



# Weakened aerosol–radiation interaction exacerbating ozone pollution in eastern China since China’s clean air actions

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**Abstract.** Since China’s clean air action, PM<sub>2.5</sub> (particulate matter with an aerodynamic equivalent diameter of 2.5 µm or less) air quality has improved, while ozone (O<sub>3</sub>) pollution has become more severe. Here we apply a coupled meteorology–chemistry model (WRF-Chem: Weather Research and Forecasting model coupled to Chemistry v3.7.1) to quantify the responses of aerosol–radiation interaction (ARI) to anthropogenic emission reductions from 2013 to 2017, including aerosol–photolysis interaction (API) related to photolysis rate change and aerosol–radiation feedback (ARF) related to meteorological field change and their contributions to O<sub>3</sub> increases over eastern China in summer and winter. Sensitivity experiments show that the decreased anthropogenic emissions play a more prominent role in the increased daily maximum 8 h average (MDA8) O<sub>3</sub> in both summer (+1.96 ppb vs. +0.07 ppb) and winter (+3.56 ppb vs. −1.08 ppb) than the impacts of changed meteorological conditions in urban areas. The decreased PM<sub>2.5</sub> caused by emission reductions can result in a weaker impact of ARI on O<sub>3</sub> concentrations, which superimposes its effect on the worsened O<sub>3</sub> air quality. The weakened ARI due to decreased anthropogenic emissions aggravates the summer (winter) O<sub>3</sub> pollution by +0.81 ppb (+0.63 ppb), averaged over eastern China, with weakened API contributing 55.6 % (61.9 %) and ARF contributing 44.4 % (38.1 %), respectively. This superimposed effect is more significant for urban areas during summer (+1.77 ppb). Process analysis indicates that the enhanced chemical production is the dominant process for the increased O<sub>3</sub> concentrations caused by weakened ARI in both summer and winter. This study innovatively reveals the adverse effect of weakened aerosol–radiation interaction due to decreased anthropogenic emissions on O<sub>3</sub> air quality, indicating that more stringent coordinated air pollution control strategies should be implemented for significant improvements in future air quality.

## 1 Introduction

Since the implementation of clean air action in 2013, PM<sub>2.5</sub> (particulate matter with an aerodynamic equivalent diameter of 2.5 µm or less) concentrations have decreased significantly in China (Zhai et al., 2019; Zhang et al., 2019). However, ozone (O<sub>3</sub>) pollution is becoming worse and poses a significant challenge over eastern China, especially in the developed city clusters including Beijing–Tianjin–Hebei (BTH), the Yangtze River Delta (YRD), the Pearl River Delta (PRD), and the Sichuan Basin (SCB) (Lu et al., 2018; Dang and Liao, 2019; Li et al., 2019, 2021). According to observation data, Li et al. (2020) found that the daily maximum 8 h average O<sub>3</sub> concentrations (MDA8 O<sub>3</sub>) increased at a rate of 1.9 ppb a<sup>−1</sup> from 2013 to 2019 over eastern China. Elevated O<sub>3</sub> concentrations can not only decrease crop yield but also damage human health (Lelieveld et al., 2015; Yue et al., 2017; Mills et al., 2018). Therefore, it is essential to gain a comprehensive understanding of factors driving the increasing trend of O<sub>3</sub> in China in order to formulate effective prevention strategies.

As a secondary air pollutant, troposphere O<sub>3</sub> can be produced by nitrogen oxides (NO<sub>x</sub> = NO + NO<sub>2</sub>), carbon monoxide (CO), methane (CH<sub>4</sub>), and volatile organic compounds (VOCs) in the presence of solar radiation through photochemical reactions (Atkinson, 2000; Seinfeld and Pandis, 2006). The concentration of O<sub>3</sub> in the troposphere is influenced by changes in meteorological conditions (e.g., high temperature and low relative humidity) and its precursors emissions (e.g., NO<sub>x</sub> and VOCs) (Wang et al., 2019; Liu and Wang, 2020a, b; Shu et al., 2020). Most precursors are from anthropogenic sources, but some precursors can come from natural sources such as biogenic VOCs and soil and lightning NO<sub>x</sub>. Moreover, particulates can also affect O<sub>3</sub> concentrations through aerosol–radiation interaction (ARI), including aerosol–photolysis interaction (API) and aerosol–radiation feedback (ARF) (Liao et al., 1999; Wang et al., 2016; Zhu et al., 2021; Yang et al., 2022) as well as heterogeneous chemistry on the aerosol surface (Lou et al., 2014; Li et al., 2019; Liu and Wang, 2020b). Many studies have found that decreased PM<sub>2.5</sub> can be one of the driving factors contributing to increased O<sub>3</sub> concentrations (Li et al., 2019; Liu and Wang, 2020b; Shao et al., 2021). Li et al. (2019) analyzed GEOS-Chem simulation results and pointed out that the reductions in PM<sub>2.5</sub> concentrations from 2013 to 2017 over the North China Plain (NCP) could have decreased the sink of HO<sub>2</sub> on the aerosol surface, which would result in an increase in O<sub>3</sub> concentrations. When heterogeneous reactions were considered in WRF-CMAQ, Liu and Wang (2020b) found that decreased PM<sub>2.5</sub> concentrations weakened the uptake of reactive gases (mainly HO<sub>2</sub> and O<sub>3</sub>), which led to the increase in O<sub>3</sub> concentrations over China from 2013 to 2017. However, the contribution of weakened aerosol–radiation interaction to the increased O<sub>3</sub> due to substantial decreases in PM<sub>2.5</sub> under clean air action has not been systematically quantified. Furthermore, previous studies mainly focus on in-

creased summer O<sub>3</sub> (Li et al., 2019; Liu and Wang, 2020a, b; Shu et al., 2020; Shao et al., 2021), but underlying reasons driving the changes in winter O<sub>3</sub> are unclear. Li et al. (2021) pointed out that O<sub>3</sub> pollution has extended into cold seasons under the emission control measures. Therefore, this study aims to quantify the response of aerosol–radiation interaction to anthropogenic emission reduction from 2013 to 2017, with the main focus on the contribution to changed O<sub>3</sub> concentrations over eastern China in both summer and winter.

Aerosol–radiation interaction (ARI) can alter photolysis rates through aerosol–photolysis interaction (API) and meteorological variables through aerosol–radiation feedback (ARF) to influence the formation of O<sub>3</sub> (Yang et al., 2022). API can affect O<sub>3</sub> directly by reducing the photochemical reactions that weaken the chemical contribution and reduce the surface O<sub>3</sub> concentrations. ARF indirectly affects O<sub>3</sub> concentrations by altering meteorological variables, e.g., by reducing the height of the planetary boundary layer. The suppressed planetary boundary layer can weaken the vertical mixing of O<sub>3</sub> by turbulence and affect the concentration of O<sub>3</sub> precursors. Hong et al. (2020) used WRF-CMAQ in conjunction with future emission scenarios to find that weakened ARF due to reduced aerosol concentration had either negative or positive impacts on the daily maximum 1 h average O<sub>3</sub> concentration in eastern China from 2010 to 2050 due to the changed precursor level caused by the weakened ARF. Using WRF-CMAQ, Liu and Wang (2020b) reported that weakened API could increase the MDA8 O<sub>3</sub> concentrations by 0.3 ppb in urban areas from 2013 to 2017. Zhu et al. (2021) used WRF-Chem to investigate the impact of weakened ARF on air pollutants over the NCP during COVID-19 lockdown and reported that the weakened ARF increased the O<sub>3</sub> concentrations by 7.8 % due to the increased northwesterly and planetary boundary layer height caused by the weakened ARF. In general, previous studies mainly examined the impact of either weakened ARF or API; systematic analysis of the total and the respective impacts of changed API and/or ARF on O<sub>3</sub> over eastern China in both summer and winter from 2013 to 2017 has not been conducted.

The objective of this paper is to examine the impacts of aerosol–radiation interaction (ARI), including the effects of aerosol–photolysis interaction (API) and aerosol–radiation feedback (ARF), on O<sub>3</sub> concentrations over eastern China in both summer and winter using the online coupled WRF-Chem model, with the main focus on ARI, API, and ARF responses to clean air action. Process analysis is also applied to explore the prominent physical–chemical process responsible for the changed impacts of API and/or ARF on surface O<sub>3</sub>. This study is believed to provide insights into the role of weakened ARI in O<sub>3</sub> levels over eastern China not only in summer but also in winter. In Sect. 2, we describe the model configuration, numerical experiments, observational data, and integrated process rate analysis. Model evaluation is presented in Sect. 3. Results and discussion are presented in Sect. 4. Conclusions are provided in Sect. 5.

## 2 Methodology

### 2.1 Model configuration

The model used in this study is an online coupled meteorology–chemistry model, the Weather Research and Forecasting model coupled to Chemistry (WRF-Chem v3.7.1), which can simulate meteorological fields and concentrations of gases and aerosols simultaneously (Grell et al., 2005; Skamarock et al., 2008). Figure S1 in the Supplement shows the simulated domain that covers most regions of China with a horizontal resolution of 27 km and grid points of 167 (west–east)  $\times$  167 (south–north). The model contains 32 vertical levels extending from the surface to 50 hPa, with the first 16 layers located below 2 km to resolve fine boundary layer processes. The black line in Fig. S1 represents eastern China (22–41.5° N, 102–123° E), and four heavily polluted regions are also selected for analysis, including BTH (36.0–41.5° N, 113–119.5° E), YRD (29.5–32.5° N, 118–122° E), PRD (21–23.5° N, 112–116° E), and SCB (27.5–31.5° N, 102.5–107.5° E).

