



# Impact of Short-Term Emission Control Measures on Air Quality in Nanjing During the Jiangsu Development Summit

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This study analyzed the effectiveness of temporary emission control measures on air quality of Nanjing, China during the Jiangsu Development Summit (JDS). We employed a regional chemistry model WRF-Chem to simulate air pollutants in Nanjing and compared the results to surface observations and satellite retrievals. During the JDS, air pollutant emissions from industry and transportation sectors largely decreased by 50-67% due to the short-term emission control measures such as reducing coal combustions, shutting down factories, and partially limiting traffic. Benefiting from the emission control, the simulated concentrations of PM<sub>2.5</sub>, NO<sub>2</sub>, SO<sub>2</sub>, CO and VOCs in Nanjing decreased by 17%, 20%, 20%, 19%, and 15% respectively, consistent with the surface and satellite observations. However, both the observed and simulated  $O_3$  increased by 3–48% during the JDS, which was mainly due to the remarkable NO<sub>x</sub> emission reduction (26%) in the downtown of Nanjing where the O3 production regime was mainly VOC-controlled. In addition, the atmospheric oxidation capacity and further the sulfur oxidation ratio, were facilitated by the elevated O3, which led to variable mitigation efficiencies of different secondary PM<sub>2.5</sub> compositions. Our study offers an opportunity for understanding the coordinated control of PM<sub>2.5</sub> and O<sub>3</sub> in typical city clusters, and can provide implications for future mitigation actions.

# Keywords: PM<sub>2.5</sub>, O<sub>3</sub>, emission control, WRF-Chem, satellite remote sensing

# INTRODUCTION

In past several years, China has been threatened by severe air pollution mainly resulted from fine particulate matter ( $PM_{2.5}$ , aerodynamic diameter is less than 2.5 µm) and ozone ( $O_3$ ). High  $PM_{2.5}$  concentration can induce haze events Huang et al. (2014), Wang et al. (2016), Li et al.( 2019), and further reduce visibility Zhang et al. (2015), Chen et al. (2016), adversely affect human health Hu et al. (2017), Zhang et al. (2017) and climate change (Bond et al., 2013; IPCC, 2013). Compared to  $PM_{2.5}$ ,  $O_3$  is a kind of gaseous pollutant with great oxidation. The oxidation of  $O_3$  may make damage to crops or plants as well as increases the risk of pulmonary function damage (Tao et al., 2012).

In order to alleviate the  $PM_{2.5}$  and  $O_3$  pollution, the Chinese government enacted a stricter air quality standard in 2013, i.e. National Ambient Air Quality Standard (NAAQS, GB3095-2012). At the same time, the Chinese State Council also promulgated a series of long-term projects to improve

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Zhang H, Tang K, Feng W, Yan X, Liao H and Li N (2021) Impact of Short-Term Emission Control Measures on Air Quality in Nanjing During the Jiangsu Development Summit. Front. Environ. Sci. 9:693513. doi: 10.3389/fenvs.2021.693513 air quality such as Air Pollution Prevention and Control Action Plan and Three-year Action Plan on Defending the Blue Sky (Zhai et al., 2019). These specific projects include desulfurization and denitration of flue gas, elimination of backward industry and ban on high-emission motor vehicles (Li et al., 2017; Tang et al., 2019; Zhang et al., 2019).

At the same time, short-term emergency emission controls were also applied during some important conferences or activities for ensuring air quality. For example the Asia-Pacific Economic Cooperation (APEC) summit in 2016, the summit for the Group of Twenty (G20) in 2016 and the 19th National Congress of the Communist Party of China (NCCPC) in 2017. These case studies suggested that temporary control measures were effective in improving air quality (Xu et al., 2017; Ansari et al., 2019; Wang et al., 2019; Ma et al., 2020; Zhang et al., 2020). However, the internal mechanisms of pollution reduction may still be complicated. For example, based on in-situ field measurements, Chang et al. (2020) pointed out that secondary aerosols significantly increased during the coronavirus disease (COVID-19) outbreak. Although the nationwide lockdown reduced primary emissions unprecedentedly. Haze episodes were even observed in Beijing (Huang et al., 2020).

In this study, we examined the effectiveness of a short-term emission control case during the Jiangsu Development Summit (JDS). JDS was held from 20 to 22, May 2017 in Nanjing, the capital of Jiangsu province, China. Source apportionment of PM2.5 is crucial to air pollution control. Numerous studies about source apportionment were conducted in east Aisa (Ge et al., 2017; Jain et al., 2017; Ye et al., 2017; Jain et al., 2020; Jain et al., 2021). Previous literatures identified that vehicles and industrial processes contributed considerable PM2.5 in urban regions of China and India. In this study, local emission profile revealed that vehicles and industrial processes contributed 84% of primary PM2.5 in Nanjing. Temporary emission control measures were implemented in Nanjing by reducing coal combustions, shutting down high-emission factories, prohibiting construction and partially limiting traffic, etc. In addition, cooperative emission reductions were also carried out by the neighboring cities. Overall, the aim of this study is: 1) to investigate the effects of emission control measures on air quality during the JDS; 2) to explore mechanisms that drive changes due to emission control measures. This study provided a detailed analysis of the effectiveness and mechanisms based on a classic short-term emission control case. The results provide insights into further mitigation of air pollution in other urban areas.

