant concentrations in the base scenario without considering any emission control, the decrease of pollutant concentrations in an emission control scheme is defined as the reduction ratio to evaluate the effect of different emission control schemes. The reduction ratio of PM_{2.5} ($RR_{\text{PM}_{2.5}}$) and O₃ (RR_{O_3}) are calculated as follows:

$$RR_{\text{PM}_{2.5}} = \left(\frac{\text{PM}_{2.5_base} - \text{PM}_{2.5_s}}{\text{PM}_{2.5_base}} \right) \times 100\% \quad (2)$$

$$RR_{\text{O}_3} = \left(\frac{\text{O}_{3_base} - \text{O}_{3_s}}{\text{O}_{3_base}} \right) \times 100\% \quad (3)$$

where $\text{PM}_{2.5_base}$ and $\text{PM}_{2.5_s}$ represent the PM_{2.5} daily average concentration in the base scenario and an emission control scenario, and O_{3_base} and O_{3_s} are the O₃ daily maximum 8-hour-averaged concentration (MDA8h O₃) in the base scenario and one emission control scenario.

In this paper, the PM_{2.5} and O₃ pollution days are defined as the PM_{2.5} daily average concentration being greater than 75 $\mu\text{g m}^{-3}$ and the MDA8h O₃ being greater than 160 $\mu\text{g m}^{-3}$, respectively.

RESULTS AND DISCUSSION

Validation of the Simulation Results

The observation data of the main meteorological factors (surface temperature, relative humidity and wind direction and speed) and pollutants (PM_{2.5} and O₃) in Jinan, Beijing, Tianjin and Taiyuan were used for comparison with the simulated results. To evaluate the simulated results, some statistical parameters were used for verification and analysis in this paper. The formulas of the statistical parameters are shown in the supplementary material.

Fig. 2, Fig. S1 and Table S3 show the comparison and statistical parameters of the simulated results and observation data of the main meteorological factors in Jinan, Beijing, Tianjin and Taiyuan in June 2015. The WRF model can be observed to better reproduce the variation of the 4 meteorological factors in the simulation period and not only

Table 1. The classification criteria for different response levels.

The average contribution ratio to O ₃ or PM _{2.5}	Response level
≥ 10%	I
≥ 2% and < 10%	II
≥ 1% and < 2%	III

Fig. 2. Comparison of hourly variation of simulated and observed surface temperature (T), relative humidity (RH), wind and wind speed (WS) at Jinan in June 2015. The simulated results are from the simulation with 3 km × 3 km horizontal resolution.

reflects the changing trend of meteorological factors but also represents the observation values accurately. The simulation of the surface temperature works well with the correlation coefficient (r) of 0.86–0.89 and the mean bias (MB) of 0.1–2.0°C. The correlation coefficient of relative humidity is 0.79–0.88 except for a slight underestimation in Beijing. Compared with temperature and relative humidity, the simulation of wind speed is slightly worse, with the correlation coefficient of 0.39–0.70 and the MB of 0.4–1.8 m s⁻¹. For wind direction, the benchmark suggested by Emery *et al.* (2001) is the MB ≤ ±10°. For wind direction in June 2015, except the relatively large bias in Beijing (MB = 21.6°), the simulation of the wind direction works well with MB of 0.4° in Jinan, 3.7° in Tianjin and 7.5° in Taiyuan.

Fig. 3, Fig. S2 and Table S4 show the comparison and statistical parameters between the simulated results and observation data of PM_{2.5} and O₃ in Jinan, Beijing, Tianjin and Taiyuan in June 2015. The model shows a good simulation capability for PM_{2.5} and O₃ with correlation coefficients of 0.43–0.75 and 0.55–0.74, respectively, making it basically able to reproduce the variation of the characteristics of pollutants. The simulated PM_{2.5} are consistent with the observations with the MB within ±15 µg m⁻³. Compared with the observations, the simulated results of O₃ concentrations are lower, especially at night. But the biases are within a reasonable range. On the whole, NAQPMS has good simulation ability for the concentration range and

variation trend of PM_{2.5} and O₃, which provides a good data basis for analyzing the spatiotemporal variation and regional transport process of PM_{2.5} and O₃ in the study period.