The National Center for Environmental Prediction (NCEP) Final (FNL) analysis dataset, with a spatial resolution of 1°  $\times$  1° and 6 h temporal resolution, is used to provide the meteorological initial and lateral boundary conditions. The chemical initial and boundary conditions for the WRF-Chem model are taken from the outputs of the Community Atmosphere Model with Chemistry (CAM-Chem).

The Carbon Bond Mechanism Z (CBM-Z) is applied as the gas-phase chemical mechanism (Zaveri and Peters, 1999), and the full eight-bin MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) module with aqueous chemistry is used to simulate aerosol evolution (Zaveri et al., 2008). In the MOSAIC module, aerosols are assumed to be internally mixed into eight bins (0.039–0.078, 0.078–0.156, 0.156–0.312, 0.312–0.625, 0.625–1.25, 1.25–2.5, 2.5–5.0, and 5.0–10  $\mu$ m), and each bin considers all major aerosol species, such as sulfate ( $\text{SO}_4^{2-}$ ), nitrate ( $\text{NO}_3^-$ ), ammonium ( $\text{NH}_4^+$ ), black carbon (BC), organic carbon (OC), and other inorganic mass. The impacts of aerosols on photolysis rates are calculated using the Fast-J scheme (Wild et al., 2000). The following physical parameterizations are used in WRF-Chem. The rapid radiative transfer model for general circulation models (RRTMG) is used to treat both shortwave and longwave radiation in the atmosphere (Iacono et al., 2008). The Purdue Lin microphysics scheme (Lin et al., 1983) and the Grell 3D ensemble scheme (Grell, 1993) are used to describe the cloud microphysical processes and cumulus convective processes. The Noah land surface scheme (Chen and Dudhia, 2001) and the Monin–Obukhov surface scheme (Foken, 2006) are used to simulate land–atmosphere interactions. The planetary boundary layer is characterized by the Yonsei University PBL scheme (Hong et al., 2006). The main physical and chemical schemes used in this study are summarized in Table S1 in the Supplement.

In this study, the Multi-resolution Emission Inventory model for Climate and air pollution research (MEIC; <http://www.meicmodel.org/>, last access: 21 March 2024) data from 2013 and 2017 are used as the anthropogenic emissions of particles and gases (Zheng et al., 2018). Biogenic emissions are calculated using the online Model of Emissions of Gases and Aerosols from Nature (MEGAN) developed by Guenther et al. (2006).

### 2.2 Numerical experiments

Seven sensitivity experiments were designed (Table 1). Here are the detailed descriptions.

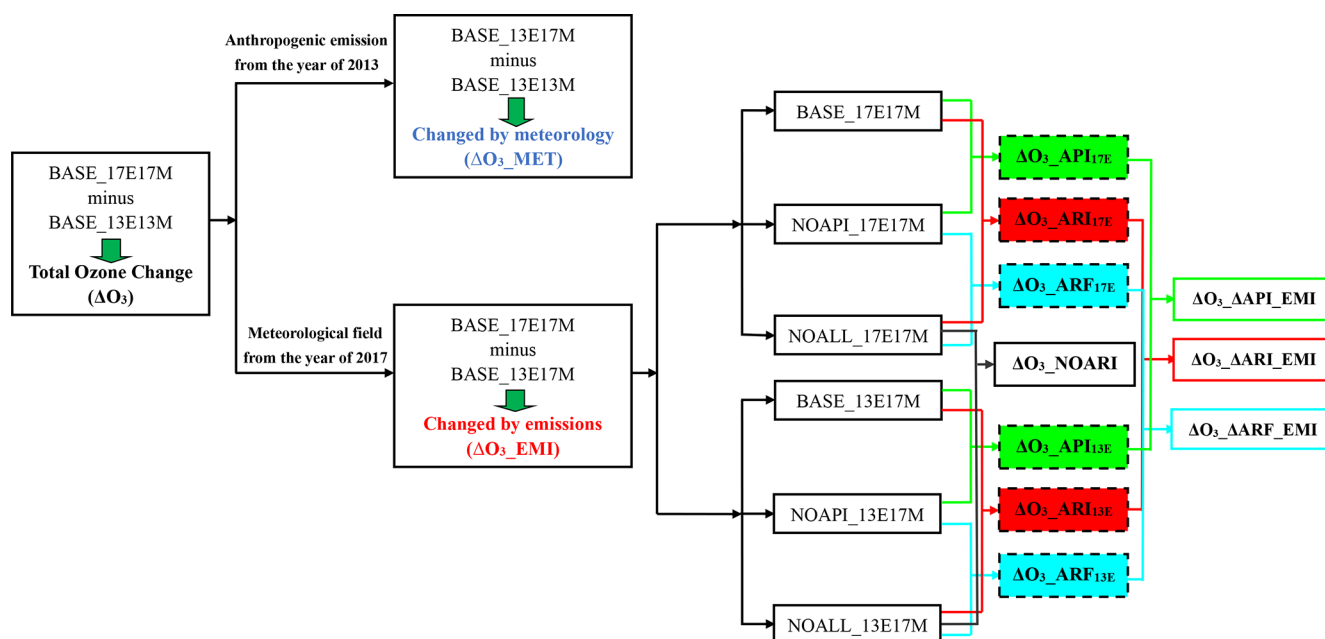
1. BASE\_17E17M is the baseline experiment coupled with the interactions between aerosol and radiation, which include the impacts of API and ARF. Both the meteorological field and anthropogenic emissions are from 2017.
2. BASE\_13E13M is the same as BASE\_17E17M, but the meteorological field and anthropogenic emissions are from 2013.
3. NOAPI\_17E17M is the same as BASE\_17E17M, but the impact of API is not considered by turning off the aerosol effect in the photolysis module, following the method described in Yang et al. (2022).
4. NOALL\_17E17M is the same as BASE\_17E17M, but neither the impact of API nor that of ARF is considered by setting the aerosol optical properties in the optical module to zero, following the method described in Yang et al. (2022).
5. BASE\_13E17M is the same as BASE\_17E17M, but the anthropogenic emissions are from 2013.
6. NOAPI\_13E17M is the same as NOAPI\_17E17M, but the anthropogenic emissions are from 2013.
7. NOALL\_13E17M is the same as NOALL\_17E17M, but the anthropogenic emissions are from 2013.

Figure 1 presents the detailed schematic overview of designed numerical experiments. As shown in Fig. 1, the differences between BASE\_17E17M and BASE\_13E13M (BASE\_17E17M minus BASE\_13E13M) represent the changed  $\text{O}_3$  ( $\Delta\text{O}_3$ ) due to variations in meteorology and anthropogenic emissions from 2013 to 2017. The differences between BASE\_13E17M and BASE\_13E13M (BASE\_13E17M minus BASE\_13E13M) show the impact of changed meteorological conditions on  $\text{O}_3$  ( $\Delta\text{O}_{3\_MET}$ ) from 2013 to 2017. The differences between BASE\_17E17M and BASE\_13E17M (BASE\_17E17M minus BASE\_13E17M) indicate the impact of anthropogenic emission reductions on  $\text{O}_3$  ( $\Delta\text{O}_{3\_EMI}$ ) from 2013 to 2017.

**Table 1.** Descriptions of model sensitivity experiments.

| Cases        | Anthropogenic emissions | Meteorological field | API* | ARF* |
|--------------|-------------------------|----------------------|------|------|
| BASE_17E17M  | 2017                    | 2017                 | On   | On   |
| BASE_13E13M  | 2013                    | 2013                 | On   | On   |
| NOAPI_17E17M | 2017                    | 2017                 | Off  | On   |
| NOALL_17E17M | 2017                    | 2017                 | Off  | Off  |
| BASE_13E17M  | 2013                    | 2017                 | On   | On   |
| NOAPI_13E17M | 2013                    | 2017                 | Off  | On   |
| NOALL_13E17M | 2013                    | 2017                 | Off  | Off  |

\* API means aerosol–photolysis interaction; ARF means aerosol–radiation feedback.



**Figure 1.** Schematic overview of numerical experiments. The term 17E17M (13E13M) means that meteorological fields and anthropogenic emissions are from 2017 (2013). Similarly, 13E17M means anthropogenic emissions are from 2013 but meteorological fields are from 2017.  $\Delta O_3\_MET$ ,  $\Delta O_3\_EMI$ , and  $\Delta O_3$  mean the impacts of changed meteorological conditions, changed anthropogenic emissions, and their combined effects on  $O_3$ , respectively.  $\Delta O_3\_API_{17E(13E)}$ ,  $\Delta O_3\_ARF_{17E(13E)}$ , and  $\Delta O_3\_ARI_{17E(13E)}$  mean the impacts of aerosol–photolysis interaction, aerosol–radiation feedback, and aerosol–radiation interaction on  $O_3$  under the different emission conditions, respectively.  $\Delta O_3\_NOARI$  means the changed  $O_3$  concentration due to reduced anthropogenic emissions without considering aerosol–radiation interaction.  $\Delta O_3\_ΔAPI\_EMI$ ,  $\Delta O_3\_ΔARF\_EMI$ , and  $\Delta O_3\_ΔARI\_EMI$  represent the impacts of weakened aerosol–photolysis interaction, aerosol–radiation feedback, and aerosol–radiation interaction due to decreased anthropogenic emissions on  $O_3$  concentration, respectively.

