Model analyses of changes in spring surface ozone concentrations over Shandong Province in the period of 2014–2017

Jialin Li 1, Juzhen Cai3,* Houfeng Liu5, Xiao Han1,4, Yongfu Xu 1,4, Xiaofang Cai6, Meigen Zhang 1,2,4,*

1 State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China
2 Center for Excellence in Urban Atmospheric Environment, Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, China
3 Zhejiang Climate Center, Hangzhou 310017, China
4 University of Chinese Academy of Sciences, Beijing 100049, China
5 School of Geography and Environment, Shandong Normal University, Jinan 250358, China
6 Taiyuan Meteorological Service, Taiyuan 030082, China

ABSTRACT

The concentrations of surface ozone (O₃) in eastern China have increased significantly in recent years, resulting in the earlier appearance of more serious O₃ pollution. Measurements at 14 stations in Shandong Province showed that the monthly mean O₃ concentrations in the late spring (May) increased by 22.2 µg m⁻³ from 2014 to 2017. To investigate the reasons of the increase of O₃ in springtime from
2014 to 2017, the changes of \( \text{O}_3 \) concentrations due to meteorological conditions and emissions in May were studied based on ambient measurements and simulations with the RAMS-CMAQ modeling system. By analyzing the observed data, it was found that the variations in wind field were conducive to the accumulation of \( \text{O}_3 \), while the effects of other meteorological parameters on \( \text{O}_3 \) concentrations were different at the same site between years. Further to perform a series of simulations with only the meteorological conditions changed in May from 2014 to 2017, the results showed that the effects of variations in meteorological conditions had become more and more important in the changes of \( \text{O}_3 \) concentrations in May between years, especially the factors that affected the photochemical generation of \( \text{O}_3 \). For example, the percentage of the sites where the changes of \( \text{O}_3 \) concentrations were dominated by the variations of the meteorological conditions increased from 28.6% to 78.6% over the region in May between years. Besides, the changes in \( \text{NO}_x \) emissions had a close relationship with the variations in \( \text{O}_3 \) concentrations when the changes of \( \text{O}_3 \) were dominated by the emission variations.

**Keywords:** Ozone, Meteorological conditions, Emissions effects, RAMS-CMAQ, Shandong

1. **Introduction**

Tropospheric ozone (\( \text{O}_3 \)), a major oxidant, is mainly produced by the photochemical oxidation between the volatile organic compounds (VOCs) and nitrogen oxides (\( \text{NO}_x \)). If ozone concentration in the troposphere exceeds the natural...
level, it has adverse effects on human health (Canella et al., 2016), vegetation (Feng et al., 2015; Van Dingene et al., 2009) and climate (Stevenson et al., 2013; Worden et al., 2008). According to the available monitoring data from 1950–1979 until 2000–2010 for the Northern Hemisphere, surface O$_3$ concentration has increased globally during the 20th century with an increase of 1–5 ppbv per decade (Sun et al., 2019). Thus, attentions have been attracted to tropospheric O$_3$ worldwide (e.g. Marais et al., 2014; Monks et al., 2015; Zeng et al., 2018).

With the rapid growth of domestic economy and urbanization, a significant increase has appeared in O$_3$ concentration of China since the 1990s (Xing et al., 2011; Sun et al., 2019). Ma et al. (2016) showed that the maximum daily 8-hour average (MDA8) O$_3$ concentration increased by 1.1 ppbv per year from 2003 to 2015 at the rural site of Beijing called Shangdianzi. Sun et al. (2016) presented that there was an increase of 1.7–2.1 ppbv yr$^{-1}$ at Mt. Tai during summertime from 2003 to 2015. Wang et al. (2017b) reported that when compared to 2013, yearly average MDA8 O$_3$ concentrations increased by 12%, 25%, 34% and 22% in Beijing, Chengdu, Lanzhou, and Shanghai in 2015, respectively. Though a series of stringent emission control measurements have been taken in China (Li et al., 2019a; Zhao et al., 2013), O$_3$ pollution has become worse. According to the report of O$_3$ in 74 major cities from the China Ministry of Ecology and Environment, the annual averaged O$_3$ concentration increased from 139 µg m$^{-3}$ in 2013 to 166 µg m$^{-3}$ in 2018. Previous studies have revealed the likely causes for the variations of tropospheric O$_3$ in different regions. Lou et al. (2015) presented that the variations of
meteorological conditions played a more important role in the interannual variability in surface O₃ than the changes of anthropogenic emissions over eastern China from 2004 to 2012. Lu et al. (2019) clarified that the increase of O₃ in 2017 compared to 2016 had something to do with the hotter and dryer weather conditions over major Chinese city clusters. Wang et al. (2019a) presented that the sensitive regime of O₃ formation in eastern China changed from VOC control to the mixed control due to the significant reduction in NOₓ emissions (25%) from 2012 to 2016 in eastern China, which resulted in more serious O₃ pollution. Wang et al. (2019b) has attributed the increase of O₃ to the decrease in PM₂.₅, which led to more solar actinic flux. Li et al. (2019c) reported that the increase of O₃ in the North China Plain was probably affected by the slowing down of the aerosol sink of hydroperoxyl (HO₂) radicals due to the decrease of PM₂.₅. Thus, O₃ formation is complex and varies regionally, which cannot be simply or uniformly attributed to one single factor. It is necessary to investigate the reasons for the variations of O₃ concentrations separately in each region of China to develop further action on O₃ control.