# MATERIALS AND METHODS

The study period was from 17 to May 22, 2017. Based on the bottomup emission inventory, we categorized air pollutants into five sectors, i.e. industry, power, residential, transportation and agriculture. In terms of air pollutants, nine species were included, such as SO<sub>2</sub>, NO<sub>x</sub> (x = 1, 2), CO, ammonia (NH<sub>3</sub>), non-methane volatile organic compounds (NMVOC<sub>s</sub>), elemental carbon (EC), organic carbon (OC), PM<sub>2.5</sub>. The local government mainly controlled emissions by limiting transportation and industrial activities. Industrial activities contributed 63% of SO<sub>2</sub> emission as well as 70% of VOCs emission. During JDS, a total of 116 key petrochemical, chemical, steel and other enterprises in Nanjing were reduced emissions by more than 50%. Meanwhile, the implementation of production restrictions or phased shutdown program was also conducted (Yan, 2019). The specific proportions of emission reductions are listed in **Table 1**. Emissions from industry and transportation sectors were decreased by 50–67% and 11–79% for various air pollutants. The reduction in primary emission was estimated based on the statistics by calculating the limited emission activities, which was obtained from the local government (Yan, 2019).

To evaluate the effectiveness of short-term emission control measures, we obtained surface measurements (*Surface Measurements*) and satellite remote sensing products (*Satellite retrievals*) during the JDS and the spring (March, April and May 2017). The satellite retrievals contained the vertical column density (VCD) for gaseous species and the aerosol optical depth (AOD) for PM<sub>2.5</sub>. In this study, we referred spring to the normal period. The effects of emission control measures could be accessed by comparing the observations during the JDS and the normal period. In addition, we utilized Weather Research and Forecast coupled with Chemistry (WRF-Chem, *WRF-Chem Model*) model to quantitatively investigate the impacts and the internal mechanisms of short-term emission control measures.

# **Surface Measurements**

The Chinese government has established a high spatio-temporal resolution air quality monitoring network since 2013. Data are available through the public website of the Ministry of Ecology and Environment of China (http://106.37.208.233:20035/). We collected hourly surface measurements of five criteria pollutants during the study period, including PM<sub>2.5</sub>, O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub>, and CO. In this study, we analyzed the records of nine monitoring sites in Nanjing. All monitoring sites in Nanjing are located in urban areas with the exception of the rural site, Pukou (PK). In addition to the surface observations of six criteria pollutants, we also obtained surface meteorological observations (http://www.meteomanz.com/) for temperature, relative humidity and wind vectors in Nanjing. More descriptions of these sites are provided in **Supplementary Table S1**. Geographical locations are also available in **Supplementary Figure S1**.

# Satellite Retrievals

The satellite products used in this study include the VCD of gaseous species, i.e. SO<sub>2</sub>, NO<sub>2</sub> and CO, as well as column AOD. The VCD profiles of CO were obtained from Infrared Atmospheric Sounding Interferometer (IASI, https://iasi.aerisdata.fr) based on the Metop-B satellite. AOD, SO<sub>2</sub> and NO<sub>2</sub> VCD were derived from The Ozone Monitoring Instrument (OMI) based on the Aura satellite Dobber et al. (2006), provided by NASA Goddard Earth Sciences Data and Information Services Center (GESDISC, https://urs.earthdata.nasa.gov). Detailed descriptions for satellite products are provided in Supplementary Table S2. These satellite retrievals have been optimized via the improved algorithms. For SO<sub>2</sub>, OMSO2003 was inversed on the basis of a principal component analysis (PCA) algorithm with more flexible Jacobians rather than a fixed factor or profile (Li et al., 2013; Li et al., 2017; Li et al., 2020). For NO<sub>2</sub>,

	Industry (%)	Power (%)	<b>Residential (%)</b>	Transportation (%)	Agriculture (%)	Total (%)
Species						
SO <sub>2</sub>	53	0	0	52	0	33
NO <sub>x</sub>	60	0	0	18	0	26
CO	51	0	0	11	0	36
$NH_3$	50	0	0	0	0	0
NMVOC <sub>s</sub>	51	0	0	23	0	35
EC	52	0	0	79	0	52
OC	55	0	0	71	0	40
PM <sub>2.5</sub>	63	0	0	76	0	57

TABLE 1 | Reductions of the air pollutant emissions in Nanjing during the First Jiangsu Development Summit.

this study selected the latest version of  $NO_2$  retrievals with the algorithm based on geometry-dependent surface Lambertian equivalent reflectivity (Lamsal et al., 2020). For AOD, the quality of the data was improved by (van den Oord et al., 2006). For CO, it was processed through the methods of Hurtmans et al. (Hurtmans et al., 2012).