The impacts of aerosol–radiation interaction (ARI) on  $O_3$  under different anthropogenic emission scenarios (i.e., strong anthropogenic emission levels in 2013 and weaker anthropogenic emission levels in 2017) can be analyzed as the differences between BASE\_17E17M and NOALL\_17E17M (BASE\_17E17M minus NOALL\_17E17M, denoted as  $\Delta O_3\_ARI_{17E}$ ) and BASE\_13E17M and NOALL\_13E17M (BASE\_13E17M minus NOALL\_13E17M, denoted as  $\Delta O_3\_ARI_{13E}$ ). The term  $\Delta O_3\_ARI_{17E}$  means that the impact of ARI on  $O_3$  under the conditions of both the meteorological field and anthropogenic emissions is from 2017,

and  $\Delta O_3\_ARI_{13E}$  means the effect of ARI on  $O_3$  with the meteorological field used in 2017 and anthropogenic emissions applied in 2013. In order to quantify the impacts caused by the decreased anthropogenic emissions from 2013 to 2017, the impacts of changed meteorological variables should be removed by fixing the meteorological fields in 2017 using sensitivity experiments. Thus, the impact of weakened ARI due to decreased anthropogenic emissions from 2013 to 2017 on  $O_3$  (denoted as  $\Delta O_3\_ΔARI\_EMI$ ) can be quantified using the differences between  $\Delta O_3\_ARI_{17E}$  and  $\Delta O_3\_ARI_{13E}$ . Similarly, the impacts of weakened

API (denoted as  $\Delta O_3\_API\_EMI$ ) and ARF (denoted as  $\Delta O_3\_ARF\_EMI$ ) due to decreased anthropogenic emissions on  $O_3$  can also be estimated using the differences between BASE\_17E17M minus NOAPI\_17E17M (denoted as  $\Delta O_3\_API_{17E}$ ) and BASE\_13E17M minus NOAPI\_13E17M (denoted as  $\Delta O_3\_API_{13E}$ ), as well as between NOAPI\_17E17M minus NOALL\_17E17M (denoted as  $\Delta O_3\_ARF_{17E}$ ) and NOAPI\_13E17M minus NOALL\_13E17M (denoted as  $\Delta O_3\_ARF_{13E}$ ). Detailed descriptions can be found in Fig. 1.

Simulation periods are integrated from 30 May to 30 June (denoted as summer) and 29 November to 31 December (denoted as winter) in both 2013 and 2017. To avoid potential deviations caused by long-term model integration, each simulation is re-initialized every 8 d, with the first 40 h as the model spin-up. The complete simulation includes five model cycles. Simulation results from the BASE\_17E17M case during summer and winter are used to evaluate the model performance. If not otherwise specified, the time in this paper is the local time, and the synergetic impacts of ARF and API are equal to the impact of ARI (i.e.,  $ARI = ARF + API$ ).

### 2.3 Observational data

Meteorological observations of temperature ( $T_2$ ), relative humidity ( $RH_2$ ), wind speed ( $WS_{10}$ ), and wind direction ( $WD_{10}$ ) provided by the NOAA National Climatic Data Center (<https://www.ncdc.noaa.gov/>, last access: 21 March 2024) were used to validate the meteorological performance of the model. In this study, 353 meteorological stations are selected, and the locations are shown as red dots in Fig. S1. Observed surface  $PM_{2.5}$ ,  $O_3$ , and  $NO_2$  concentrations in eastern China are obtained from the China National Environmental Monitoring Centre, which can be downloaded from <https://quoteoft.net/air/> (last access: 21 March 2024). To ensure the data quality, a single site with at least 500 actual observations during the simulated period was used for model evaluation. A total of 1296 sites, as shown in Fig. 2a, were obtained. Photolysis rates of nitrogen dioxide ( $NO_2$ ) ( $J[NO_2]$ ) measured at the Peking University site (39.99° N, 116.31° E) were also used to evaluate the model performance.

### 2.4 Integrated process rate analysis

Process analysis techniques, i.e., integrated process rate (IPR) analysis, can be used in grid-based Eulerian models (e.g., WRF-Chem) to obtain contributions of each physical–chemical process to variations in pollutant concentrations. Eulerian models utilize the numerical technique of operator splitting to resolve continuity equations for each species into several simple ordinary differential equations or partial differential equations that only contain the influence of one or two processes (Gipson, 1999).

In order to quantitatively elucidate individual contributions of physical and chemical processes to  $O_3$  concentra-

tion changes due to weakened ARI, the integrated process rate (IPR) methodology is applied in this study. IPR analysis is an advanced tool to evaluate the key process for  $O_3$  concentration variation (Shu et al., 2016; Zhu et al., 2021; Yang et al., 2022). In this study, the IPR analysis tracks the hourly (e.g., one time step) contribution to  $O_3$  concentration variation from four main processes: vertical mixing (VMIX), net chemical production (CHEM), horizontal advection (ADVH), and vertical advection (ADVZ). VMIX is initiated by a turbulent process and is closely related to PBL development, which influences  $O_3$  vertical gradients. CHEM represents the net  $O_3$  chemical production (chemical production minus chemical consumption). ADVH and ADVZ represent transport by wind. We define ADV as the sum of ADVH plus ADVZ.

## 3 Model evaluation

Simulation results of BASE\_17E17M are compared to the observations to evaluate the model performance before interpreting the impacts of aerosol–radiation interaction on surface-layer ozone concentration.

### 3.1 Evaluation for meteorology

Figure S2 shows the time series of observed and simulated  $T_2$ ,  $RH_2$ ,  $WS_{10}$ , and  $WD_{10}$  averaged over the 353 meteorological stations in China during summer and winter in 2017. Statistical performance of simulated meteorological parameters compared to ground-based observations is shown in Table 2. Simulations track well with observed  $T_2$ , with a correlation coefficient ( $R$ ) of 0.99 and 0.92, but underestimate  $T_2$  with a mean bias (MB) of  $-1.0$  and  $-2.0$  K in summer and winter, respectively. Simulated  $RH_2$  agrees reasonably well with observations, with  $R$  of 0.97 and 0.87, and small normalized mean biases (NMBs) are found in summer and winter with values of 3.2 % and 3.5 %, respectively.  $WS_{10}$  is slightly overpredicted, with an MB of  $1.6$ – $2.1$  m s $^{-1}$ . The  $R$  and root mean square error (RMSE) of  $WS_{10}$  are 0.77–0.82 and  $1.6$ – $2.1$  m s $^{-1}$ . A large bias in wind speed could be partly caused by unresolved topographical features (Jimenez and Dudhia, 2012). The NMB of  $WD_{10}$  ranges from  $-3.9$  % to  $-2.6$  % and  $R$  ranges from 0.40 to 0.69. As shown in Fig. S3, the predicted  $J[NO_2]$  matches the observations well, with  $R$  of 0.93–0.94 and NMB of 4.8 %–12.3 %. In general, the simulated meteorological variables agree fairly well with the observations.

### 3.2 Evaluation for air pollutants

Figure 2 shows the spatiotemporal variations in observed and simulated near-surface  $PM_{2.5}$ ,  $O_3$ , and  $NO_2$  concentrations averaged over eastern China during summer and winter in 2017. As demonstrated in Fig. 2a1 and c1, the WRF-Chem model reproduces the spatial distribution of observed

**Table 2.** Statistical parameters of the simulated 2 m temperature ( $T_2$ , K), 2 m relative humidity ( $RH_2$ , %), 10 m wind speed ( $WS_{10}$ ,  $\text{m s}^{-1}$ ), 10 m wind direction ( $WD_{10}$ , °), photolysis rate of  $\text{NO}_2$  ( $J[\text{NO}_2]$ ,  $10^{-3} \text{ s}^{-1}$ ),  $\text{PM}_{2.5}$  ( $\mu\text{g m}^{-3}$ ),  $\text{O}_3$  (ppb), and  $\text{NO}_2$  (ppb) versus observations during summer and winter in 2017. There are 1296 air pollutant monitoring stations and 353 meteorological stations.

| Variable          | Summer |       |       |               |                    |                 | Winter |       |       |               |                    |                 |
|-------------------|--------|-------|-------|---------------|--------------------|-----------------|--------|-------|-------|---------------|--------------------|-----------------|
|                   | $O^a$  | $M^a$ | $R^b$ | $\text{MB}^c$ | $\text{NMB}^d$ (%) | $\text{RMSE}^e$ | $O^a$  | $M^a$ | $R^b$ | $\text{MB}^c$ | $\text{NMB}^d$ (%) | $\text{RMSE}^e$ |
| $T_2$             | 295.3  | 294.2 | 0.99  | −1.0          | −3.2               | 1.0             | 275.0  | 272.8 | 0.92  | −2.0          | −74.1              | 2.5             |
| $RH_2$            | 68.1   | 71.0  | 0.97  | 2.2           | 3.2                | 3.6             | 58.1   | 60.6  | 0.87  | 2.1           | 3.5                | 6.5             |
| $WS_{10}$         | 2.6    | 4.2   | 0.77  | 1.6           | 61.6               | 1.6             | 2.6    | 4.7   | 0.82  | 2.1           | 83.2               | 2.1             |
| $WD_{10}$         | 175.7  | 170.9 | 0.40  | −4.6          | −2.6               | 16.9            | 192.6  | 184.6 | 0.69  | −7.5          | −3.9               | 17.4            |
| $J[\text{NO}_2]$  | 2.6    | 2.7   | 0.93  | 0.1           | 4.8                | 1.2             | 1.0    | 1.2   | 0.94  | 0.1           | 12.3               | 0.6             |
| $\text{PM}_{2.5}$ | 31.0   | 24.8  | 0.63  | −6.3          | −20.2              | 8.3             | 69.0   | 58.9  | 0.80  | −10.1         | −14.6              | 15.6            |
| $\text{O}_3$      | 39.7   | 38.9  | 0.90  | −0.6          | −1.6               | 6.9             | 17.7   | 20.5  | 0.86  | 2.8           | 15.7               | 5.0             |
| $\text{NO}_2$     | 12.7   | 11.2  | 0.73  | −1.5          | −12.0              | 4.5             | 23.3   | 18.7  | 0.83  | −4.5          | −19.4              | 5.6             |

<sup>a</sup>  $O$  and  $M$  are the averages for observed and simulated results, respectively.  $O = \frac{1}{n} \times \sum_{i=1}^n O_i$ ,  $M = \frac{1}{n} \times \sum_{i=1}^n M_i$ .