Characteristics of the Combined Pollution Episode

Fig. 4 shows the variation of PM_{2.5} and O₃ concentrations and meteorological factors over Jinan in June. It can be found from the variation of PM_{2.5} daily average concentration and MDA8h O₃ in Fig. 4(a) that Jinan experienced severe air pollution in June 2015 with 15 O₃ pollution days and 9 PM_{2.5} pollution days. And there were 8 days when simultaneous pollutions of O₃ and PM_{2.5} occurred (on June 7, 10, 16, 17, 22, 26, 27 and 28). It indicated that almost all PM_{2.5} pollutions were accompanied by O₃ pollutions.

As shown in Fig. 4(a), the ozone concentration exceeded 160 µg m⁻³ slightly on June 1, and then the two pollutant concentrations continued to decline until June 5. Starting from June 5, the concentration of pollutants gradually increased, and the PM_{2.5} and O₃ combined pollution occurred on June 7, with the PM_{2.5} daily average concentration and MDA8h O₃ of 82 µg m⁻³ and 233 µg m⁻³, respectively. According to the change of wind field, the concentrations of pollutants increased gradually with the high wind speed in the early stage. The temperature was higher and the relative humidity was lower under the influence of southward or southwest airflow. On June 7, the pollutant concentrations reached the maximum, with the decrease in wind speed and

Fig. 3. Comparison of hourly variation of simulated and observed $\text{PM}_{2.5}$ and O_3 concentrations at Jinan in June 2015. The simulated results are from the simulation with $3 \text{ km} \times 3 \text{ km}$ horizontal resolution.

Fig. 4. Time series of observed pollutant concentrations and major meteorological parameters: (a) the $\text{PM}_{2.5}$ daily average concentration and the O_3 daily maximum 8-hour-averaged concentration (MDA8h O_3), (b) hourly $\text{PM}_{2.5}$ concentration and wind speed, (c) hourly O_3 concentration and wind speed, (d) hourly wind, and (e) hourly surface temperature (T) and relative humidity (RH).

change in wind direction. As seen from the hourly variation of pollutant concentrations and wind field, when the wind direction changed, that is, from southerly wind to a weak northerly wind, the concentrations of $\text{PM}_{2.5}$ and O_3 both rose sharply. The sea-level synoptic pressure patterns over central and eastern China on June 6 and 7 are displayed in Fig. S3. It can be seen that on June 7, there existed a weak high-pressure system over northern China, which put Jinan between the high-pressure and low-pressure systems. Jinan and surrounding areas were mainly dominated by weaker northeasterly winds. And the south of Jinan was dominated by southerly wind. Such weather conditions might result in the accumulation of pollutants in Jinan. The north wind increased on June 8, resulting in a decrease in the pollutant concentrations. Then, Jinan was affected by strong south

wind after the wind direction changed again, and the pollutant concentrations again rose. The O_3 concentrations markedly exceeded the standard on June 9 and 10 with MDA8h O_3 of $207 \mu\text{g m}^{-3}$ and $217 \mu\text{g m}^{-3}$, respectively. Moreover, $\text{PM}_{2.5}$ pollution occurred on the 10th. On June 11, Jinan was dominated by the strong northwesterly wind which effectively removed the pollutants.

Starting from June 14, the concentrations of O_3 and $\text{PM}_{2.5}$ began to rise gradually. There were 6 days (June 16, 17, 22, 26, 27 and 28) when co-pollution occurred. The peak of the MDA8h O_3 was $255 \mu\text{g m}^{-3}$ and the peak of $\text{PM}_{2.5}$ daily average concentration was $111 \mu\text{g m}^{-3}$, which both occurred on June 16. From June 14 to 30, Jinan was affected by low wind speed ($\leq 2 \text{ m s}^{-1}$) continuously throughout the pollution episode, especially in the period of high pollutant

concentrations with a mostly westerly or southwest wind. When the pollutant concentrations dropped to low values, easterly wind or southeast wind was the dominant wind over Jinan. The clean air mass with a lower temperature and higher humidity from the ocean played a positive role in the removal of pollutants.