As presented above, high O₃ concentrations have been widely observed in eastern China in recent years (Lu et al., 2018; Wang et al., 2017a; Xue et al., 2014). Based on measurements from the Chinese National Environmental Monitoring Center (CNEMC), it was found that O₃ pollution became more serious in springtime and the peak of O₃ concentrations appeared earlier in Shandong Province. The observed monthly average O₃ concentrations increased by 22.2 µg m⁻³ in May from 2014 to 2017. However, few studies have focused on this compared with that in the developed
regions of eastern China, like Beijing–Tianjin–Hebei, the Yangtze River Delta and the
Pearl River Delta. To investigate the reasons of the increase of O$_3$ in springtime from
2014 to 2017, the roles of meteorology variations and emission changes on O$_3$
concentrations in May were identified based on ambient measurements and
simulations with the RAMS-CMAQ (the Regional Atmospheric Modeling System
coupled with the Community Multiscale Air Quality) modeling system in Shandong
Province during the time period 2014–2017.

2. Data and methods

2.1 Observational data

We collected the measured surface O$_3$, NO$_2$ and NO$_x$ concentrations at 14
monitoring sites (Fig. 1) in Shandong Province from January 2014 to December 2017
from the CNEMC. The corresponding meteorological factors (including temperature
[T], relative humidity [RH], wind speed [WS] and wind direction [WD]) were from
the Meteorological Information Comprehensive Analysis and Process System
(MICAPS). The measured data of cloud fraction (CF) was obtained from the
MODeRate Resolution Imaging Spectroradiometer (MODIS) through https://ladsweb.
modaps.eosdis.nasa.gov/search/ (last accessed 19 April, 2021).

2.2 Model description

The version of CMAQv4.7.1 coupled with the gas-phase photochemical
mechanism SAPRC99 (1999 Statewide Air Pollutant Research Center) (Carter, 2000)
was used to simulate and trace the evolution of the concentrations of pollutants.
Different inventories were combined to make the emission sources. The anthropogenic emissions over China were from the Multi-resolution Emission Inventory for China (MEIC) of 2014 (0.25° × 0.25°) (www.meicmodel.org, last accessed 17 November 2019), while the emissions of surrounding countries were from the MIX inventory (Li et al., 2017). The biogenic emissions provided by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2012) were derived from the Emissions of atmospheric Compounds and Compilation of Ancillary Data (ECCAD) database (https://eccad3.sedoo.fr, last accessed 8 October, 2021), namely MEGAN-MACC Biogenic emission inventory (0.5° × 0.5°, the year of 2010). The open biomass burning emissions of 2014 were collected from the Global Fire Emissions Database, Version 4 (Randerson et al., 2015). The monthly soil emissions of NOx were derived from the Regional Emission inventory in ASia, Version 2.1 (0.25° × 0.25°, the year of 2008) (Kurokawa et al., 2013), while the monthly lightning NOx emissions were obtained from the Global Emissions Inventory Activity (1° × 1°, the year of 2000) (Benkovitz et al., 1996). According to Han et al. (2004) and Gong et al. (2003), the online calculation of dust and sea salt emissions were added in the model. The output of RAMS provided the meteorological inputs to drive CMAQ. The initial and lateral boundary conditions used for the RAMS were from the National Center for Environmental Prediction reanalysis datasets.

Fig. 1 presents the two nested domains used in our simulations. The outer domain (D01), covering most of East Asia, was divided into 105 × 86 grid cells with a horizontal resolution of 64 km. The center of D01 was located at (35° N, 110° E). The
nested domain (D02) was a 16 km × 16 km horizontal resolution domain that covered the North China Plain (1504 km × 1440 km) with the center located at (40° N, 116° E).