<sup>b</sup>  $R$  is the correlation coefficient between observations and model results.  $R = \frac{\sum_{i=1}^n (O_i - O) \times (M_i - M)}{\sqrt{\sum_{i=1}^n (O_i - O)^2 + \sum_{i=1}^n (M_i - M)^2}}$ .

<sup>c</sup>  $\text{MB}$  is the mean bias between observations and model results.  $\text{MB} = \frac{1}{n} \times \sum_{i=1}^n (M_i - O_i)$ .

<sup>d</sup>  $\text{NMB}$  is the normalized mean bias between observations and model results.  $\text{NMB} = \frac{1}{n} \times \sum_{i=1}^n \frac{M_i - O_i}{O_i} \times 100\%$ .

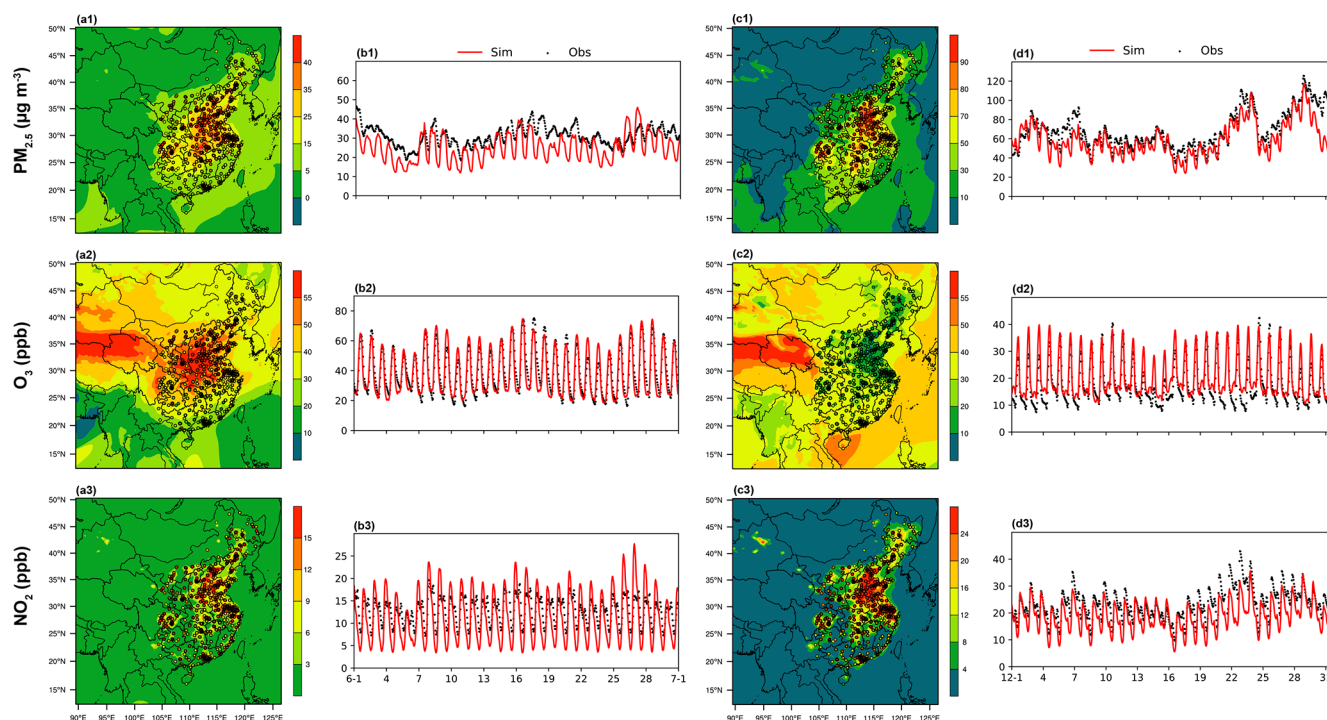
<sup>e</sup>  $\text{RMSE}$  is the root mean square error in observations and model results.  $\text{RMSE} = \sqrt{\frac{1}{n} \times \sum_{i=1}^n (M_i - O_i)^2}$ .

In the above  $O_i$  and  $M_i$  are the hourly observed data and simulated data, respectively, and  $n$  is the total number of hours.

$\text{PM}_{2.5}$  reasonably well, with high values over large city clusters. The predicted  $\text{O}_3$  concentrations can also reproduce the spatial variation in the observed concentrations (Fig. 2a2 and c2).  $\text{NO}_2$  is an important precursor of  $\text{O}_3$  and aerosol; thus a good performance for  $\text{NO}_2$  is necessary. From Fig. 2a3 and c3, the model can reproduce the spatial distribution of observed  $\text{NO}_2$  well. Although the distributions of simulated air pollutants are in good agreement with the observations, biases still exist, which may be due to the uncertainty in the emission inventories. Figure 2b1–b3 and d1–d3 show the temporal profiles of observed and simulated surface-layer air pollutants averaged over monitoring sites and the grid cell containing the monitoring site in eastern China. The statistical metrics are also shown in Table 2. As shown in Fig. 2b1 and d1, the model tracks well with the diurnal variation in  $\text{PM}_{2.5}$  over eastern China, with  $R$  of 0.63 and 0.80. However, the model slightly underestimates the concentrations of  $\text{PM}_{2.5}$ , with  $\text{MB}$  of  $-6.3$  and  $-10.1 \mu\text{g m}^{-3}$  in summer and winter, respectively. Simulated  $\text{O}_3$  agrees reasonably well with observations, with  $R$  of 0.90 and 0.86, and small  $\text{MB}$  is found in summer and winter with values of  $-0.6$  and  $2.8$  ppb, respectively. The model tracks the daily variation in observed  $\text{NO}_2$  reasonably well, with  $R$  of 0.73 and 0.83. However, the model slightly underestimates the  $\text{NO}_2$  versus measurements, with  $\text{MB}$  of  $-1.5$  and  $-4.5$  ppb in summer and winter, respectively. In general, the WRF-Chem model can reproduce the features of observed meteorology and air pollutants well over eastern China.

### 3.3 Evaluation for changes in air pollutants from 2013 to 2017

Figure 3 demonstrates the spatial distribution of changed summer (left) and winter (right) surface  $\text{PM}_{2.5}$  (panels a, b) and MDA8  $\text{O}_3$  (panels c, d) from 2013 to 2017. As shown in Fig. 3a and b, the observed concentrations of  $\text{PM}_{2.5}$  in eastern China are significantly reduced in both summer ( $-16.2 \mu\text{g m}^{-3}$ ) and winter ( $-56.0 \mu\text{g m}^{-3}$ ), and these changes can be captured by the model well ( $-14.3 \mu\text{g m}^{-3}$  for summer and  $-49.8 \mu\text{g m}^{-3}$  for winter). Therefore, the model can reproduce the observed decrease in  $\text{PM}_{2.5}$  levels from 2013 to 2017. As shown in Fig. 3c and d, the model reproduces the seasonal patterns of changed surface MDA8  $\text{O}_3$  reasonably well over eastern China during summer and winter from 2013 to 2017. In summer, both the observations and simulations showed the increased (decreased) MDA8  $\text{O}_3$  in YRD (PRD and SCB), while the model could not simulate the positive changes in MDA8  $\text{O}_3$  over BTH; the potential reason may be that this study did not consider the effect of changes in heterogeneous aerosol reactions. Li et al. (2019) found that the weakened uptake of  $\text{HO}_2$  on aerosol surfaces was the main reason for the  $\text{O}_3$  increase over BTH. In contrast to the changes in summer, observed MDA8  $\text{O}_3$  in winter generally increased over eastern China, which can be reproduced well by the model.



**Figure 2.** Spatial distributions of observed (circle) and simulated (colored)  $\text{PM}_{2.5}$ ,  $\text{O}_3$ , and  $\text{NO}_2$  concentrations averaged over (a1–a3) summer and (c1–c3) winter in 2017. Time series of observed (black dots) and simulated (red lines) hourly  $\text{PM}_{2.5}$ ,  $\text{O}_3$ , and  $\text{NO}_2$  concentrations averaged over all the observation sites in eastern China during (b1–b3) summer and (d1–d3) winter in 2017.