Source Contribution Analysis of O_3 and $PM_{2.5}$

The contributions to O_3 and $PM_{2.5}$ from various regions can be quantified by the online pollutant source-tagging module implemented in the NAQPMS. The contributions from the surrounding regions to $O_3/PM_{2.5}$ concentrations on $O_3/PM_{2.5}$ pollution days in Jinan are shown in Fig. 5. For O_3 , the contributions from different regions vary greatly over time. The local contribution ratio of Jinan is between 6.0% and 56.0%. The following source region with a great contribution is the other areas of Shandong (OSD) with the contribution ratio between 20.8% and 40.9%. Among the neighboring provinces, Jiangsu and Anhui are 2 important sources of O_3 with the average contribution ratio of 6.5% and 9.0% respectively. In addition, the long-distance transport makes an important contribution to O_3 concentrations in

Jinan with the contribution ratio of OIC ranging from 2.6% to 42.7%. And the average contribution ratio of Others is 8.9%.

Different from O_3 , the sources of $PM_{2.5}$ in Jinan are relatively consistent over time. The average contribution ratio of OSD is 57.7%. The local contribution ratio of Jinan is between 14.0% and 33.3%. Moreover, Jiangsu and Anhui are important source regions with the maximum contribution ratios reaching 12.2% and 21.2% respectively.

The result of source tagging indicated that the contribution of inter-regional transport is greater than that of local emission to O_3 and $PM_{2.5}$ in Jinan. Therefore, in order to resolve the O_3 and $PM_{2.5}$ pollution problems, Jinan should cooperate with the neighboring cities to control the emissions of primary pollutants.

Selection of the Emission Control Region

In order to control the pollutions of O_3 and $PM_{2.5}$ effectively in the whole month of June 2015, we take the average contribution ratio as the standard to select emission control regions. The cities whose average contribution ratio to $O_3/PM_{2.5}$ concentrations on $O_3/PM_{2.5}$ pollution days is

Fig. 5. (a) The average contribution ratio of different regions to O_3 concentrations on O_3 pollution days in Jinan in June 2015. The black line represents the O_3 daily maximum 8-hour-averaged concentration (MDA8h O_3). (b) The average contribution ratio of different regions to $PM_{2.5}$ concentrations on $PM_{2.5}$ pollution days in Jinan in June 2015. The black line represents the $PM_{2.5}$ daily average concentration. The simulated results are from the simulation with $3 \text{ km} \times 3 \text{ km}$ horizontal resolution. OIC: other areas in China; OSD: other areas of Shandong; Others: other areas outside China.

more than 1% are presented in Fig. 6. Since the emission control of OIC and Others is too difficult to implement, the other cities excluding these 2 regions in Fig. 6 were chosen as the emission control region in the Source-tagging Control scenario.

Jinan and its neighboring cities Taian and Jining are important source regions of O₃ and PM_{2.5}. Especially for PM_{2.5}, the contributions from Jinan (23.6%), Taian (24.5%) and Jining (14.6%) are much higher than that from other cities and regions. By comparing the contributions of different regions to the two pollutants, it can be found that there are two major differences between the source regions of O₃ and PM_{2.5}: (1) OIC and Others contribute significantly to O₃ in Jinan with an average contribution ratio of 13.1% and 8.9%, respectively. But the 2 regions contribute less to PM_{2.5}. (2) Besides Jinan, Taian and Jining, the contributions of neighboring cities to PM_{2.5} are mainly caused by Linyi (4.0%), Xuzhou (3.5%), Dezhou (3.5%), Zaozhuang (2.7%) and Laiwu (2.3%). However, the O₃ concentration comes from widely distributed source regions with 9 cities having an average contribution ratio between 1–2%. The differences indicate that controlling O₃ pollution can be more difficult because of the large contribution of long-distance transport and its widely distributed source regions.

Since our goal is to control O₃ and PM_{2.5} pollutions simultaneously, we take the average contribution ratios of both O₃ and PM_{2.5} into account when choosing emission control regions. Jinan, Taian and Jining are the cities whose average contribution ratio to O₃ or PM_{2.5} is more than 10%. These 3 cities are supposed to be Level I cities in the Source-tagging Control region. Cities with an average contribution ratio to O₃ or PM_{2.5} less than 10% and greater than or equal to 2% are Dezhou, Binzhou, Xuzhou, Suzhou, Laiwu, Linyi and Zaozhuang, which are Level II cities. The other cities in the Source-tagging Control region are considered as Level III cities (with an average contribution ratio to O₃ or PM_{2.5} less than 2% and greater than or equal to 1%). The locations of 16 cities in the Source-tagging Control region are shown in Fig. 7(a). And following the conventional regional

emission control measures, the 18 cities shown in Fig. 7(b) are supposed to be the Conventional Control region.

