Simulation-based Design of Regional Emission Control Experiments under Simultaneous Pollution of O$_3$ and PM$_{2.5}$ in Jinan, China

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Abstract

High O$_3$ and PM$_{2.5}$ concentrations were frequently observed in Jinan during June 2015. The simultaneous pollutions of O$_3$ and PM$_{2.5}$ occurred on 8 days in June with the maximum 8-hour average O$_3$ concentration of 255$\mu$g m$^{-3}$ and the maximum daily average PM$_{2.5}$ concentration of 111$\mu$g m$^{-3}$. In order to investigate how to perform simultaneous control of the 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 (named conventional control) was reducing the emissions over Jinan and surrounding areas with the strictest control measures. The results suggested that this control scenario resulted in 15.7% reduction of O$_3$ concentrations on O$_3$ polluted days and 21.3% reduction of PM$_{2.5}$ concentrations on PM$_{2.5}$ polluted days. The other emission control scenario (named source-tagging control) was designed based on the online source tagging modeling results from NAQPMS. Different from the conventional control, the emission reduction region was selected based on their source contributions to O$_3$ and PM$_{2.5}$ concentrations in Jinan. And the emission reduction intensity over the areas with small source contributions to O$_3$ and PM$_{2.5}$ concentrations in Jinan were significantly lower than that in conventional control. The total reduction emissions of primary pollutants were only 61% of that in the conventional control, and the areas and population affected of this control were also smaller than the conventional control by 12% and 31%.

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respectively. The modeling results suggest that the source-tagging control can bring out a slightly better control effect than the conventional control. 16.2% reduction of O₃ was obtained on O₃ polluted days and 22.8% reduction of PM₂.₅ concentrations on PM₂.₅ polluted days. This highlights the advantages and potentials of using the source tagging modeling results in designing the regional control measures under the simultaneous pollution of O₃ and PM₂.₅.

**Keywords:** Emission control; Source tagging method; Simultaneous pollution; O₃; PM₂.₅.

**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 (PM₂.₅) 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 PM₂.₅ 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 PM₂.₅ 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 PM₂.₅ and O₃ pollution.

The co-occurrence of PM₂.₅ 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 PM₂.₅ in summer. Zhang et al. (2015) have found that when the concentrations of PM₂.₅ precursors (SO₂, NOₓ, etc.) were high, NOₓ could destroy O₃ at night, causing a negative correlation between O₃ and PM₂.₅. However,
Shi et al. (2015) reported a positive-correlation ($R \sim 0.59$) 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 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; Wen et al., 2016; Liu et al., 2016; Li et al., 2017). 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 O₃–NOₓ–hydrocarbon chemistry. For aerosol chemistry, an aerosol thermodynamic model (ISORROPIA1.7) is used to calculate the composition of an inorganic aerosol system (Nenes et al., 1998). Based on Odum and Seinfeld (1997) and Pandis et al. (1991), the reaction rates and methods for two anthropogenic precursors and four biogenic precursors was calculated to form six secondary organic aerosols (SOA). 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 km × 27 km horizontal resolution on 106 × 106 grids. The second domain includes eastern and northern China with a 9 km × 9 km horizontal resolution on 163 × 151 grids. The third domain covers Shandong Province and its surrounding provinces with a 3 km × 3 km horizontal resolution on 271 × 241 grids. The simulation period of air pollutants occurs from 25 May 2015 to 30 June 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°×1° from the National Center for Environment 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°×0.25° resolution. The HTAP dataset is a bottom-up database that includes seven 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., 2017). 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). VOCs emissions from the ocean are obtained from Precursors of Ozone and their Effects in the Troposphere (POET) database (Granier et al., 2005). NO\textsubscript{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\textsubscript{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\textsubscript{2} and NO\textsubscript{x}. Table S1 shows the total emissions of CO, SO\textsubscript{2} and NO\textsubscript{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 pollutants observation data are provided by the China National Environmental Monitoring Centre. 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 PM\textsubscript{2.5}, PM\textsubscript{10}, O\textsubscript{3}, SO\textsubscript{2}, NO\textsubscript{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 PM\textsubscript{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\textsubscript{2.5}, O\textsubscript{3} 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, 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:

\[
\frac{dF_r}{dt}_i = (E_{r,i})_{emis} + (P_{r,i})_{adv+conv+diff} + (C_{r,i})_{chem}
\]  