### WRF-Chem Model

WRF-Chem (v3.9.1) is an open-source regional chemical transport model (CTM) developed by the US National Center for Atmospheric Research (NCAR). WRF-Chem is capable of simulating air quality with fully coupled meteorology and chemistry (Grell et al., 2005). Initial and boundary meteorological conditions were derived from FNL reanalysis data (https://rda.ucar.edu/datasets/ds083.2/) by the National Centers for Environmental Prediction (NCEP). The anthropogenic emissions were based on the Multi-resolution Emission Inventory for China (MEIC, http://meicmodel.org/), which was developed by Tsinghua University (Li et al., 2017). Online biogenic and real-time biomass burning emissions were calculated from Model of Emissions of Gases and Aerosols from Nature (MEGAN, v2.1) tool (Guenther et al., 2006) and Fire INventory from NCAR (FINN, https://www2.acom.ucar.edu/ modeling/finn-fire-inventory-ncar) respectively. WRF-Chem incorporates a variety of chemical parameterizations. We chose Statewide Air Pollution Research Center (SAPRC99) mechanism as the gas phase reaction scheme (Carter, 2000). SAPRC99 comprised 74 gases and 211 gaseous reactions. VOCs are lumped to describe O3 and SOA production processes. According to the previous studies, SAPRC showed superior performance in reproducing O<sub>3</sub> and SOA concentrations (Li et al., 2011; Feng et al., 2016; Hu et al., 2016). Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) was selected for the modeling of aerosols (Zaveri et al., 2008). MOSAIC places aerosol species, including sulfate (SO<sub>4</sub><sup>2-</sup>), nitrate  $(NO_3^{-})$ , ammonium  $(NH_4^{+})$ , EC, OC, sea salt  $(Na^{+} and Cl^{-})$  and other inorganic aerosol (OIN) into eight size bins (0.039-0.078, 0.078-0.156, 0.156-0.313, 0.313-0.625, 0.625-1.250, 1.250-2. 500, 2.500-5.000 and 5.000-10.000 µm for dry diameter). We combined the mass concentrations of sea salt with OIN as the mass concentrations of dust. All aerosol species are assumed to be internally mixed within each size bin. Finally, the initial and boundary chemical conditions were determined by MOZART global model (Emmons et al., 2010).

The start date of the simulation was set on 15 May. The first 48 h was spin-up period. The domain of WRF-Chem model covers the Yangtze River Delta region (YRD, see **Supplementary Figure S1A**), with a horizontal resolution of  $4 \times 4$  km. Vertically, 28 layers extended from the surface to a height of 50 hPa, with seven layers below the bottom 1 km (refer to the scale of planetary boundary layer). Details of the configuration of model are listed in **Supplementary Table S3**.

### RESULTS

### Model Evaluation

We evaluated the model performance by comparing with surface meteorology and air pollutant observations, as well as satellite products. Comprehensive comparisons between observations and simulations are shown in **Table 2**. We adopted three statistical indexes, i.e. the index of agreement (IOA, varies from 0 to 1), the normalized mean bias (NMB, varies from -1 to  $+\infty$ ) and the root mean square error (RMSE, varies from 0 to  $+\infty$ ), to describe the performance of the model in simulating surface species. The equations for the calculations of these statistics are shown in **Eqs 1–3**, where *s*, *o*, *C* and *N* indicates the simulation, observation, concentration and total number of samples, respectively.

$$IOA = 1 - \frac{\sum_{i=1}^{N} (C_S - C_o)^2}{\sum_{i=1}^{N} \left( \left| C_S - \overline{C_o} \right| + \left| C_o - \overline{C_o} \right| \right)^2}$$
(1)

$$NMB = \frac{\sum_{i=1}^{N} (C_{S} - C_{o})}{\sum_{i=1}^{N} C_{o}}$$
(2)

$$RMSE = \sqrt{\frac{\sum\limits_{i=1}^{N} (C_s - C_o)^2}{N}}$$
(3)

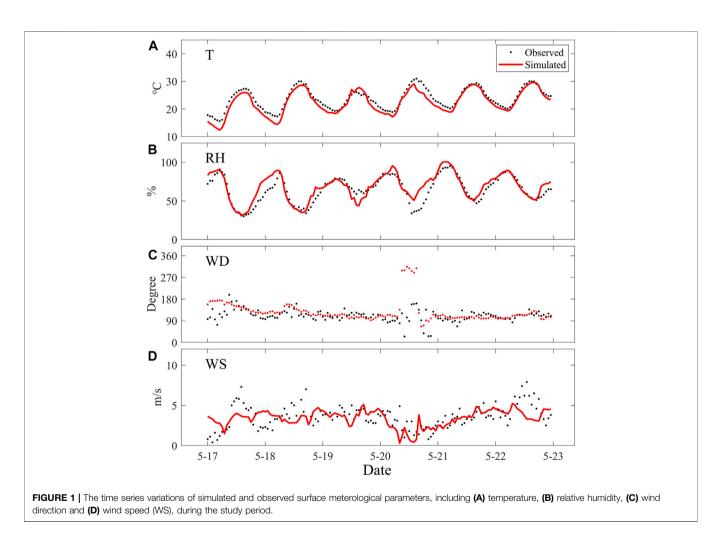
### Meteorology

Firstly, we examined the agreement between observation and simulation in terms of meteorological parameters. The time series diagrams for temperature, relative humidity, wind direction and