Fig. 1. Model domain and geographical location of the 14 monitoring sites in Shandong Province. BZ, Binzhou; DY, Dongying; HZ, Heze; JN, Jinan; LC, Liaocheng; LW, Laiwu; LY, Linyi; QD, Qingdao; RZ, Rizhao; TA, Tai’an; WF, Weifang; WH, Weihai; ZB, Zibo; ZZ, Zaozhuang.

2.3 Sensitivity experiments

Fig. 2 shows the observed seasonal variations of O₃ concentrations in Shandong Province from 2014 to 2017. As shown in Fig. 2, the period of maximum of O₃ concentration (90.1–91.7 µg m⁻³) lasted from May to August in 2014, which was broader than that in 2017 (May and June, 112.3–117.8 µg m⁻³). Unlike the continuous increase of O₃ in early summer (June) between years, there was a significant and sudden increase of O₃ in the late spring (May) of 2017. It indicated that more serious ozone pollution appeared in springtime over Shandong Province. Thus, we decided to identify the roles of meteorology variations and emission changes on O₃ concentrations in May based on ambient measurements and simulations with the
Rams-CMAQ modeling system in Shandong Province during the time period 2014–2017.

Fig. 2. Observed seasonal variations of $O_3$ concentrations in Shandong Province from 2014 to 2017.

Four sensitivity experiments (Table 1) were designed to analyze the effects of emissions and meteorological conditions on pollutant concentrations over Shandong Province in May during the time period 2014–2017 with the RAMS-CMAQ modeling system.

We used the changes in the observed pollutant concentrations between different time spaces as the total changes (Eq. (1)). We kept the emissions of the CMAQ simulations unchanged between years and only changed the meteorological inputs. Thus, the changes in the predicted pollutant concentrations were due to differences in the meteorological conditions (Eq. (2)). The changes caused by different emissions were obtained by excluding the changes due to meteorological conditions from the
total changes (Eq. (3)). The scheme was proved to be reasonable in Wang et al. (2019b).

\[ \Delta O_{i,j} = \text{Obs}_{i,j} - \text{Obs}_{i,k} \]  
(1)

\[ \Delta M_{i,j} = \text{Sim}_{i,j} - \text{Sim}_{i,k} \]  
(2)

\[ \Delta E_{i,j} = \Delta O_{i,j} - \Delta M_{i,j} \]  
(3)

where \( i \) represents the pollutant; \( j \) and \( k \) represent the year \((j > k)\); \( \Delta O_{i,j} \) is the total changes in the observed concentration of pollutant \( i \) from year \( k \) to \( j \); \( \text{Obs}_{i,j} \) and \( \text{Obs}_{i,k} \) represent the observed concentration of pollutant \( i \) in year \( j \) and \( k \), respectively; \( \Delta M_{i,j} \) is the change in the simulated concentration of pollutant \( i \) in year \( j \) due to changes in the meteorological conditions compared to year \( k \); \( \text{Sim}_{i,j} \) and \( \text{Sim}_{i,k} \) represent the modeled concentration of pollutant \( i \) in year \( j \) and \( k \), respectively; and \( \Delta E_{i,j} \) is the difference in the simulated concentration of pollutant \( i \) between the year \( j \) and \( k \) due to the changes in emissions.

The contribution of the meteorological conditions and the changes in emissions cannot be completely differentiated as a result of the complex atmospheric processes and therefore our results were only approximations. This analysis was used to give an overview of Shandong Province.

**Table 1.** Model sensitivity experiments in this study.

<table>
<thead>
<tr>
<th>case</th>
<th>Name</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2014 standard (14E14M)</td>
<td>The standard simulation of ozone concentrations based on the emissions and meteorology in 2014</td>
</tr>
<tr>
<td>2</td>
<td>14E15M</td>
<td>Same as 1 but with the meteorology in 2015</td>
</tr>
<tr>
<td>3</td>
<td>14E16M</td>
<td>Same as 1 but with the meteorology in 2016</td>
</tr>
<tr>
<td>4</td>
<td>14E17M</td>
<td>Same as 1 but with the meteorology in 2017</td>
</tr>
</tbody>
</table>

**2.4 Model evaluation**
To evaluate the reliability of the modeled results, we compared the modeled and observed meteorological factors (including temperature, relative humidity, wind direction and wind speed) in May of each year and the hourly concentrations of pollutants (including O\textsubscript{3}, NO\textsubscript{2} and NO\textsubscript{x}) in May 2014 derived from 14 monitoring sites over Shandong (Tables 2 and 3).