(1)

where \(i\) is the index of the model cell and \(r\) is the tagged region, and \((E_{r,i})_{emis}\) and
\((C_{r,i})_{chem}\) 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. \((P_{r,i})_{adv+conv+diff}\) 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 more 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, 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 O$_3$. 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 O$_3$/PM$_{2.5}$ concentrations on O$_3$/PM$_{2.5}$ pollution days of Jinan were calculated respectively. Cities whose average contribution ratio to O$_3$ 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 O$_3$ or PM$_{2.5}$, the cities in source-tagging control are divided into I, II and III response levels. 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 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$_x$, SO$_2$ and VOCs. The response level I corresponds to the strictest emission reduction, the response level II corresponds to the smaller reduction percentages, and the 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$_{2.5}$ ($RR_{PM_{2.5}}$) and O$_3$ ($RR_{O_3}$) are calculated as follows:

$$RR_{PM_{2.5}} = \left( \frac{PM_{2.5,\text{base}} - PM_{2.5,\text{er}}} {PM_{2.5,\text{base}}} \right) \times 100\%$$
where $PM_{2.5,\text{base}}$ and $PM_{2.5,s}$ represent the PM$_{2.5}$ daily average concentration in the base scenario and an emission control scenario, and $O_{3,\text{base}}$ and $O_{3,s}$ are the O$_3$ daily maximum 8-hour average concentration (MDA8h O$_3$) in the base scenario and one emission control scenario.

In this paper, the PM$_{2.5}$ and O$_3$ pollution days are defined as the PM$_{2.5}$ daily average concentration greater than 75 $\mu$g m$^{-3}$ and the MDA8h O$_3$ greater than 160 $\mu$g m$^{-3}$, respectively.

RESULTS AND DISCUSSION

Validation of the simulation results

The observation data of the main meteorological factors (surface temperature, relative humidity, wind direction and speed) and pollutants (PM$_{2.5}$ and O$_3$) 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 four meteorological factors in the simulation period and not only reflects the changing trend of meteorological factors but also represents the observation values accurately. The simulation of the surface temperature works well with correlation coefficients ($r$) of 0.86-0.89 and average mean...
Biases (MB) of 0.1~2.0°C. The correlation coefficient of relative humidity is 0.79-0.88 except for a slight underestimation of the RH in Beijing. Compared with temperature and relative humidity, the simulation of wind speed is slightly worse, with a correlation coefficient of 0.39-0.70 and average mean bias of 0.4~1.8 m s⁻¹. For wind direction, the benchmark suggested by Emery et al. (2001) is the average 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 PM₂.₅ and O₃ in Jinan, Beijing, Tianjin and Taiyuan in June 2015. The model shows a good simulation capability for PM₂.₅ 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 PM₂.₅ are consistent with the observations with the average biases 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 PM₂.₅ and O₃, which provides a good data basis for analyzing the spatiotemporal variation and regional transport process of PM₂.₅ and O₃ in the study period.

Characteristics of the combined pollution episode

Fig. 4 shows the variation of PM₂.₅ and O₃ concentrations and meteorological factors over Jinan in June. It can be found from the variation of PM₂.₅ 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 PM₂.₅ pollution days. And there were 8 days occurred simultaneous
pollutions of O$_3$ and PM$_{2.5}$ (on June 7, 10, 16, 17, 22, 26, 27, 28). It indicated that almost all PM$_{2.5}$ pollutions were accompanied by O$_3$ pollutions.

As shown in Fig. 4(a), the ozone concentration exceeded 160 $\mu$g m$^{-3}$ slightly on June 1, and then the two pollutant concentrations continued to decline until June 5. Starting from 5 June, the concentration of pollutants gradually increased, and the PM$_{2.5}$ and O$_3$ combined pollution occurred on 7 June, with the PM$_{2.5}$ daily average concentration and MDA8h O$_3$ of 82 $\mu$g m$^{-3}$ and 233 $\mu$g 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 7 June, the pollutant concentrations reached the maximum, with the decrease in wind speed and 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 PM$_{2.5}$ and O$_3$ both rose sharply. The sea-level synoptic pressure patterns over central and eastern China on 6 and 7 June are displayed in Fig. S3. It can be seen that on 7 June, there existed a weak high-pressure system over the north 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 8 June, 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 the June 9 and 10 with MDA8h O$_3$ of 207 $\mu$g m$^{-3}$ and 217 $\mu$g m$^{-3}$, respectively. Moreover, PM$_{2.5}$ pollution
occurred on 10th. On 11 June, Jinan was dominated by the strong northwesterly wind which effectively removed the pollutants.

Starting from the June 14, the concentrations of O₃ and PM₂.₅ began to rise gradually. There were six days (June 16, 17, 22, 26, 27 and 28) when co-pollution occurred. The peak of the MDA₈h O₃ was 255 μg m⁻³ and the peak of PM₂.₅ daily average concentration was 111 μg m⁻³, which both occurred on 16 June. From June 14 to 30, Jinan was affected by low wind speed (≤2m s⁻¹) continuously throughout the pollution episode, especially in the period of high pollutant concentrations with a mostly westerly or southwest wind direction. 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 O₃ and PM₂.₅**

The contributions to O₃ and PM₂.₅ from various regions can be quantified by the online pollutant source tagging module implemented in the NAQPMS modeling system. The contributions from the surrounding regions to O₃/PM₂.₅ concentrations on O₃/PM₂.₅ pollution days in Jinan are shown in Fig.5. For O₃, 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 two important sources of O₃ with the average contribution ratio of 6.5% and 9.0% respectively. In addition, the long-distance transport makes an important contribution to O₃ 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 neighbor 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 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 two regions in Fig.6 were chosen as the emission control region in source-tagging control scenario.