#### TABLE 2 | The comparison between simulations against surface observations or satellite retrievals.

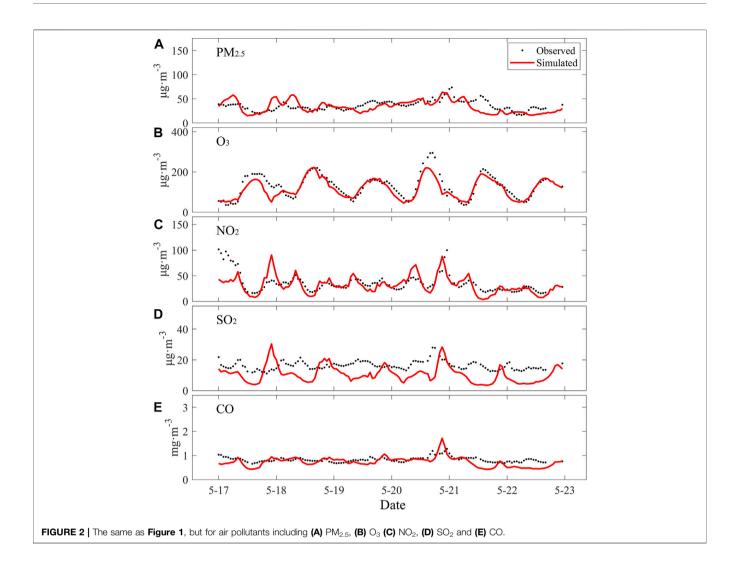
	Mean		IOA	NMB	RMSE
	Observation	Simulation			
Model vs Surface observations					
Temperature (°C)	23.8	22.4	0.90	-6%	1.7
Relative humidity (%)	64.6	68.1	0.85	5%	9.0
Wind speed (m/s)	3.6	3.4	0.73	-5%	1.5
PM <sub>2.5</sub> (µg⋅m <sup>-3</sup> )	35.8	33.8	0.64	-6%	12.3
O <sub>3</sub> (µg⋅m <sup>-3</sup> )	126.3	113.9	0.87	-10%	28.8
NO₂ (µg⋅m <sup>-3</sup> )	35.7	32.4	0.75	-9%	16.5
SO₂ (µg⋅m <sup>-3</sup> )	16.2	10.5	0.41	-35%	8.2
CO (mg⋅m <sup>-3</sup> )	0.8	0.7	0.53	-11%	0.2
Model vs Satellite retrievals <sup>a</sup>					
AOD	0.8	0.9	_	_	_
NO <sub>2</sub> VCD	12.1	15.4	_	_	_
SO <sub>2</sub> VCD	13.7	12.0	_	_	_
CO VCD	3.1	3.7	_	_	_

<sup>a</sup>AOD is unitless. For SO<sub>2</sub> and NO<sub>2</sub> VCD, unit is 10<sup>15</sup> mol cm<sup>-2</sup>. For CO VCD, unit is 10<sup>18</sup> molecules cm<sup>2</sup>.



wind speed are shown in **Figure 1**. The average of the observed temperature, relative humidity and wind speed during the study period was 23.8°C, 64.6% and 3.6 m/s, respectively. The leading wind came from the southeast. WRF-Chem successfully captured

both the magnitude and the varying trends for these meteorological parameters in general. Among these three meteorological parameters, the temperature had a maximum IOA, with the value is 0.9. Meanwhile, the NMB for all



meteorological parameters were less than  $\pm 10\%$ , emphasizing that WRF-Chem is capable to reproduce weather conditions. Precise simulated meteorology is crucial to reflect the real vertical diffusion and horizontal advection.

### Chemistry

**Figure 2** illustrates the comparison between the simulated chemical species and the observed ones, including  $PM_{2.5}$ ,  $O_3$ ,  $NO_2$ ,  $SO_2$  and CO. The mean observation for these species was  $35.8 \,\mu g \, m^{-3}$ ,  $126.3 \,\mu g \, m^{-3}$ ,  $35.7 \,\mu g \, m^{-3}$ ,  $16.2 \,\mu g \, m^{-3}$  and  $0.8 \,m g \, m^{-3}$  respectively. As **Table 2** shown, all the chemical species were well predicted except for SO<sub>2</sub>, with a range of IOA from 0.53 (CO) to 0.87 (O<sub>3</sub>). The corresponding NMB varies from -11% (CO) to -6% (PM<sub>2.5</sub>). In addition, the diurnal patterns of two typical photochemical products (i.e. O<sub>3</sub> and NO<sub>2</sub>) were also successfully simulated by model (**Supplementary Figure S2**).

### Satellite Products

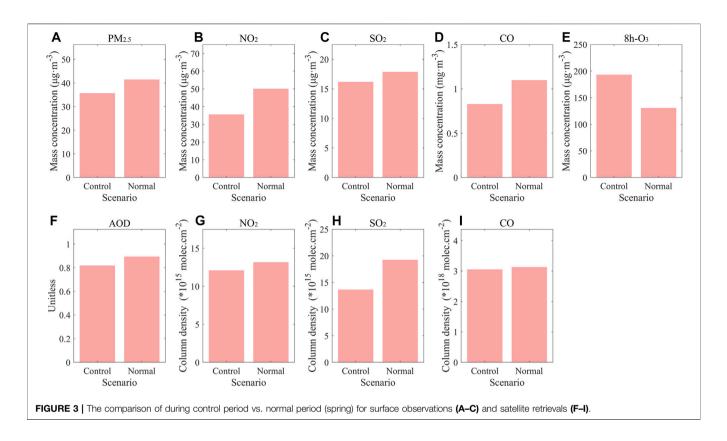
We also compared VCDs and AOD between the satellite retrievals and the model simulations. In WRF-Chem, the AOD

properties were calculated at wavelengths of 300, 400, 600, and 999 nm. However, we adopted the OMI AOD products at a wavelength of 500 nm. Thus the simulated AOD at 500 nm should be derived from the simulated AOD at 300, 600, 999 nm based on the Ångström power law, see **Eq. (4)**. (Kumar et al., 2014). In **Eq. (4)**, A,  $\lambda$  and  $\alpha$  denotes AOD, wavelength and Ångström exponent respectively. The calculation for  $\alpha$  followed **Eq. (5)**.

$$A(\lambda) = A(600) \times \left(\frac{\lambda}{600}\right)^{-\alpha}$$
(4)

$$\alpha = \frac{\ln\left(\frac{A(300)}{A(999)}\right)}{\ln\left(\frac{999}{300}\right)} \tag{5}$$

The comparisons between simulated and observed VCDs are provided in **Table 2**. The mean inversed VCD of NO<sub>2</sub>, SO<sub>2</sub> and CO based on satellite remote sensing was  $12.1 \times 10^{15}$  molecules·cm<sup>-2</sup>,  $13.7 \times 10^{15}$  molecules·cm<sup>-2</sup> and  $3.1 \times 10^{18}$  molecules·cm<sup>-2</sup>, respectively. For AOD, the mean value was 0.8. Model simulations were in good agreement with satellite



retrievals in terms of magnitude. It was highlighted that WRF-Chem was capable to simulate reliable vertical diffusion as well.