## 4 Results and discussion

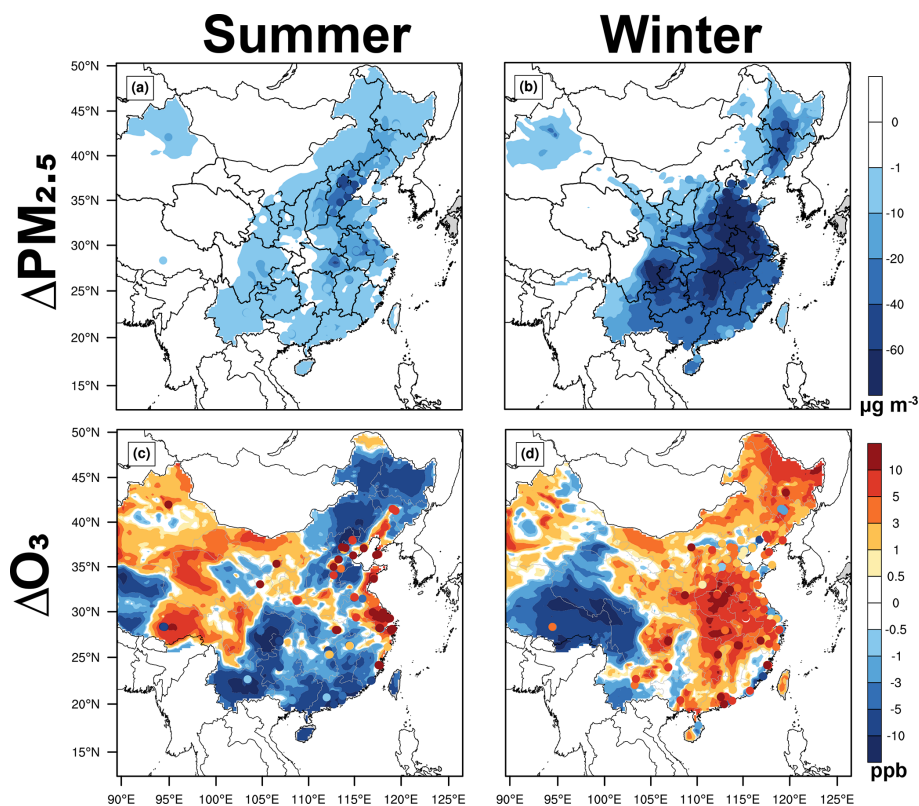
### 4.1 Impacts of changed meteorology and anthropogenic emissions on $\text{O}_3$

The strategy of the clean air action decreased the anthropogenic emission of  $\text{NO}_x$ , but the changes in anthropogenic VOC emissions were non-obvious (Fig. S4), which might influence the sensitive  $\text{O}_3$  formation regime and the  $\text{O}_3$  concentration. Figure 4 shows the spatial distributions of changed summer and winter MDA8  $\text{O}_3$  concentrations from 2013 to 2017 due to changed anthropogenic emissions alone and due to changed meteorological conditions alone. As shown in Fig. 4a, the concentration of summer MDA8  $\text{O}_3$  from 2013 to 2017 increased in city clusters but decreased in rural regions. This discrepancy might be explained by the ozone formation regimes in urban areas typically being VOC-limited, while rural areas are typically  $\text{NO}_x$ -limited during summer (Li et al., 2019; Wang et al., 2019). Contrary to the phenomenon in summer, decreased anthropogenic emissions lead to a uniform increase in winter MDA8  $\text{O}_3$  over the whole of eastern China (Fig. 4c). These different spatial variation characteristics in summer and winter may be explained by the different ozone formation regimes in winter (VOC-limited) and summer ( $\text{NO}_x$ -limited) (Fig. S5; Jin and Holloway, 2015). From Fig. 4b and d, the impacts of changed meteorological conditions on MDA8  $\text{O}_3$  varied by region, ranging from  $-24.9$

( $-14.0$ ) to  $17.0$  ( $7.3$ ) ppb in summer (winter). Focusing on the four developed city clusters, compared to 2013, the meteorological conditions in the summer of 2017 promoted the generation of  $\text{O}_3$  in the YRD region (Fig. 8a3) but suppressed the generation of  $\text{O}_3$  in the BTH (Fig. 8a2), PRD (Fig. 8a4), and SCB (Fig. 8a5) regions. In PRD and SCB, the changes in MDA8  $\text{O}_3$  due to meteorology have an even greater impact than those due to emission changes, which highlights the significant role of meteorology in summer  $\text{O}_3$  variations.

### 4.2 Impacts of weakened aerosol–radiation interaction on $\text{O}_3$

Figures S6a (S7a) and S6b (S7b) present the spatial distribution of the impacts of ARF, API, and ARI on surface MDA8  $\text{O}_3$  concentrations in summer (winter) under different anthropogenic emission conditions in 2017 and 2013. As shown in Fig. S6, summer MDA8  $\text{O}_3$  is significantly reduced over eastern China: ARF, API, and ARI decrease the surface MDA8  $\text{O}_3$  concentrations by  $0.23$  ( $0.59$ ),  $1.09$  ( $1.54$ ), and  $1.32$  ( $2.13$ ) ppb, respectively, under low (high) anthropogenic emission conditions in 2017 (2013). The changes in MDA8  $\text{O}_3$  concentrations due to aerosol–radiation interaction under low emission conditions are weaker than those under high emission conditions. This is because the concentration of aerosols in 2013 was higher than that in 2017, and thus its impact on meteorological conditions and  $J[\text{NO}_2]$



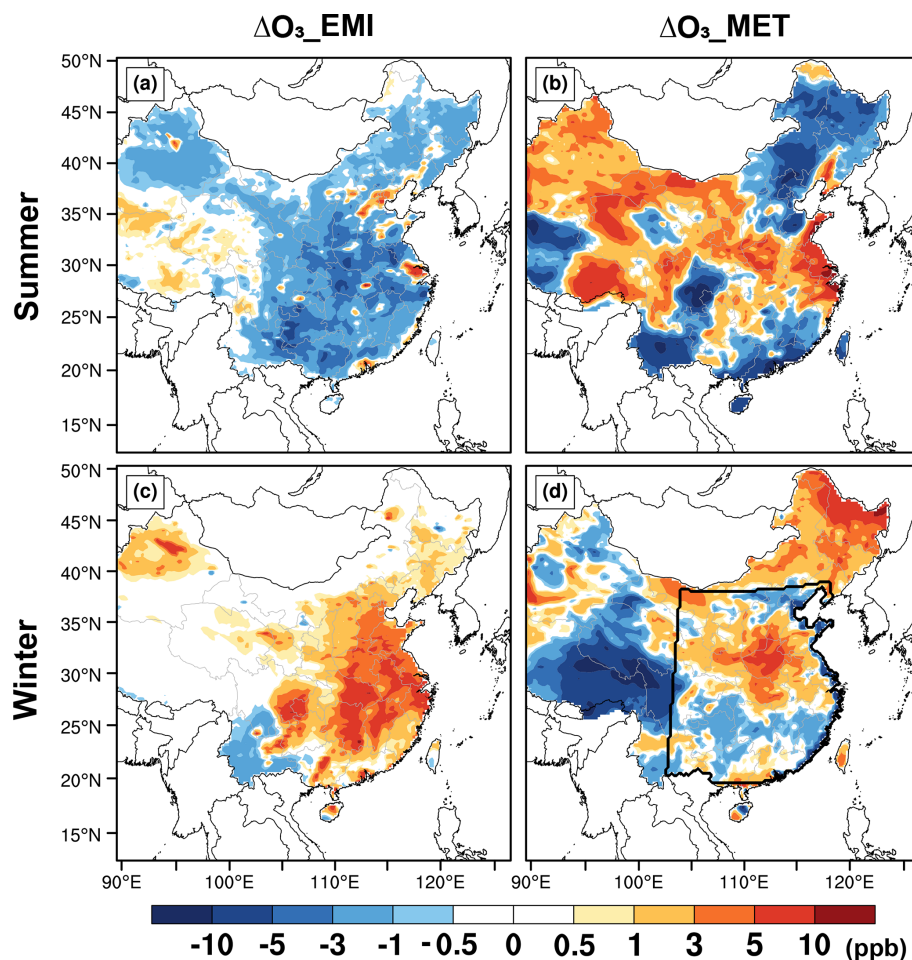
**Figure 3.** Spatial distribution of changed summer (a, c) and winter (b, d) surface (a, b)  $\text{PM}_{2.5}$  and (c, d) MDA8  $\text{O}_3$  from 2013 to 2017. Observed changes in surface  $\text{PM}_{2.5}$  MDA8  $\text{O}_3$  are also marked with colored circles.

was greater (Fig. S8). As shown in Fig. S7a, 2017 winter MDA8  $\text{O}_3$  concentrations were decreased by ARF, API, and ARI by 0.38 ppb (−0.9 %), 1.59 ppb (−4.1 %), and 1.96 ppb (−5.1 %), respectively. Compared to the impacts under relatively high anthropogenic emission conditions in 2013, the reduction in surface MDA8  $\text{O}_3$  concentrations caused by ARF, API, and ARI is also greater, with values of 0.62 ppb (−1.6 %), 1.98 ppb (−5.4 %), and 2.59 ppb (−7.1 %), respectively. Both API and ARF reduce  $\text{O}_3$  concentrations, and the reduction in  $\text{O}_3$  caused by API is greater than that caused by ARF in both summer and winter.