Assessment of Emission Control Schemes

In order to evaluate the effect of implementing emission control in the source-tagging region on O₃ and PM_{2.5} concentrations in Jinan and compare the effect with that of conventional control measures, we devise two experiment scenarios (“Source-tagging Control 1” and “Conventional Control 1”). The cities and the emission reduction percentages in Source-tagging Control 1 and Conventional Control 1 are given in Table 2 and 3 respectively. Referring to the selected source-tagging region, we propose the emission control scheme in Source-tagging Control 1: The various pollutants are reduced by ER1 (ER: emission reduction percentage) in the source-tagging region with different response levels. Similarly, the conventional emission control scheme is proposed in Conventional Control 1: The various pollutants are reduced by ER1 in the Conventional Control region. According to the Emergency Plans for Heavy Pollution Weather issued by the government of involved provinces, the ER1 of various pollutants in cities of different response level are tentatively set. And considering the negative effect of NO_x emission reduction on O₃ reduction (Sillman, 1999; Jiménez *et al.*, 2004; Tang *et al.*, 2010; Kanaya *et al.*, 2016), the emission reduction percentages of NO_x and VOCs have been slightly adjusted.

As shown in Figs. 8(a) and 8(b), these two schemes have significant differences in the degree of emission control. The total reduction emissions of various pollutants in the source-tagging region are obviously less than that in the Conventional Control region, especially for PM_{2.5}, NO_x and SO₂. The total reduction emissions of PM_{2.5}, NO_x and SO₂ are 15.6 kg, 32.1 kg and 62.4 kg in Source-tagging Control 1 which are 55.5%, 50.1% and 52.7% of those in Conventional Control 1, respectively. Fig. 8(d) gives the areas and population of the Source-tagging Control region and Conventional Control region. It can be found that the emission control measures in Conventional Control 1 need to involve more

Fig. 6. The cities whose average contribution ratio to O₃/PM_{2.5} concentrations on O₃/PM_{2.5} pollution days in Jinan in June 2015 is more than 1%. The red dotted lines represent the thresholds for different response levels of emission control. The simulated results are from the simulation with 3 km × 3 km horizontal resolution.

Fig. 7. (a) The locations of 16 cities in DST region: 1) Jinan, 2) Taian, 3) Jining, 4) Dezhou, 5) Binzhou, 6) Laiwu, 7) Linyi, 8) Zaozhuang, 9) Xuzhou, 10) Suzhou, 11) Liaocheng, 12) Heze, 13) Shangqiu, 14) Suqian, 15) Bozhou and 16) Chuzhou. (b) The locations of 18 cities in the conventional emission control region: 1) Jinan, 2) Taian, 3) Jining, 4) Liaocheng, 5) Dezhou, 6) Binzhou, 7) Dongying, 8) Zibo, 9) Laiwu, 10) Linyi, 11) Zaozhuang, 12) Heze, 13) Puyang, 14) Handan, 15) Xingtai, 16) Hengshui, 17) Cangzhou and 18) Weifang. SD: Shandong; HB: Hebei; HN: Henan; AnH: Anhui; JS: Jiangsu; DST region: the emission control region defined by source tagging.

Fig. 8. (a) The total reduction in emissions of various pollutants in the emission control region in June 2015. (b) The area and population of the emission control region. (c) The reduction ratio of PM_{2.5} ($RR_{PM2.5}$) on PM_{2.5} pollution days. (d) The reduction ratio of O₃ (RR_{O3}) on O₃ pollution days. Source-tagging Control 1: the Source-tagging Control scenario with the various pollutants reduced by ER1 (see Table 2); Conventional Control 1: the Conventional Control scenario with the various pollutants reduced by ER1 (see Table 3). The simulated results are from the simulation with 3 km × 3 km horizontal resolution.

people and more areas. The affected areas and the population of Source-tagging Control 1 were smaller than those of Conventional Control 1 by 12% and 31% respectively. Another important advantage of Source-tagging Control 1 is that the meteorological factors were taken into account to select the Source-tagging Control region. Therefore, it is a more flexible emission reduction scheme. For different pollution cases, the more targeted Source-tagging Control regions can be selected according to the source-tagging modeling results to achieve better effect on controlling pollutions.