Table 2 shows statistical results of the meteorological parameters, where: \( N \) presents the total number of samples; \( C_{\text{sim}} \) and \( C_{\text{obs}} \) are the averaged results of simulations and observations, respectively; MB is the mean bias; RMSE and GE are the root-mean-square and gross error, respectively; \( R \) is the correlation coefficient between the measured and simulated values; \( P_{22.5^\circ} \) and \( P_{45^\circ} \) represent the percentages of compared points at which the absolute biases between the modeled and measured wind directions are within 22.5° and 45°, respectively.

<table>
<thead>
<tr>
<th>variables</th>
<th>2014</th>
<th>2015</th>
<th>2016</th>
<th>2017</th>
</tr>
</thead>
<tbody>
<tr>
<td>T(ºC)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( N )</td>
<td>10387</td>
<td>9591</td>
<td>10296</td>
<td>4628</td>
</tr>
<tr>
<td>( C_{\text{obs}} )</td>
<td>21.7</td>
<td>19.8</td>
<td>19.9</td>
<td>21.6</td>
</tr>
<tr>
<td>( C_{\text{sim}} )</td>
<td>21.3</td>
<td>19.3</td>
<td>19.6</td>
<td>21.7</td>
</tr>
<tr>
<td>MB</td>
<td>-0.4</td>
<td>-0.5</td>
<td>-0.3</td>
<td>0.1</td>
</tr>
<tr>
<td>GE</td>
<td>2.3</td>
<td>2.1</td>
<td>2.0</td>
<td>2.2</td>
</tr>
<tr>
<td>RMSE</td>
<td>2.9</td>
<td>2.6</td>
<td>2.6</td>
<td>2.8</td>
</tr>
<tr>
<td>( R )</td>
<td>0.88</td>
<td>0.84</td>
<td>0.84</td>
<td>0.87</td>
</tr>
<tr>
<td>RH(%)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( N )</td>
<td>10386</td>
<td>9591</td>
<td>10295</td>
<td>4628</td>
</tr>
<tr>
<td>( C_{\text{obs}} )</td>
<td>50.8</td>
<td>61.2</td>
<td>58.8</td>
<td>57.6</td>
</tr>
<tr>
<td>( C_{\text{sim}} )</td>
<td>53.2</td>
<td>62.5</td>
<td>58.1</td>
<td>57.7</td>
</tr>
<tr>
<td>MB</td>
<td>2.4</td>
<td>1.3</td>
<td>-0.7</td>
<td>0.1</td>
</tr>
<tr>
<td>GE</td>
<td>11.6</td>
<td>10.7</td>
<td>10.8</td>
<td>11.1</td>
</tr>
<tr>
<td>RMSE</td>
<td>15.0</td>
<td>14.3</td>
<td>14.2</td>
<td>14.8</td>
</tr>
<tr>
<td>( R )</td>
<td>0.78</td>
<td>0.79</td>
<td>0.77</td>
<td>0.73</td>
</tr>
<tr>
<td>WS(m/s)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( N )</td>
<td>10387</td>
<td>9591</td>
<td>10296</td>
<td>4628</td>
</tr>
<tr>
<td>( C_{\text{obs}} )</td>
<td>2.7</td>
<td>2.4</td>
<td>2.3</td>
<td>2.3</td>
</tr>
<tr>
<td>( C_{\text{sim}} )</td>
<td>2.5</td>
<td>2.2</td>
<td>2.1</td>
<td>1.8</td>
</tr>
<tr>
<td>MB</td>
<td>-0.2</td>
<td>-0.2</td>
<td>-0.2</td>
<td>-0.5</td>
</tr>
<tr>
<td>GE</td>
<td>1.6</td>
<td>1.3</td>
<td>1.3</td>
<td>1.4</td>
</tr>
<tr>
<td>RMSE</td>
<td>2.0</td>
<td>1.8</td>
<td>1.7</td>
<td>1.8</td>
</tr>
</tbody>
</table>
$N$ is the total number of samples; $C_{\text{sim}}$ and $C_{\text{obs}}$ are the average values of modeled and observed results, respectively; MB is the mean bias; GE is the gross error; RMSE is the root-mean-square error; $R$ is the correlation coefficient between the observed and simulated results; $P_{22.5^\circ}$ and $P_{45^\circ}$ represent the proportions of compared results that the absolute biases between the simulated and measured wind directions are within $22.5^\circ$ and $45^\circ$, respectively.