Further, the significant reduction in  $\text{PM}_{2.5}$  due to the clean air action (Fig. S9) led to an increase in  $\text{O}_3$  concentrations as there were weakened effects of aerosols on  $\text{O}_3$ . Therefore, this study further quantifies the effects of  $\Delta\text{O}_3_{\Delta\text{ARF\_EMI}}$ ,  $\Delta\text{O}_3_{\Delta\text{API\_EMI}}$ , and  $\Delta\text{O}_3_{\Delta\text{ARI\_EMI}}$  ( $\Delta\text{O}_3_{\Delta\text{ARI\_EMI}} = \Delta\text{O}_3_{\Delta\text{ARF\_EMI}} + \Delta\text{O}_3_{\Delta\text{API\_EMI}}$ ) on  $\text{O}_3$  air quality. As shown in Fig. 5a1–a3, the surface MDA8  $\text{O}_3$  in summer increased over most of eastern China due to  $\Delta\text{O}_3_{\Delta\text{ARF\_EMI}}$ ,  $\Delta\text{O}_3_{\Delta\text{API\_EMI}}$ , and  $\Delta\text{O}_3_{\Delta\text{ARI\_EMI}}$ . The largest increases in MDA8  $\text{O}_3$  concentrations due to  $\Delta\text{O}_3_{\Delta\text{ARF\_EMI}}$  and  $\Delta\text{O}_3_{\Delta\text{API\_EMI}}$  were found in the four developed city clusters, with increases larger than 4 ppb. Overall,  $\Delta\text{O}_3_{\Delta\text{ARF\_EMI}}$ ,

$\Delta\text{O}_3_{\Delta\text{API\_EMI}}$ , and  $\Delta\text{O}_3_{\Delta\text{ARI\_EMI}}$  led to increases in surface MDA8  $\text{O}_3$  by 0.36, 0.45, and 0.81 ppb, respectively, averaged over eastern China during summer. As shown in Fig. 5b1–b3, the  $\Delta\text{O}_3_{\Delta\text{ARF\_EMI}}$ ,  $\Delta\text{O}_3_{\Delta\text{API\_EMI}}$ , and  $\Delta\text{O}_3_{\Delta\text{ARI\_EMI}}$  can also cause an increase of 0.24, 0.39, and 0.63 ppb, respectively, in winter MDA8  $\text{O}_3$  concentrations. In general, weakened aerosol–radiation interaction due to reduced anthropogenic emissions from 2013 to 2017 can exacerbate ozone pollution in both summer and winter.

In order to explore the mechanism of the impacts of  $\Delta\text{O}_3_{\Delta\text{ARI\_EMI}}$  on MDA8  $\text{O}_3$ , we resolve the changed  $\text{O}_3$  into the contributions from chemical and physical processes. Figure 6 presents the accumulated changes in  $\text{O}_3$  and each process contribution by  $\Delta\text{O}_3_{\Delta\text{API\_EMI}}$ ,  $\Delta\text{O}_3_{\Delta\text{ARF\_EMI}}$ , and  $\Delta\text{O}_3_{\Delta\text{ARI\_EMI}}$  from 09:00 to 16:00 LST during summer and winter. As shown in Fig. 6, enhanced chemical production is the dominant process leading to the increase in  $\text{O}_3$  concentrations over eastern China and the four city clusters in both summer and winter. The leading factor of enhancement in  $\text{O}_3$  over BTH is inconsistent with that over eastern China, and the enhancement of  $\text{O}_3$  concentration over BTH is mainly due to  $\Delta\text{O}_3_{\Delta\text{ARF\_EMI}}$ . But the leading factor of enhancement in  $\text{O}_3$  over SCB is consistent with that over eastern China; the enhancement of  $\text{O}_3$  concentration is mainly due to  $\Delta\text{O}_3_{\Delta\text{API\_EMI}}$  in



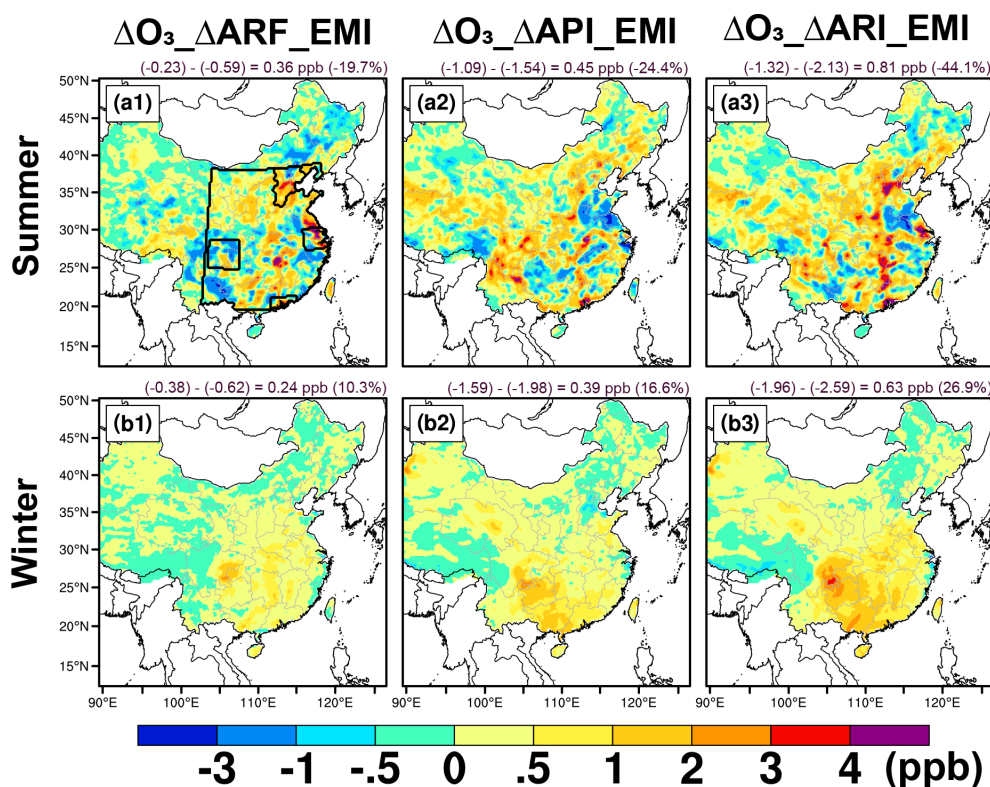
**Figure 4.** Spatial distribution of changed summer (a, b) and winter (c, d) surface-layer MDA8 O<sub>3</sub> from 2013 to 2017 due to (a, c) changed anthropogenic emissions alone and (b, d) changed meteorological fields alone. The thick black line in panel (d) represents eastern China.

both summer and winter. Moreover, the enhancement of O<sub>3</sub> concentration in BTH, YRD, and PRD is mainly due to  $\Delta O_3\_ \Delta ARF\_EMI$  during winter, which is opposite to that of eastern China. The leading factors for the increase in O<sub>3</sub> concentration in different city clusters are different. The enhancement of O<sub>3</sub> concentration in most areas was caused by  $\Delta O_3\_ \Delta API\_EMI$ , whereas the increase in O<sub>3</sub> concentration in the BTH, YRD, and PRD areas is dominated by  $\Delta O_3\_ \Delta ARF\_EMI$  in winter. In general, the weakened aerosol–radiation interaction caused by emission reduction promotes the chemical production of O<sub>3</sub> and increases the O<sub>3</sub> concentrations over eastern China in summer and winter.

In order to explore the reason for the increase in O<sub>3</sub> chemical production, we further analyzed the variation in HO<sub>x</sub> (HO + HO<sub>2</sub>) concentration from 2013 to 2017. As the aerosol concentration decreases, its influence on solar radiation is weakened and photolysis is enhanced, leading to an increase in HO<sub>x</sub> levels. It can be seen from Fig. S10 that the concentration of HO<sub>x</sub> increases in both winter and summer.

The increase in HO<sub>x</sub> promotes the conversion of NO to NO<sub>2</sub>, which leads to the accumulation of O<sub>3</sub> concentration.

Figure 7 shows the changed summer and winter surface-layer MDA8 O<sub>3</sub> concentrations caused by anthropogenic emission reduction from 2013 to 2017 with ( $\Delta O_3\_EMI$ ) and without ( $\Delta O_3\_NOARI$ ) ARI, including the effects of weakened ARI on the effectiveness of emission reduction for O<sub>3</sub> air quality ( $\Delta O_3\_ \Delta ARI\_EMI$ , which is also equal to  $\Delta O_3\_EMI$  minus  $\Delta O_3\_NOARI$ ). As shown in Fig. 7a1 and b1, the surface-layer MDA8 O<sub>3</sub> concentrations increased mainly in urban areas during summer and increased uniformly in winter due to anthropogenic emission reduction from 2013 to 2017 without the impact of ARI. When the effect of ARI is considered, the concentrations of MDA8 O<sub>3</sub> increased more than when ARI was not taken into account (Fig. 7a2 and b2). The consequences of weakened ARI as a result of anthropogenic emission reduction on MDA8 O<sub>3</sub> concentrations are shown in Fig. 7a3 and b3. From Fig. 7a3 and b3 we can see that the concentrations of MDA8 O<sub>3</sub> increased in both summer and winter over eastern China.