Jinan and its neighbor cities Taian and Jining are important source regions of O$_3$ 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 O$_3$ and PM$_{2.5}$: (1) OIC and Others contribute significantly to O$_3$ in Jinan with an average contribution ratio of 13.1% and 8.9%, respectively. But the two regions contribute less to
PM$_{2.5}$. (2) Besides Jinan, Taian and Jining, the contributions of neighbor 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 O$_3$ concentration comes from widely distributed source regions with 9 cities have an average contribution ratio between 1%-2%. The differences indicate that controlling O$_3$ 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 O$_3$ and PM$_{2.5}$ pollutions simultaneously, we take the average contribution ratios of both O$_3$ and PM$_{2.5}$ into account when choosing emission control regions. Jinan, Taian and Jining are the cities whose average contribution ratio to O$_3$ or PM$_{2.5}$ is more than 10%. These three cities are supposed to be level I cities in source-tagging control region. Cities with an average contribution ratio to O$_3$ 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 source-tagging control region are considered as level III cities (with an average contribution ratio to O$_3$ or PM$_{2.5}$ less than 2% and greater than or equal to 1%). The locations of 16 cities in 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 measures in source-tagging region on O$_3$ and PM$_{2.5}$ concentrations in Jinan and compare the effect with that of conventional control, 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 Table 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 in 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 O\(_3\) 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 Fig. 8(a) and Fig. 8(b), these two schemes have significant differences in the degree of emission control. The total reduction emissions of various pollutants in source-tagging region are obviously less than that in conventional control region, especially for PM\(_{2.5}\), NO\(_x\) and SO\(_2\). The total reduction emissions of PM\(_{2.5}\), NO\(_x\) and SO\(_2\) are 15.6kg, 32.1kg and 62.4kg 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 source-tagging control region and conventional control region. It can be found that the emission control measures in conventional control 1 need to be involve more 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 Fig. 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 $RR_{O_3}$ are -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 two 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 Table 2 and Table 3, the various pollutants are reduced by ER2 in 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 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 Fig. 9(a) and Fig. 9(b), the total reduction emissions of primary pollutants in source-tagging control 2 were 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 Fig. 9(c) and Fig. 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$_3$ 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 VOCs emissions are more effective for lowering O$_3$ concentrations than controlling NO$_x$.

**CONCLUSIONS**

This study has provided a case study of numerical emission control experiments based on the NAQPMS source tagging modeling results and has explored the more flexible and efficient emission control measures under simultaneous pollutions from urban O$_3$ and PM$_{2.5}$.

The large contributions from the surrounding cities indicating that the inter-regional transports had a dominant contribution to the O$_3$ and PM$_{2.5}$ pollutions in Jinan. Designing the emission control region based on the source tagging modeling results is proved to be a more efficient and flexible method of developing regional control measures. The total reduction emissions of...
primary pollutants in source-tagging control were only 61% of those in conventional control, and
the affected areas and the population of source-tagging control were also smaller by 12% and 31%
than conventional control respectively. The modeling results suggest that the source-tagging
control can bring out a slightly better control effect than the conventional control with 16.2% and
22.8% reductions of O$_3$ and PM$_{2.5}$ concentrations on the O$_3$ polluted days and PM$_{2.5}$ polluted days
respectively. Moreover, the meteorological factors were taken into account to select the
source-tagging control region, which makes the more targeted source-tagging control regions can
be selected for different pollution processes according to the source tagging modeling results to
achieve better control effect on pollutions. This highlight the advantages and potentials of using
the source tagging modeling results in designing the regional control measures under the
simultaneous pollution of O$_3$ and PM$_{2.5}$.

The control of O$_3$ pollution faces great challenges due to the time variation of its source regions,
the widely distributed source regions and the complexity of O$_3$ chemistry. The negative effect of
NO$_x$ emission reduction on controlling O$_3$ cannot be ignored. Furthermore, the dynamic control
of emission reduction region is urgently needed to achieve more effective control of O$_3$ pollution.