Table 3. Performance statistics for hourly O$_3$, NO$_2$ and NO$_x$ derived from the 14 monitoring sites over Shandong in May of 2014.

<table>
<thead>
<tr>
<th></th>
<th>$N$</th>
<th>$C_{\text{obs}}$</th>
<th>$C_{\text{sim}}$</th>
<th>MB</th>
<th>RMSE</th>
<th>NMB</th>
<th>NME</th>
<th>$R$</th>
</tr>
</thead>
<tbody>
<tr>
<td>O$_3$ ($\mu$g m$^{-3}$)</td>
<td>9936</td>
<td>90.2</td>
<td>96.1</td>
<td>5.9</td>
<td>55.2</td>
<td>0.06</td>
<td>0.48</td>
<td>0.5</td>
</tr>
<tr>
<td>NO$_2$ ($\mu$g m$^{-3}$)</td>
<td>9650</td>
<td>40.8</td>
<td>39.0</td>
<td>-1.8</td>
<td>25.8</td>
<td>-0.04</td>
<td>0.45</td>
<td>0.6</td>
</tr>
<tr>
<td>NO$_x$ ($\mu$g m$^{-3}$)</td>
<td>10008</td>
<td>46.7</td>
<td>42.2</td>
<td>-4.5</td>
<td>32.3</td>
<td>-0.10</td>
<td>0.47</td>
<td>0.6</td>
</tr>
</tbody>
</table>

NMB and NME are the normalized mean bias and error.

As shown in Table 2, the model reproduced the variation and magnitude trend of the temperature and relative humidity quite well according to the statistical results. For T, though the absolute GE values were a little higher than the benchmark (2.0), the absolute MB values were within the benchmark (0.5) suggested by Emery et al. (2001) and the values of $R$ were no less than 0.8. Besides, the RMSE of T were comparable to that in Gao et al. (2016), also suggesting a reasonable simulation of T. While for RH, MB and R values were no more than 2.5% and no less than 0.7, respectively. Compared to Wang et al. (2019b), RMSE and GE values for RH in this study were even smaller. All these indicated the good reproduction of the relative humidity by the model. For WS, both the values of GE and MB met the benchmarks...
(GE \leq 2.0 \text{ and } |MB| \leq 0.5), suggesting a better simulation than Feng et al. (2016).

The values of RMSE for WS were also within the benchmark (\leq 2.0) for a good performance. For WD, P_{22.5^\circ} and P_{45^\circ} were larger than 40% and 60%, which was comparable to Li et al. (2019b), indicating the reasonable simulation of wind directions. Thus, the model was performed well to provide a reasonable meteorological field.

The evaluated results for the modeled O_3, NO_2 and NO_x concentrations are shown in Table 3. MB values of O_3, NO_2 and NO_x in May of 2014 were small (5.9, -1.8 and -4.5 \text{ µg m}^{-3}) with R values equaling 0.5, 0.6 and 0.6, respectively. Besides, the normalized mean bias (NMB) and the normalized mean error (NME) values of O_3 were comparable to those reported by Wang et al. (2019a), while NMB and NME values of NO_2 were smaller than the results of Wang et al. (2019a). Thus, the simulated pollutant concentrations were reliable.

3. Results and discussion

According to previous studies (e.g. Atkinson, 2000; Meleux et al., 2007), temperature, relative humidity, wind speeds and cloud fraction played important roles in O_3 formation. Thus, we first examined the relationships between the observed 90th percentile of hourly O_3 concentration (O_3-h_90) and the measured temperature, relative humidity, wind field and cloud fraction in May at the 14 monitoring sites during the time period 2014–2017. Fig. 3 shows the variations of observed O_3-h_90, temperature, relative humidity and CF, while Fig. 4 shows the statistical results for the
observed wind direction and wind speed.

**Fig. 3.** Observed (a) 90th percentile of hourly O₃ concentrations (O₃-h₉₀), the monthly mean (b) temperature (T) and relative humidity (RH) and (c) the cloud fraction (CF) in May at the 14 observation sites in Shandong Province during the time period 2014–2017. The different color columns in (a) represent different years, which is also used to indicate that in (b) and (c).

**Fig. 4.** Statistical results for wind direction and wind speed measured in May at the 14 observation
sites in Shandong Province during the time period 2014–2017.