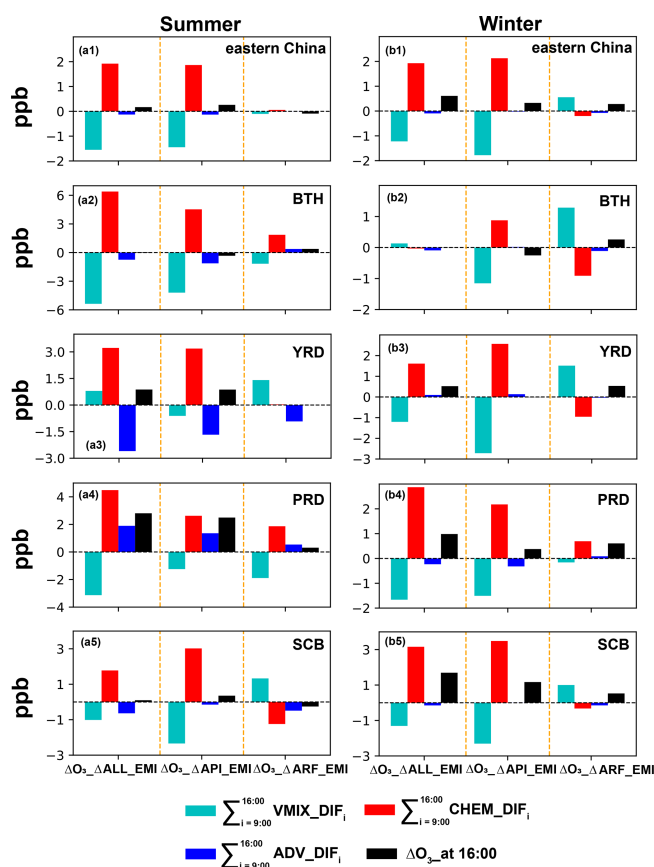


**Figure 5.** Impacts of  $\Delta O_3_{\Delta ARF\_EMI}$ ,  $\Delta O_3_{\Delta API\_EMI}$ , and  $\Delta O_3_{\Delta ARI\_EMI}$  on summer (a1, a2, a3) and winter (b1, b2, b3) surface-layer MDA8  $O_3$  concentrations. The thick black lines in (a1) represent eastern China and the four developed city clusters. The mean changes over eastern China are also shown at the top of each panel. Detailed information about  $\Delta O_3_{\Delta ARF\_EMI}$ ,  $\Delta O_3_{\Delta API\_EMI}$ , and  $\Delta O_3_{\Delta ARI\_EMI}$  can be found in Fig. 1.

Therefore,  $\Delta O_3_{\Delta ARI\_EMI}$  creates the superimposed impact on the effectiveness of anthropogenic emission reduction for the increased MDA8  $O_3$  concentrations from 2013 to 2017 over eastern China. However, during summer, the worsened  $O_3$  air quality due to weakened ARI can only be found in scattered city clusters (e.g., BTH, YRD, and PRD in Fig. 7a3). During winter, it caused increased MDA8  $O_3$  concentrations over nearly the whole of eastern China (Fig. 7b3).

We also average the observed MDA8  $O_3$  concentrations of monitoring sites in the urban areas and the simulation value for the grid cell containing the monitoring site to further examine the impacts of changed meteorological conditions, anthropogenic emissions, and ARI on  $O_3$  levels in densely populated urban areas (Fig. 8). Given that most of the monitoring stations with 5 years of continuous observations are located in urban areas, these monitoring stations and the grid cells containing the monitoring stations can be considered urban areas in this study (Liu and Wang, 2020b). As shown in Fig. 8a1 and b1, the changes in observed MDA8  $O_3$  over urban areas in eastern China from 2013 to 2017 can be captured well by WRF-Chem in both summer and winter. In summer, changed meteorological conditions from 2013 to 2017 had little impact on the variations in MDA8  $O_3$  over the urban areas, while the contribution of emission

reductions to increased MDA8  $O_3$  is significant. In winter, changed meteorological conditions were unfavorable for the increase in MDA8  $O_3$  from 2013 to 2017, indicating that the worsened ozone pollution was driven by the changed anthropogenic emissions. What is more,  $\Delta O_3_{\Delta ARI\_EMI}$  had a significant effect on the increased MDA8  $O_3$  in summer from 2013 to 2017, with a value of +1.77 ppb (87.6%), but its impacts in winter were smaller at only +0.42 ppb (11.8%), which is consistent with the results in Li et al. (2021). The increased MDA8  $O_3$  concentration over urban areas in summer caused by  $O_3_{\Delta ARI\_EMI}$  in this study was 1.77 ppb compared to the 2.12 ppb increase caused by weakened aerosol heterogeneous reactions quantified by Liu and Wang (2020b). Meanwhile, the contributions of  $\Delta O_3_{\Delta API\_EMI}$  and  $\Delta O_3_{\Delta ARF\_EMI}$  to the increase in  $O_3$  concentration averaged over urban areas in eastern China were almost the same in summer (0.79 vs. 0.98) and winter (0.20 vs. 0.22). In general, we find that the enhancement of  $O_3$  concentrations in both summer and winter was mainly caused by reduced anthropogenic emissions. Furthermore, the contributions of  $\Delta O_3_{\Delta API\_EMI}$  and  $\Delta O_3_{\Delta ARF\_EMI}$  to the increases in  $O_3$  concentrations from 2013 to 2017 over urban areas were almost the same during summer and winter.



**Figure 6.** Accumulated changes in each process from 09:00 to 16:00 LST and the changed  $O_3$  concentrations due to  $\Delta O_3_{\Delta ARI\_EMI}$  in summer (a1–a5) and winter (b1–b5). The location, i.e., eastern China, Beijing–Tianjin–Hebei (BTH), the Yangtze River Delta (YRD), the Pearl River Delta (PRD), and the Sichuan Basin (SCB), is indicated on the upper right side of each panel.

### 4.3 Discussion

1. The CBMZ gas-phase chemistry coupled with MOZART aerosol module (CBMZ–MOZAIC for short) used in this study does not include secondary organic aerosol (SOA); thus, we applied three additional chemical mechanisms that consider SOA, namely RADM2 gas-phase chemistry coupled with the MADE/SORGAM aerosol module (RADM2–MADE/SORGAM for short), CBMZ gas-phase chemistry coupled with the MADE/SORGAM aerosol module (CBMZ–MADE/SORGAM for short), and MOZART gas-phase chemistry coupled with the MOZART aerosol module (MOZART–MOZAIC for short), to test the impact of ARI on  $O_3$  with and without SOA for the BASE\_17E17M scenario.

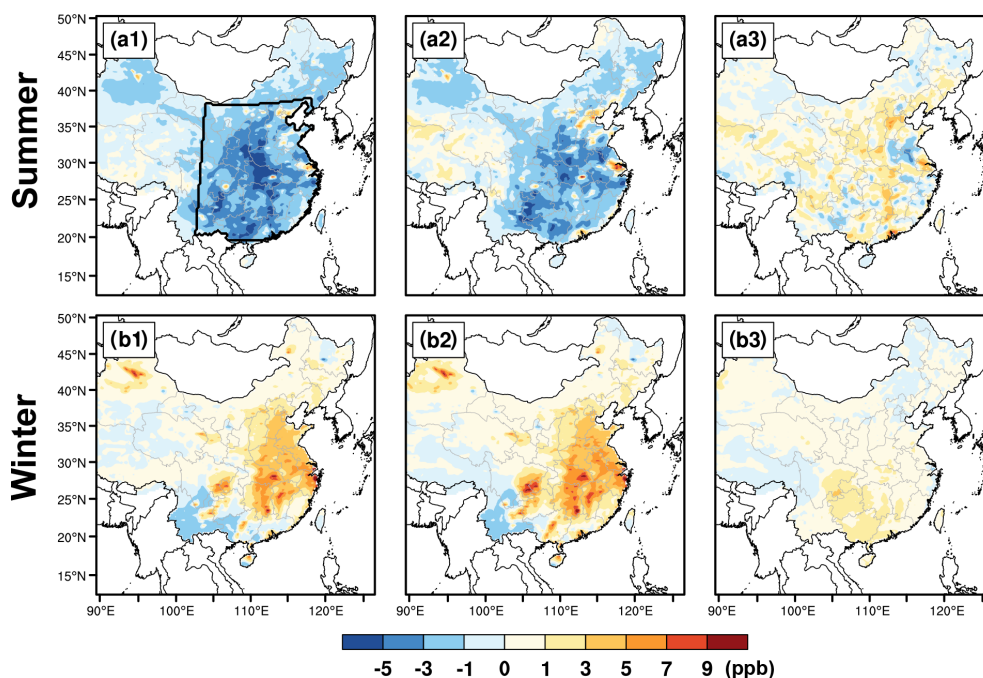
Figure S11 shows the temporal variations in observed and simulated  $PM_{2.5}$  and  $O_3$  concentrations over eastern China for the three additional chemical mecha-

nisms. Compared with the observed  $PM_{2.5}$  ( $O_3$ ) concentrations, MOZART–MOZAIC showed the best performance in December 2017, with  $R$  of 0.73 (0.79) and NMB of  $-18.7\%$  ( $-20.5\%$ ). Therefore, we used this mechanism to further simulate the air pollutant concentrations during the period of June 2017. As shown in Fig. S11 (a4, b4), the temporal variations in observed  $PM_{2.5}$  ( $O_3$ ) can be captured by this mechanism well, with  $R$  of 0.56 (0.91) and NMB of  $-1.7\%$  ( $-20.3\%$ ).

Finally, we investigated the effect of ARI on  $O_3$  from the results of CBMZ–MOZAIC (the mechanism applied in this paper, which does not include SOA) and MOZART–MOZAIC (this mechanism includes SOA and performs the best compared with RADM2–MADE/SORGAM and CBMZ–MADE/SORGAM). As shown in Fig. S12, summer (winter) MDA8  $O_3$  is significantly reduced over eastern China. ARI reduces the surface MDA8  $O_3$  concentrations by 1.32 (1.96) and 1.85 (1.60) ppb in CBMZ–MOZAIC and MOZART–MOZAIC, respectively. The  $O_3$  reductions are of comparable magnitude in these two schemes. Therefore, we can conclude that although the CBMZ–MOZAIC mechanism applied in this paper does not take into account the formation of SOA and its associated effects, the aerosol radiative effects on  $O_3$  concentrations, not only in the pattern of spatiotemporal distribution but also in the order of magnitude, are consistent with the results when the SOA simulation mechanism is considered.