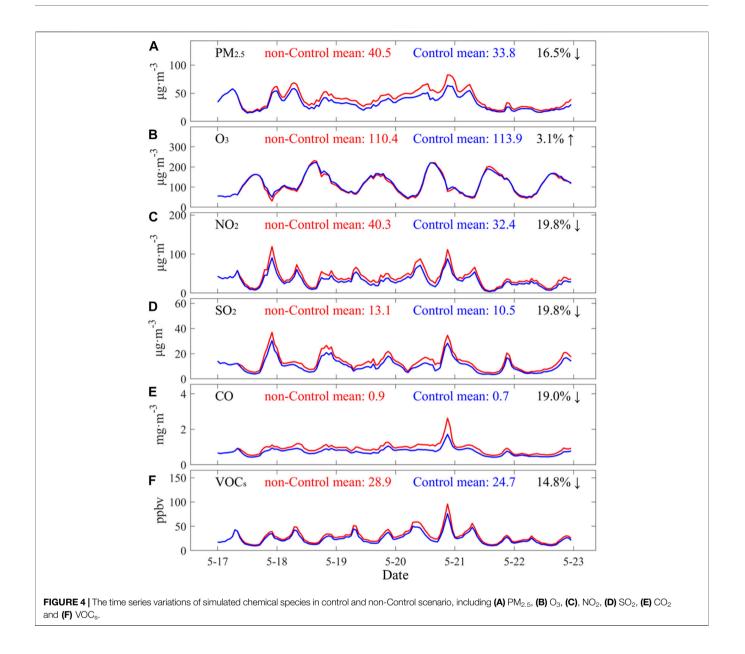
# Air Quality Improvement During the JDS

The corresponding diffusion condition of JDS was close to that in Spring because there was no obvious special weather process such as gale or precipitation during JDS. The seasonal mean concentrations could indicate the representative level of air pollutants. To evaluate the effectiveness short-term control measures, our study therefore compared the seasonal mean concentration and the average during JDS in terms of both surface concentrations and satellite retrievals. Figure 3 compares the air quality in Nanjing during the JDS control period and the spring of the same year. For surface observations, PM<sub>2.5</sub>, NO<sub>2</sub>, SO<sub>2</sub> and CO posed a decreasing trend during the control period (Figure 3A-D). PM<sub>2.5</sub>, NO<sub>2</sub>,  $SO_2$  and CO decreased from 41.5 to 35.8 µg m<sup>-3</sup> (14%), from 50.2 to 35.7  $\mu g$  m  $^{-3}$  (29%), from 17.9 to 16.2  $\mu g$  m  $^{-3}$  (9%) and from 1.1 to  $0.8 \text{ mg m}^{-3}$  (24%) respectively. The unifying declines indicated that short-term emission control measures were effective, particularly for NO<sub>2</sub>. However, O<sub>3</sub> showed an increase. The mean 8h-O<sub>3</sub> concentration during the JDS was 193.6  $\mu$ g m<sup>-3</sup>, which was 48% higher than the seasonal mean value in spring (Figure 3E). We also assessed the air quality improvements from the perspective of satellite. As Figure 3F-I shown, AOD and gaseous VCD presented a decline of 3-29% which also can be attributed to the massive reduction of ground emissions.

# Impacts of the Short-Term Measures During the JDS

### Five Criteria Air Pollutants and VOCs

We conducted two sensitivity simulations to assess the effectiveness of short-term emission control measures on the air quality during the JDS, emission control scenario (considering the emission control as shown in Table 1) and non-control scenario (none emission control). As shown in Figure 4, the concentrations of SO<sub>2</sub>, NO<sub>2</sub>, CO and VOCs decreased by 15-20% as the result of the emission control measures. The decrease in those four species could be explained by the reduction of emissions in transportation (11-52%) and industrial activities (51-60%). Especially for SO<sub>2</sub>, the strict control measures in industry, which was dominant source, induced 20% decline in its concentration. NO<sub>2</sub> shows the highest mitigation efficiency. The 26% decline in NO<sub>x</sub> emissions resulted in a 20% decrease in NO2 concentrations. VOCs was the most difficult species to control, for which a 35% emission reduction only led to a 15% decrease in concentration. Similarly, PM<sub>2.5</sub> concentration was reduced by 17% due to the emission control of its precursors, however, the magnitude of the PM2.5 decrease was relatively smaller compared to its precursors. The day-to-day variation of meteorology conduced the variations in air pollutant concentrations. We summarized the daily average of meteorological parameters in Supplementary Table S4. The variations of meteorological parameters, especially for the wind speed, determined the advection condition and further contributed to the difference in the pollutants from day to