Fig. 3a shows that O$_3$-h$_{90}$ decreased at most of the sites (except Binzhou, Liaocheng, Tai’an, Heze and Linyi) in May 2015 compared with 2014. Fig. 3b shows that the temperature decreased and the relative humidity either increased or remained stable in May 2015 compared with 2014. According to the previous studies (e.g. Atkinson, 2000; Hu et al., 2008; Zhang et al., 2015), lower T and higher RH were unfavorable for ozone production. The rate of ozone formation was repressed in May 2015 compared with 2014 due to the increase in CF, which can reduce the intensity of surface illumination (Fig. 3c). We therefore concluded that the temperature, relative humidity and CF in May 2015 reduced the formation of ozone compared with 2014 over Shandong.

O$_3$-h$_{90}$ in May 2016 decreased at most sites compared with 2015 (except Dongying, Laiwu, Tai’an, Zibo, Qingdao and Weihai) (Fig. 3a). A significant increase in the CF (Fig. 3c) led to a decrease in O$_3$ at all sites in May 2016 compared with 2015. Different from the CF, the effects from the changes in temperature and relative humidity were not consistent at all sites. However, it was easy to find that both the two factors had positive effects on the formation of O$_3$ at most sites in May 2016 compared with 2015 (Fig. 3b) due to the decrease of relative humidity and slight increase of temperature.

O$_3$-h$_{90}$ in May 2017 (Fig. 3a) significantly increased at most sites compared with 2016 (except Weihai and Zibo). Cloud fraction can affect the surface temperature and photochemical production of surface ozone due to its impacts on the amount of
insolation (Meleux et al., 2007; Lee et al., 2015); thus the decreased cloud fraction (Fig. 3c) at most sites (except Weihai) contributed to the increase of O₃ production. Besides, the decrease in the relative humidity (Fig. 3b) at most sites in May 2017 compared with 2016 was also conducive to the formation of O₃. As shown in Fig. 4, the observed distribution of winds in May was similar between the years with the southerly winds as the main winds. The continuous reduction in the mean wind speed (i.e. 2.7, 2.4, 2.3, and 2.3 m s⁻¹) of May from 2014 to 2017 can result in the less air mass of O₃ taken away from Shandong Province. Thus, the changes in wind field were conducive to the accumulation of O₃ in May during the time period 2014−2017.

In conclusion, the variations of meteorological parameters had different effects on the formation of O₃ in May between years. It was easy to identify the individual effect of the changes in each meteorological factor on O₃ production at each site, but the combined effects from all the meteorological conditions could not be determined. It was necessary to use the modeling system to evaluate the integrated effects. Thus, secondly, as described in Section 2.3, four sensitivity experiments (Table 1) were performed with RAMS-CMAQ modeling system to analyze the effects of emissions and meteorological conditions on pollutant concentrations over Shandong Province in May during the time period 2014−2017.

Fig. 5 shows the changes in the 90th percentile of the hourly O₃ concentrations (ΔO₃) in May between each year and the year before resulting from changes in meteorological conditions (ΔMₒ₃) and emissions (ΔEₒ₃) during the time period...

Fig. 5. Changes of 90th percentile of hourly O₃ concentrations (ΔO₃) due to changes in meteorological conditions (ΔM) and emissions (ΔE) in (a) May 2015 compared with May 2014, (b) May 2016 compared with May 2015, and (c) May 2017 compared with May 2016.

As shown in Fig. 5a, the variations of O₃ in May due to changes in meteorological conditions and emissions between 2015 and 2014 were -35.9−35.2 µg m⁻³ and -43.4−92.7 µg m⁻³, respectively. The changes in meteorological conditions in May 2015 could have caused the increase in O₃ (ΔM was positive) at most sites (11.8−35.2 µg m⁻³) (except Qingdao, Weihai and Rizhao). Our analyses of the results from Fig. 3 and 4 suggested that, except the effects of wind field, the changes of temperature, relative humidity and cloud fraction had negative effects on O₃ formation in May 2015 compared to 2014. Therefore, it can be concluded that the changes of wind field have dominated the effects of changes in meteorological conditions in May 2015. Furthermore, the wind field transport had the greatest effects...
on $\Delta M_{\text{ozone}}$ (positive) in northwestern, central and most parts of southern Shandong Province, whereas the meteorological factors affecting the photochemical generation of $O_3$ (e.g. temperature and CF) had the greatest effect on $\Delta M_{\text{ozone}}$ (negative) over Byland and a small part of southern Shandong Province in May 2015.