This is a preliminary study to provide some useful findings regarding emission control under
simultaneous pollutions from urban O$_3$ and PM$_{2.5}$. Nevertheless, further analyses are merited.
First, in order to identify suitable reduction emission percentages of NO$_x$ and VOCs, more O$_3$
production sensitivity studies are needed to explore the response of O$_3$ production to NO$_x$ and
VOCs emission reduction. Secondly, the differences between O$_3$ and PM$_{2.5}$ source regions suggest
the possibility for the dynamic control of emission reduction regions. Moreover, a model
involving total reduction emissions, population, economy and other relevant factors can be
developed to assess the cost of an emission control measure, thus providing scientific support for making more efficient control measures.

ACKNOWLEDGEMENTS

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Table 1. The classification criteria for different response levels.

<table>
<thead>
<tr>
<th>The average contribution ratio to O₃ or PM₂.₅</th>
<th>Response level</th>
</tr>
</thead>
<tbody>
<tr>
<td>≥10%</td>
<td>I</td>
</tr>
<tr>
<td>≥2% and &lt; 10%</td>
<td>II</td>
</tr>
<tr>
<td>≥1% and &lt; 2%</td>
<td>III</td>
</tr>
</tbody>
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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.

<table>
<thead>
<tr>
<th>Response level</th>
<th>City</th>
<th>Species</th>
<th>ER1 (%)</th>
<th>ER2 (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
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<td>BC</td>
<td>50</td>
<td>50</td>
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<tr>
<td></td>
<td></td>
<td>OC</td>
<td>50</td>
<td>50</td>
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<tr>
<td></td>
<td>Taian</td>
<td>PM2.5</td>
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<td>50</td>
</tr>
<tr>
<td></td>
<td>Jining</td>
<td>SO₂</td>
<td>60</td>
<td>60</td>
</tr>
<tr>
<td></td>
<td></td>
<td>NOₓ</td>
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<td>10</td>
</tr>
<tr>
<td></td>
<td></td>
<td>VOCs</td>
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<td>80</td>
</tr>
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<td></td>
<td>Dezhou</td>
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<td>40</td>
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</tr>
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<td>Binzhou</td>
<td>OC</td>
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<td></td>
<td>Xuzhou</td>
<td>PM2.5</td>
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<td>Suzhou</td>
<td>SO₂</td>
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<td>50</td>
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<tr>
<td></td>
<td>Laiwu</td>
<td>NOₓ</td>
<td>30</td>
<td>10</td>
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<tr>
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<td>VOCs</td>
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<td>70</td>
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<td>Liaocheng</td>
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<td>Bozhou</td>
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<td>Suqian</td>
<td>VOCs</td>
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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.

<table>
<thead>
<tr>
<th>Response level</th>
<th>City</th>
<th>Species</th>
<th>ER1 (%)</th>
<th>ER2 (%)</th>
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<tr>
<td>I</td>
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<tr>
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<td>Binzhou</td>
<td>VOCs</td>
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Figure 2. Comparison of hourly variation of simulated and observed surface temperature (T), relative humidity (RH) and wind speed (WS) at Jinan in June 2015. The simulated results are from the simulation with 3km × 3km horizontal resolution.
Figure 3. Comparison of hourly variation of simulated and observed PM$_{2.5}$ and O$_3$ concentrations at Jinan in June 2015. The simulated results are from the simulation with 3km × 3km horizontal resolution.
Figure 4. Time series of observed pollutant concentrations and major meteorological parameters. (a) The PM$_{2.5}$ daily average concentration and the O$_3$ daily maximum 8-hour average concentration (MDA8h O$_3$); (b) hourly PM$_{2.5}$ concentration and wind speed; (c) hourly O$_3$ concentration and wind speed; (d) hourly wind speed; (e) hourly surface temperature (T) and relative humidity (RH).
Figure 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 average 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 3km × 3km horizontal resolution. OIC: Other areas in China; OSD: Other areas of Shandong; Others: Other areas outside China.
Figure 6. The cities whose average contribution ratio to O$_3$/PM$_{2.5}$ concentrations on O$_3$/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 3km $\times$ 3km horizontal resolution.
Figure 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, 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, 18-Weifang. SD: Shandong; HB: Hebei; HN: Henan; AnH: Anhui; JS: Jiangsu; DST region: the emission control region defined by source tagging.
Figure 8. (a) The total reduction emissions of various pollutants in 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 O$_3$ ($RR_{O_3}$) on O$_3$ pollution days. source-tagging control 1: source-tagging control scenario with the various pollutants reduced by ER1 (see Table 2); conventional control 1: conventional control scenario with the various pollutants reduced by ER1 (see Table 3). The simulated results are from the simulation with 3km × 3km horizontal resolution.
Figure 9. (a) The total reduction emissions of various pollutants in 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 O$_3$ ($RR_{O_3}$) on O$_3$ pollution days. source-tagging control 2: source-tagging control scenario with the various pollutants reduced by ER2 (see Table 2); conventional control 2: conventional control scenario with the various pollutants reduced by ER2 (see Table 3). The simulated results are from the simulation with 3km $\times$ 3km horizontal resolution.