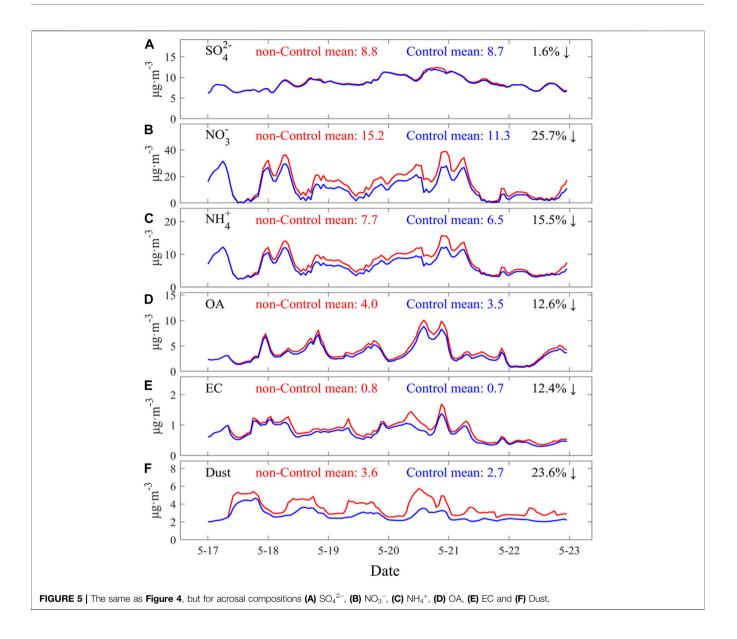


day. Unlike these air pollutants, the concentration of  $O_3$  slightly increased by ~3% due to the impacts of emission reduction measures. The increase in  $O_3$  can be attributed by its nonlinear formation regime affected by  $NO_x$  and VOCs (Wang et al., 2017). This evidence suggested that it was difficult to mitigate  $O_3$  pollution via current emission control measures. More effective controls should be considered for preventing photochemical pollution. The detailed differences in concentration between two scenarios and their relative changes are summarized in **Supplementary Table S51**.

### Aerosol Compositions

We further analyzed the changes of each aerosol composition in the sensitivity simulations. As shown in **Figure 5**, we classified six types of aerosol species, including  $SO_4^{2-}$ ,  $NO_3^{-}$ ,  $NH_4^+$ , organic aerosol (OA), EC and dust. Here the OA was inferred based on OC multiplied by an empirical ratio of 1.4 (Seinfeld and Pandis, 2006). In the case of implementing emission control measures, the secondary inorganic aerosol containing sulfate, nitrate and ammonium (SNA) contributed almost 80% of the total concentration of  $PM_{2.5}$ .  $NO_3^-$  was the dominant chemical component of the SNA. The mean simulated nitrate concentration was 11.3 µg m<sup>-3</sup>, approximately occupied 43% of the SNA. On the other hand, EC accounted for the least fraction of  $PM_{2.5}$  (less than 3%).

All these aerosol compositions showed a decline due to the control measures. The unifying decrease of  $PM_{2.5}$  concentration occurred serval hours later after the emission reduction pattern, indicating that the short-term control may not have an immediate effect on  $PM_{2.5}$ . However the reduction in primary components such as EC and dust come earlier. The secondary components tend to have a lag of several hours in the decline of



concentration due to the chemical processes. Dust and  $NO_3^-$  decreased most significantly among all  $PM_{2.5}$  compositions. Dust decreased by 24% owing to the short-term emission control such as banning construction.  $NO_3^-$  decreased by about 26%, which was attributed to the tremendous reduction of the precursor  $NO_x$ . Compared to  $NO_3^-$ , the other two SNA,  $SO_4^{-2-}$  and  $NH_4^+$ , only decreased by 2 and 16%. Particularly for  $SO_4^{-2-}$ , the absolute decline in mass concentration under control was less than 0.1 µg m<sup>-3</sup>.

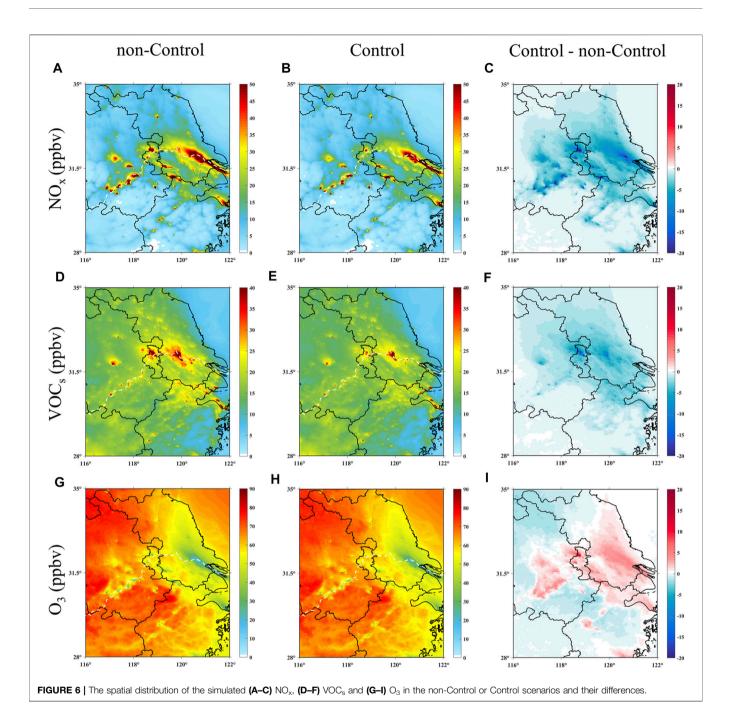
# DISCUSSION

# Mechanisms of O<sub>3</sub> Pollution Episode

 $O_3$  is the main pollutant during the study period. The maximum hourly concentration of the observed  $O_3$  was 294 µg m<sup>-3</sup> (20 May), which far exceeded 200 µg m<sup>-3</sup> (NAAQS level II). The peak