Combined with the results for $\Delta O_{\text{ozone}}$, it showed that changes in emissions dominated the variations in total $O_3$ concentrations in May 2015 over most regions of Shandong. For example, $\Delta O_{\text{ozone}}$ was positive and $\Delta M_{\text{ozone}}$ was negative at Dongying and therefore the final changes in the concentration of $O_3$ in May 2015 were dominated by the effects of emissions. As another example, both $\Delta O_{\text{ozone}}$ and $\Delta M_{\text{ozone}}$ were positive at Liaocheng, but $\Delta M_{\text{ozone}}$ contributed < 30% of $\Delta O_{\text{ozone}}$. Thus, the dominant effects were still from the changes in emissions. There were 10 such sites and most of the sites were in northwestern and central Shandong. While $O_3$ variations at other four sites (28.6%) were dominated by the changes of meteorological conditions.

In Fig. 5b, $\Delta M_{\text{ozone}}$ and $\Delta E_{\text{ozone}}$ in May between 2016 and 2015 varied from -34.1 to 43.2 $\mu g\, m^{-3}$ and from -47.6 to 123.3 $\mu g\, m^{-3}$, respectively. By contrast with 2015, the variations in meteorological conditions in May 2016 resulted in a decrease in $O_3$ (-34.1 to -4.6 $\mu g\, m^{-3}$) at most sites (except Qingdao, Weihai and Rizhao). Similarly, according to the analyzed results from Fig. 3 and 4, except the cloud fraction, the changes of temperature, relative humidity and wind field were conducive to the increase of $O_3$ at most sites over Shandong. We therefore concluded that the cloud fraction played a dominant role in the effects of changes in meteorological conditions.
on $O_3$ concentration through its impacts on the photochemical generation of $O_3$, especially the regions over northwestern, central and most parts of southern Shandong. Combined with the results for $\Delta O_{ozone}$, this showed that there were eight sites (57.1%) at which the variations in the concentration of $O_3$ were dominated by the changes in emissions. Compared with May 2015, the effects of meteorological conditions on the concentration of $O_3$ increased over Shandong Province in 2016 (6 sites, 42.9%).

Fig. 5c showed that $\Delta M_{ozone}$ and $\Delta E_{ozone}$ in May between 2017 and 2016 varied from -20.4 to 43.1 $\mu g \ m^{-3}$ and from -35.7 to 28.6 $\mu g \ m^{-3}$, respectively. The differences in meteorological conditions in May 2017 compared with May 2016 resulted in an increase in $O_3$ at most sites (7.6–43.1 $\mu g \ m^{-3}$) (except Weihai and Rizhao). Combined with the results for $\Delta O_{ozone}$, it was clear that the differences of meteorological conditions contributed more to the changes of $O_3$ concentration than that of emissions at most sites (11 sites, 78.6%) in May 2017. $\Delta M_{ozone}$ was positive at 10 of the 11 sites. According to the analyzed results from Fig. 3 and 4, except the relative humidity, the changes of temperature and cloud fraction had positive effects on $O_3$ formations at nearly all the sites. Since there were few differences in the effects of wind field transport in May between 2016 and 2017, it can be concluded that the meteorological parameters affecting the photochemical generation of $O_3$ dominated the increases in $O_3$ concentrations over most regions of Shandong Province in May 2017.

The effects of changes in meteorological conditions have become more and more important in the variation of $O_3$ concentrations from May 2014 to May 2017,
especially the factors that affected the photochemical generation of O$_3$.

NO$_x$ is one of the photochemical precursors of O$_3$. Previous studies (e.g. Wang et al., 2019a) presented that changes in the formation of O$_3$ were closely related to variations in NO$_x$ emissions over eastern China, thus we further investigated the effects of meteorological conditions and emissions on NO$_x$ concentrations, respectively.

![Graph](image.png)

**Fig. 6.** Changes in the monthly mean NO$_x$ concentrations ($\Delta$O$_{NO_x}$) in (a) May 2015 compared with May 2014, (b) May 2016 compared with May 2015, and (c) May 2017 compared with May 2016 as a result of changes in the meteorological conditions ($\Delta$M$_{NO_x}$) and emissions ($\Delta$E$_{NO_x}$).

Fig. 6 shows the changes in the monthly mean NO$_x$ concentrations ($\Delta$O$_{NO_x}$) in May between two adjacent years resulting from changes in the meteorological conditions ($\Delta$M$_{NO_x}$) and emissions ($\Delta$E$_{NO_x}$) during the time period 2014–2017.