The reduction ratio of $PM_{2.5}$ ($RR_{PM_{2.5}}$) and O_3 (RR_{O_3}) concentrations in Source-tagging Control 1 and Conventional Control 1 are shown in Fig. 8(c) and 8(d). It shows that the $PM_{2.5}$ concentrations on most pollution days could be controlled effectively in Source-tagging Control 1 and Conventional Control 1. But it is worth noting that except June 23 and 26, $RR_{PM_{2.5}}$ in Source-tagging Control 1 is higher than that in Conventional Control 1. The average $RR_{PM_{2.5}}$ in Source-tagging Control 1 (22.0%) is slightly higher than that in Conventional Control 1 (20.7%). Under Source-tagging Control 1, the average concentration of $PM_{2.5}$ on pollution days is $71.4 \mu g m^{-3}$ which is 22.9% lower than that in the base scenario ($92.4 \mu g m^{-3}$). Under Conventional Control 1, the average concentration of $PM_{2.5}$ on pollution days is $72.7 \mu g m^{-3}$ which is 21.3% lower than that in the base scenario.

However, the two emission reduction scenarios both have

negative effects on O_3 reduction and RR_{O_3} , -1.7% and -1.8% in Source-tagging Control 1 and Conventional Control 1, respectively. This result may be caused by the simultaneous emission control of the 2 important O_3 precursors (NO_x and VOC) with unsuitable emission reduction percentages. To explore this problem, two more experiments (“Source-tagging Control 2” and “Conventional Control 2”) were devised to produce more effective control effect on O_3 pollution. Referring to Tables 2 and 3, the various pollutants are reduced by ER2 in the Source-tagging Control region of different response levels in Source-tagging Control 2. Similarly, in Conventional Control 2, the various pollutants are reduced by ER2 in the Conventional Control region. Compared with ER1, the emission reduction percentages of NO_x and VOCs in ER2 has been adjusted to reduce more VOCs and less NO_x .

As shown in Figs. 9(a) and 9(b), the total reduction in emissions of primary pollutants in Source-tagging Control 2 was only 61% of those in Conventional Control 2, and the affected areas and the population of Source-tagging Control 2 were also smaller than that of Conventional Control 2 by 12% and 31% respectively. The reduction ratio of $PM_{2.5}$ ($RR_{PM_{2.5}}$) and O_3 (RR_{O_3}) concentrations in Source-tagging Control 2 and Conventional Control 2 are shown in Figs. 9(c) and 9(d). Compared with the results in Source-tagging Control 1 and Conventional Control 1, the $RR_{PM_{2.5}}$ in Source-tagging Control 2 and Conventional Control 2 change little with a small increase. However, the control effect of O_3

Table 2. The response levels of cities in the emission control region defined by source tagging and the corresponding emission reduction percentages of various pollutants in Source-tagging Control 1 and Source-tagging Control 2. ER: emission reduction percentage.

Response level	City	Species	ER1 ^a (%)	ER2 (%)
I	Jinan	BC	50	50
	Taian	OC	50	50
	Jining	$PM_{2.5}$	50	50
		SO_2	60	60
		NO_x	40	10
		VOCs	50	80
II	Dezhou	BC	40	40
	Binzhou	OC	40	40
	Xuzhou	$PM_{2.5}$	40	40
	Suzhou	SO_2	50	50
	Laiwu	NO_x	30	10
	Linyi	VOCs	40	70
	Zaozhuang			
III	Heze	BC	30	30
	Shangqiu	OC	30	30
	Liaocheng	$PM_{2.5}$	30	30
	Bozhou	SO_2	40	40
	Chuzhou	NO_x	20	10
	Suqian	VOCs	30	60

^a The ER1 of various pollutants are tentatively set according to the Emergency Plans for Heavy Pollution Weather issued by the government of Shandong Province (http://www.shandong.gov.cn/sdxxgk/publi/message/detail.do?identifier=ml_0123-01-2017-000545), Jiangsu Province (http://www.jiangsu.gov.cn/art/2019/1/18/art_46144_8089267.html), Henan Province (<https://www.henan.gov.cn/2018/11-16/722610.html>) and Anhui Province (<http://xxgk.ah.gov.cn/UserData/DocHtml/731/2017/5/5/962670675764.html>).