of  $O_3$  concentration tended to occur between 14:00 and 18:00 (local time, **Supplementary Figure S2**). This type of serious  $O_3$ pollution episode could be explained from the following two factors. One was the influences of weather conditions. During the periods of, the weather conditions were characterized as higher temperature and lower relative humidity. The mean temperature and relative humidity observed during  $O_3$  pollution episodes was 28°C and 46%, which is extremely favorable for the production of  $O_3$  (Liu and Wang, 2020).

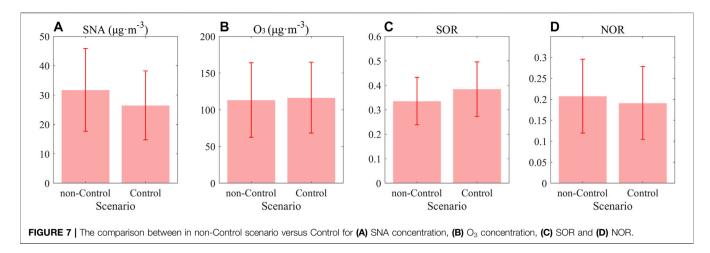
The other cause related to the non-linear production regime of  $O_3$ . **Figure 6** illustrates the spatial changes of  $O_3$  (g-i) and its precursors  $NO_x$  and VOCs (a-f). As shown in **Figure 6A–B**, the spatial high concentrations of  $NO_x$  occurred along with the Yangtze River. This could reflect the spatial distribution pattern of transportation emissions, such as shipping and motor vehicles. For VOCs, the regional high concentrations tended to exist in the central of Jiangsu province. The



emission control measures taken in Nanjing and its neighboring cities induced both NO<sub>x</sub> and VOCs spatially decease by 1.2 ppbv and 0.9 ppbv respectively. Meanwhile, the decrease was more significant in Nanjing. The decrease in NO<sub>x</sub> was more dramatic in terms of specific concentrations (**Figure 6C–F**). The control strategy about NO<sub>x</sub> and VOCs contributed to an increase in O<sub>3</sub> concentration, particularly in the coastal regions and the southern areas of Jiangsu Province.

We applied an indicator method to further infer the  $O_3$  production regime. Ozone production efficiency (OPE) is one of the most useful indicators. OPE is defined as the ratio of  $\Delta O_3$  to

 $\Delta NO_z$ .  $NO_z$  consists of nitrogenous compounds excluding  $NO_x$ , such as HONO, HNO<sub>3</sub>, HO<sub>2</sub>NO<sub>2</sub>, N<sub>2</sub>O<sub>5</sub>, NO<sub>3</sub>, peroxy acetyl nitrate (PAN) and other organic nitrates. OPE is derived from the slope of the regression between O<sub>3</sub> and NO<sub>z</sub>. It denotes the mean number of O<sub>3</sub> molecules produced when a NO<sub>2</sub> molecule is oxidized to be a NO<sub>z</sub> species. NO<sub>z</sub> therefore has a good linear correlation with O<sub>3</sub>. The O<sub>3</sub> production regime is under VOC<sub>s</sub>-controlled in the case of OPE is less than 4. On the contrary, NO<sub>x</sub> is the predominant species of O<sub>3</sub> formation if OPE is more than 7. The other OPE values between 4 and seven are correspond to the mixed-controlled status (Wang et al., 2017).



**Supplementary Figure S3** shows the scatter plots of  $O_3$  versus  $NO_z$  based on the simulated results in Nanjing urban sites during the peak time of  $O_3$  formation. The OPE values in non-control and control scenarios were 3.76 and 3.42, respectively, highlighting that the  $O_3$  formation regime in urban areas of Nanjing was under VOCs-controlled, consistent with previous studies (Jin and Holloway, 2015; Wang et al., 2017; Li et al., 2018).

### Aerosol Chemistry in PM<sub>2.5</sub> Reduction

Previous simulations have confirmed that the emission control measures during the JDS reduced SO<sub>2</sub> concentration by 20% but reduced SO<sub>4</sub><sup>2-</sup> by only 2%. This phenomenon could be one of the side effects of the elevated O<sub>3</sub> (**Figure 7B**). First, sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) has stronger capacity to neutralize NH<sub>3</sub> compared with HNO<sub>3</sub>, which ensures that the production of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> or NH<sub>4</sub>HSO<sub>4</sub> is prior to that of NH<sub>4</sub>NO<sub>3</sub> (Seinfeld and Pandis, 2006). In addition, the universal increase in O<sub>3</sub> across the YRD region induced stronger atmospheric oxidation capacity (AOC), and further facilitated SO<sub>2</sub> to produce H<sub>2</sub>SO<sub>4</sub> more efficiently. Subsequently, the efficiency of SO<sub>4</sub><sup>2-</sup> production, we adopted sulfur oxidation ratio (SOR) as **Eq. (6**).