As shown in Fig. S13, the mean SOAs simulated by RADM2–MADE/SORGAM, CBMZ–MADE/SORGAM, and MOZART–MOZAIC are 0.29, 0.45, and  $0.94 \mu g m^{-3}$ , accounting for 3.4 %, 3.8 %, and 4.4 % of  $PM_{2.5}$  concentrations in winter 2017, respectively. From Fig. S14, the mean SOA simulated in MOZART–MOZAIC is  $0.90 \mu g m^{-3}$ , accounting for 9.1 % of  $PM_{2.5}$  in summer 2017. Model-simulated SOA concentrations are generally underestimated by most current chemical transport models (Zhang et al., 2015; Zhao et al., 2015). The low SOA concentrations simulated by the model can be explained by low emission of biogenic and anthropogenic VOCs (key precursors of SOA), but a thorough investigation of this underestimation is outside the scope of this paper and will be discussed in our future work.

2. The impacts of aerosol heterogeneous reactions (HET) on  $O_3$  have not been considered in this paper due to the uncertainty and inconsistency of the heterogeneous uptake shown in previous observation and simulation studies (Liu and Wang, 2020b; Tan et al., 2020; Shao et al., 2021). Liu and Wang (2020b) found that the rapid decrease in  $PM_{2.5}$  was the primary contributor to the summer  $O_3$  increase through weakening of the heterogeneous uptake of the hydroperoxyl radical ( $HO_2$ ). However, Tan et al. (2020) launched a field campaign in



**Figure 7.** Spatial distribution of changed summer (a1, a2, a3) and winter (b1, b2, b3) surface-layer MDA8 O<sub>3</sub> concentrations from sensitivity simulations. (a1, b1) Effects of anthropogenic emission reduction on MDA8 O<sub>3</sub> without ARI. (a2, b2) Effects of anthropogenic emission reduction on MDA8 O<sub>3</sub> with ARI. (a3, b3) Effects of weakened ARI on the effectiveness of emission reduction for O<sub>3</sub> air quality.

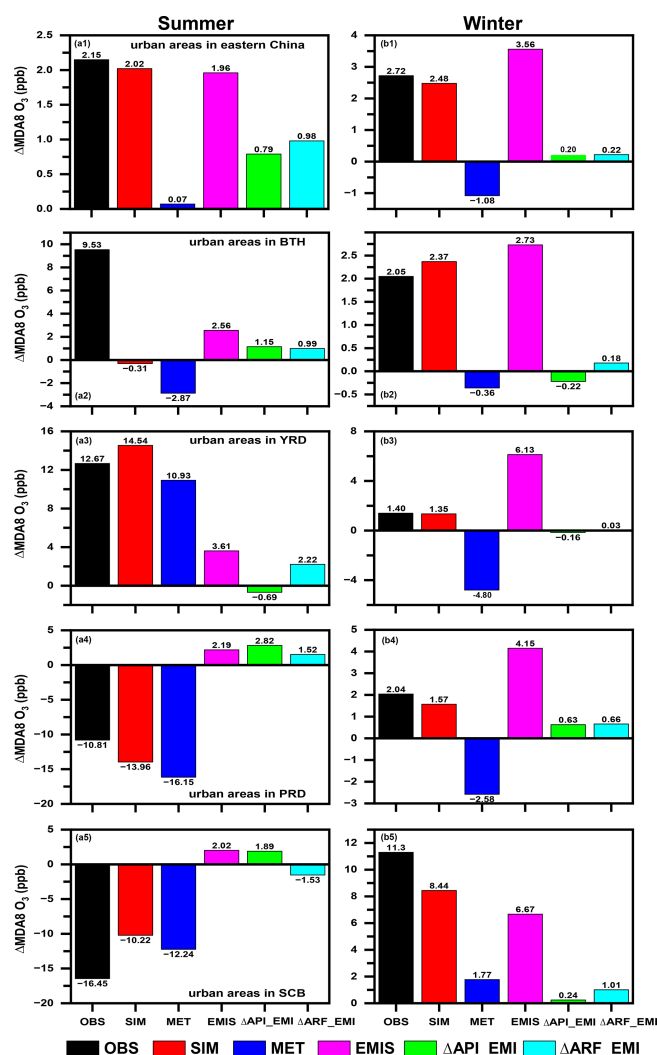
NCP and proposed a contradicting opinion about the importance of the impact of HET on O<sub>3</sub>. Shao et al. (2021) summarized the fact that different heterogeneous uptakes on the aerosol surface applied in the model simulation (e.g., 0.20 vs. 0.08) would cause significant deviations in simulated ozone concentrations (e.g., O<sub>3</sub> increased by 6 %, while O<sub>3</sub> increased by 2.5 %). Previous laboratory studies indicate that the dependence of the uptake coefficient on aerosol composition and RH means that a single assumed value for heterogeneous uptake used in numerical simulations can lead to large uncertainties (Lakey et al., 2015; Taketani et al., 2009; Zou et al., 2019). Therefore, the uncertainty in the heterogeneous uptake value used in the numerical simulation will ultimately amplify the deviation in model results. Meanwhile, our paper is devoted to quantifying the effects of ARI on O<sub>3</sub> rather than the impacts of heterogeneous reactions on O<sub>3</sub>. The absence of heterogeneous chemistry on the aerosol surface may result in underestimation of the effect of aerosol on O<sub>3</sub>, which will be considered in our future work.

- There may be an interaction between API and ARF. However, in this study we discuss the roles of API and ARF separately, which may ignore the effects of interactions between API and ARF on O<sub>3</sub>. This may affect our results, and we will discuss their interaction in our future studies.

## 5 Conclusions

In this study, the impact of weakened aerosol–radiation interaction (ARI) due to decreased anthropogenic emissions on surface O<sub>3</sub> ( $\Delta\text{O}_3_{\Delta\text{ARI\_EMI}}$ ) over eastern China is analyzed, mainly using the online coupled regional chemistry transport model WRF-Chem. Simulation results generally reproduce the spatiotemporal characteristics of observations with correlation coefficients of 0.63–0.90 for pollutant concentrations and 0.40–0.99 for meteorological parameters.

Sensitivity experiments show that the changes in MDA8 O<sub>3</sub> from 2013 to 2017 over eastern China vary spatially and seasonally and that the decreased anthropogenic emissions play a more prominent role in the MDA8 O<sub>3</sub> increase than the impact of changed meteorological conditions in both summer and winter. Furthermore, the decreased PM<sub>2.5</sub> concentrations due to reduced anthropogenic emissions can result in a weaker impact of ARI on O<sub>3</sub> concentrations, which then superimposes its effect on the worsened O<sub>3</sub> air quality. For urban areas over eastern China,  $\Delta\text{O}_3_{\Delta\text{ARI\_EMI}}$  has a significant effect on the increase in MDA8 O<sub>3</sub> in summer with a value of +1.77 ppb, accounting for 87.6 % of the increased value caused by decreased anthropogenic emissions, but the impacts in winter are smaller (+0.42 ppb), accounting for 11.8 % of the increased value caused by decreased anthropogenic emissions. For the whole region over eastern China, the enhancement of MDA8 O<sub>3</sub> by  $\Delta\text{O}_3_{\Delta\text{ARI\_EMI}}$  is +0.81 (+0.63) ppb,



**Figure 8.** The observed (OBS; black bars) and simulated (SIM; red bars) changes in (left) summer and (right) winter surface-layer MDA8 O<sub>3</sub> from 2013 to 2017. Contributions of changed meteorological conditions alone (MET; blue bars), changed anthropogenic emissions alone (EMI; purple bars), changed aerosol–photolysis interaction alone ( $\Delta\text{API\_EMI}$ ; green bars), and changed aerosol–radiation feedback alone ( $\Delta\text{ARF\_EMI}$ ; cyan bars) are also shown. Observations are calculated from the monitoring sites in the analyzed region, while the corresponding gridded simulations are averaged for SIM. Panels represent the urban areas in eastern China, Beijing–Tianjin–Hebei (BTH), the Yangtze River Delta (YRD), the Pearl River Delta (PRD), and the Sichuan Basin (SCB).

with  $\Delta\text{O}_3_{\Delta\text{API\_EMI}}$  and  $\Delta\text{O}_3_{\Delta\text{ARF\_EMI}}$  contributing 55.6 % (61.9 %) and 44.4 % (38.1 %) in summer (winter), respectively. Process analysis shows that the enhanced O<sub>3</sub> chemical production is the dominant process for the increased O<sub>3</sub> concentrations caused by  $\Delta\text{O}_3_{\Delta\text{ARI\_EMI}}$  in both summer and winter.

Generally, since China’s clean air action in 2013, the decreased PM<sub>2.5</sub> concentrations due to reduced anthropogenic

emissions have worsened O<sub>3</sub> air quality through the weakened interactions between aerosol and radiation, which is a new and an important implication for understanding the causes driving the increases in the O<sub>3</sub> level over eastern China. Therefore, our results highlight that more carefully designed multi-pollutant coordinated emission control strategies are needed to reduce the concentrations of PM<sub>2.5</sub> and O<sub>3</sub> simultaneously.

**Data availability.** The observed hourly surface concentrations of air pollutants are derived from the China National Environmental Monitoring Center (<https://air.cnemc.cn:18007/>, CNEMC, 2024). The observed surface meteorological data are obtained from <https://www.ncei.noaa.gov/maps/hourly/> (National Centers for Environmental Information, 2024). The photolysis rates of nitrogen dioxide in Beijing are provided by Xin Li ([li\\_xin@pku.edu.cn](mailto:li_xin@pku.edu.cn)). The simulation results can be accessed by contacting Lei Chen ([chen-lei@nuist.edu.cn](mailto:chen-lei@nuist.edu.cn)) or Hong Liao ([hongliao@nuist.edu.cn](mailto:hongliao@nuist.edu.cn)).

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