Table 3. The response levels of cities in the Conventional Control region and the corresponding emission reduction percentages of various pollutants in Conventional Control 1 and Conventional Control 2. ER: emission reduction percentage.

Response level	City	Species	ER1 (%)	ER2 (%)
I	Jinan	BC	50	50
	Taian	OC	50	50
	Jining	PM _{2.5}	50	50
	Liaocheng	SO ₂	60	60
	Dezhou	NO _x	40	10
	Binzhou	VOCs	50	80
	Dongying			
	Zibo			
	Laiwu			
	Linyi			
	Zaozhuang			
	Heze			
	Puyang			
	Handan			
	Xingtai			
	Hengshui			
	Cangzhou			
	Weifang

Fig. 9. (a) The total reduction in emissions of various pollutants in the emission control region in June 2015. (b) The area and population of the emission control region. (c) The reduction ratio of PM_{2.5} ($RR_{PM_{2.5}}$) on PM_{2.5} pollution days. (d) The reduction ratio of O₃ (RR_{O_3}) on O₃ pollution days. Source-tagging Control 2: the Source-tagging Control scenario with the various pollutants reduced by ER2 (see Table 2); Conventional Control 2: the Conventional Control scenario with the various pollutants reduced by ER2 (see Table 3). The simulated results are from the simulation with 3 km × 3 km horizontal resolution.

pollution has been significantly improved. In Source-tagging Control 2 and Conventional Control 2, the RR_{O_3} has been raised to 16.2% and 15.7% and pollution days were

reduced by 6 and 5 days respectively. The results indicated that controlling VOC emissions are more effective for lowering O₃ concentrations than controlling NO_x.

CONCLUSIONS

This study provides a case study of numerical emission control experiments based on the NAQPMS source-tagging modeling results and explores more flexible and more efficient measures for reducing simultaneous pollution from urban O₃ and PM_{2.5}.

The large contributions from the surrounding cities indicate that inter-regionally transported emissions are a dominant factor in the O₃ and PM_{2.5} pollution in Jinan; thus, targeting a region based on source-tagging modeling results increases the flexibility and efficiency of regional control measures. In our experiments, the total reduction in primary pollutants in Source-tagging Control was only 61% of that in Conventional Control, and the area and the population that experienced the effects of emission reduction were smaller by 12% and 31%, respectively. However, the results suggest that Source-Tagging Control can bring out a slightly better control effect than Conventional Control in reducing simultaneous pollution by O₃ and PM_{2.5}, achieving reductions of 16.2% in O₃ concentrations and 22.8% in PM_{2.5} concentrations on days polluted by O₃ and PM_{2.5}, respectively. Moreover, the meteorological factors were considered when selecting the Source-tagging Control region, enabling different regions to be targeted for different pollution processes and increasing the effectiveness of control measures.

Great challenges exist in coping with O₃ pollution due to the time variation and wide distribution of its source regions and the complexity of its chemistry. The negative effect of reducing NO_x emissions on O₃ control cannot be ignored. Furthermore, dynamically targeting emission reduction regions is urgently needed.

This preliminary study provides several useful findings on emission control during simultaneous pollution by urban O₃ and PM_{2.5}. Nevertheless, further analysis is warranted. First, more O₃ production sensitivity studies are needed in order to explore the response of this pollutant to reduced NO_x and VOC emissions and thereby identify appropriate percentages of reduction. Second, the obvious variation of the O₃ source regions with time indicate that dynamically selecting emission reduction regions is necessary. Moreover, a model that simulates the total reduction in emissions, the affected population and economy and other relevant factors should be developed to scientifically assess the cost and the effectiveness of proposed control measures.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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