$$SOR = \frac{n[SO_4^{2-}]}{n[SO_4^{2-}] + n[SO_2]}$$
(6)

$$NOR = \frac{n[NO_{3}^{-}]}{n[NO_{3}^{-}] + n[NO_{2}]}$$
(7)

$$PNR = \frac{n[NH_4^+]}{2n[SO_4^{2-}] + n[NO_3^-]}$$
(8)

The *n* in **Eq. (6)** stands for mole concentration (unit: mole). As shown in **Figure 7C**, the SOR in the control scenario was 0.38, 15% higher than that in the non-control scenario. The increased  $SO_4^{2^2}$  yields almost offset the impacts of SO<sub>2</sub> concentration reduction. The similar results were also found in previous observational or modeling studies focusing on the COVID-19 (Chang et al., 2020; Huang et al., 2020; Le et al., 2020; Liu et al., 2021).

However, the efficiency of  $NO_3^-$  production could be relatively reduced. As shown in **Figure 7D**, the nitrogen oxidation ratio (NOR, which as defined in **Eq. (7)**) decreased by 8% owing to the

emission control measures. The NOR was not promoted even though the ambient atmosphere was more oxidizing. This could be related to the limited concentration of NH<sub>3</sub>. According to the emission control measures, NH<sub>3</sub> concentration held still in two scenarios. By calculating the mole ratio of total NH<sub>4</sub><sup>+</sup> to net SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> (PNR), we can assess whether or not the ambient NH<sub>3</sub> is sufficient to neutralize all H<sub>2</sub>SO<sub>4</sub> and HNO<sub>3</sub>. The calculation of the PNR followed **Eq. (8)**. The simulated PNR was 0.23, much less than 1, indicating that ambient conditions in Nanjing are expected to be ammonia-poor (Yin et al., 2018). As the result, the more efficient SO<sub>4</sub><sup>2-</sup> production did impose negative impacts on the formation of NO<sub>3</sub><sup>-</sup> because of the competitive relationship. Both the decrease of NO<sub>x</sub> concentration and the NO<sub>3</sub><sup>-</sup> formation efficiency led to a significant decrease of 26% in NO<sub>3</sub><sup>-</sup>.

### Implications for Future Control Strategy

In this study, the emission control measures during the JDS are proved to be effective for aerosol mitigation but invalid for O<sub>3</sub>. The short-term control strategies during JDS reduced more NO<sub>x</sub>. Based on the nonlinear O<sub>3</sub> formation regime, the decrease in NO<sub>x</sub> probably may fail to suppress  $O_3$  formation and even promote  $O_3$ concentrations under VOCs-controlled condition. To alleviate O3 pollution, we need a more reasonable ratio to reduce NO<sub>x</sub> and VOCs. In addition, the control of VOCs is another difficulty. Model results indicated that VOCs have the lowest mitigation efficiency. A more detailed plan for the reduction of VOCs should be taken into account on the basis of both their accurate local source profile and ozone formation potential (OFP) (Wu and Xie, 2017). It is also noted that secondary transformation as a result of elevated O<sub>3</sub> can compensate for the reduction of PM<sub>2.5</sub>. If the offset effects due to worsen O3 are strong enough, the reduction of PM<sub>2.5</sub> is likely to be ineffective such as the unexpected haze event during COVID-19. This enhancement in O<sub>3</sub> was also witnessed in Europe and India during the COVID-19 outbreak Mertens et al. (2021), Zhang et al. (2021) which tends to be several months. Our study illustrated the similar internal mechanisms in O<sub>3</sub> formation through the short-term case, and provided an insight for improving the understanding in secondary pollution episodes.

At the same time, Li et al. pointed out that  $PM_{2.5}$  can indirectly influence  $O_3$  formation through radical chemistry and photochemistry

as well (Li et al., 2019b; Li et al., 2019c). There is therefore a complicated interaction between  $PM_{2.5}$  and  $O_3$ . This interaction calls for a more synergistic strategy for both controlling  $PM_{2.5}$  and  $O_3$ .

# CONCLUSION

This study investigated the variations of air pollutants during the JDS with observations from surface monitors and satellite retrievals, and then examined the effectiveness of the short-term emission control measures using WRF-Chem. Results showed that the short-term emission control measures could effectively reduce the concentrations of NO2, SO2, CO, VOCs and PM2.5, but failed to suppress the O<sub>3</sub> level. During the JDS, the surface concentrations of NO<sub>2</sub>, SO<sub>2</sub>, CO and VOCs decreased by 20%, 20%, 19% and 15% due to the control measures. In terms of PM2.5, the total concentration was reduced by 17%, but the different aerosol compositions show variable mitigation efficiency (2-26%) because of changes of AOC and competition for ammonia. For O<sub>3</sub>, OPE analysis showed urban areas of Nanjing was under VOClimited. Thus 26% NOx reduction and 35% VOCs reduction in emissions led to an increasement of 3% for O3 concentration. Our study is important for understanding the coordinated control of PM<sub>2.5</sub> and O<sub>3</sub> in typical city clusters, and can provide useful information for future mitigation actions.

# DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/**Supplementary Material**, further inquiries can be directed to the corresponding author.

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# **AUTHOR CONTRIBUTIONS**

Data curation, model simulation, visualization and writingoriginal draft preparation, HZ and KT; Supervision, funding acquisition, writing-review and editing, NL and HL; Data curation, FW and XY. All authors have read and agreed to the published version of the manuscript.

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# SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fenvs.2021.693513/full#supplementary-material

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