As shown in Fig. 6a, the concentration of NO$_x$ decreased ($-33.9$ to $-0.7$ µg m$^{-3}$) at most sites in May 2015 compared with May 2014, which was dominated by the
effects of changes in NO_x emissions over most regions of Shandong (|∆E_{NO_x}|>|∆M_{NO_x}|
| at 10 sites). Combined with the analysis from Fig. 5a, ∆E_{ozone} dominated the changes
in total O_3 concentrations in May 2015. There were seven sites where both ∆E_{ozone}
and ∆E_{NO_x} contributed more to corresponding changes in the concentration of
pollutants (e.g. Dongying, Tai’an and Qingdao). By comparing ∆E_{ozone} and ∆E_{NO_x} at
the seven sites, the variation tendency was same at five sites between ∆E_{ozone} and
∆E_{NO_x}, suggesting that the variations in O_3 were closely related to the changes in NO_x
emissions in May 2015 compared to 2014.

As presented in Fig. 6b, the changes in NO_x emissions still dominated the variations
in NO_x concentrations at most sites (11 sites) in May 2016 compared to 2015.
Combined with the results shown in Fig. 5b, ∆E_{ozone} also dominated the variations in
O_3 formation in May 2016 compared with 2015. There were seven sites where both
∆E_{ozone} and ∆E_{NO_x} contributed more to corresponding changes in the concentration of
pollutants (e.g. Dongying, Zibo and Rizhao). By comparing ∆E_{ozone} and ∆E_{NO_x} at the
seven sites, the variation tendency was same at six sites between ∆E_{ozone} and ∆E_{NO_x},
suggesting that the variations in O_3 were still closely related to the changes in NO_x
emissions in May 2016 compared to 2015.

In Fig. 6c, NO_x concentrations decreased at most of the sites (except Heze) in May
2017 compared with 2016, which was still dominated by the changes in NO_x
emissions. However, combined with the results shown in Fig. 5c, the variations in
meteorological conditions that affected the photochemical generation of O_3 dominated
the changes of O_3 concentrations. Therefore, the variations in NO_x emissions may
have relatively small effects on the changes in O$_3$ concentrations.

In conclusion, the changes in NO$_x$ emissions had a close relationship with the variations in O$_3$ concentrations when the changes in emissions dominated the variations of O$_3$ concentrations in Shandong.

4. Conclusions

The changes of surface O$_3$ concentrations due to meteorological conditions and emissions in the late springtime (May) of Shandong during the time period 2014–2017 were discussed based on ambient measurements and simulations with the RAMS-CMAQ modeling system.

Based on the measurements, an increase was observed in O$_3$ pollution over Shandong during the time period 2014–2017. The peak period of O$_3$ pollution gradually changed from a broad (May–August) in 2014 to a narrow (May and June) range in 2017, while the observed monthly mean O$_3$ concentrations during the peak period changed from 90.1–91.7 µg m$^{-3}$ in 2014 to 112.3–117.8 µg m$^{-3}$ in 2017. There was a significant and unexpected increase of O$_3$ in May 2017 compared to 2016, indicated that more serious ozone pollution appeared in the late springtime over Shandong Province. Further to analyze the effects of each meteorological factor on O$_3$ concentrations in May between years, it was found that the changes in wind field were conducive to the accumulation of O$_3$, while the effects of other meteorological factors on O$_3$ concentrations were different at the same site between years.

In order to identify the effects of the changes in O$_3$ concentrations due to
meteorological conditions and emissions in May during the time period 2014–2017, four sensitivity simulations were designed and performed with RAMS-CMAQ. The simulation showed that the effects of changes in meteorological conditions became more and more important in the variations of O₃ concentrations in May during the time period 2014–2017, especially the factors that affected the photochemical generation of O₃. For instance, the percentage of the sites where O₃ variations were dominated by the changes of the meteorological conditions increased from 28.6% to 78.6% over the region in May between years. The effects of meteorological parameters on O₃ concentrations were different and were positive (11.8–35.2 µg m⁻³), negative (-34.1 to -4.6 µg m⁻³) and positive (7.6–43.1 µg m⁻³) at most sites in May between years. By analyzing the effects of the changes in NOₓ concentrations due to meteorological conditions and emissions, it was found that the changes in NOₓ emissions had a close relationship with the variations in O₃ concentrations when the changes of O₃ were dominated by the emission variations in Shandong.

Acknowledgments

This study was founded by the Strategic Priority Research Program (A) of the Chinese Academy of Sciences (Grant XDA19040204), the National Natural Science Foundation of China (41830109), the China Postdoctoral Science Foundation and the Key Research and Development Plan of Ningxia Hui Autonomous Region (Grants 2019BFG02025).
References


https://doi.org/10.1038/nclimate2317


