Surface ozone changes during the COVID-19 outbreak in China: An insight into the pollution characteristics and formation regimes of ozone in the cold season

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Abstract

The countrywide lockdown in China during the COVID-19 pandemic provided a natural experiment to study the characteristics of surface ozone (O₃). Based on statistical analysis of air quality across China before and during the lockdown, the tempo-spatial variations and site-specific formation regimes of wintertime O₃ were analyzed. The results showed that the O₃ pollution with concentrations higher than air quality standards could occur widely in winter, which had been aggravated by the emission reduction during the lockdown. On the national scale of China, with the significant decrease (54.03%) in NO₂ level from pre-lockdown to COVID-19 lockdown, the maximum daily 8-h average concentration of O₃ (MDA8h O₃) increased by 39.43% from 49.05 to 64.22 μg/m³. This increase was comprehensively contributed by attenuated NOₓ suppression and favorable meteorological changes on O₃ formation during the lockdown. As to the pollution states of different monitoring stations, surface O₃ responded oppositely to the consistent decreased NO₂ across China. The O₃ levels were found to increase in the northern and central regions, but decrease in the southern region, where the changes in both meteorology (e.g. temperature drops) and precursors (reduced emissions) during the lockdown had diminished local O₃ production. The spatial differences in NOₓ levels generally dictate the site-specific O₃ formation regimes in winter, with NOₓ-titration/VOCs-sensitive regimes being dominant in northern and central China, while VOCs-sensitive/transition regimes being dominant in southern China. These findings highlight the influence of NOₓ saturation levels on winter O₃ formation and the necessity of VOCs emission reductions on O₃ pollution controls.

Keywords Surface ozone · Nitrogen dioxide · COVID-19 · Tempo-spatial variation · Formation regime · Winter
1 Introduction

Tropospheric ozone ($O_3$) is one of the most threatening air pollutants in terms of its adverse effects on human health (e.g. inducing respiratory and cardiovascular diseases) and vegetation growth (e.g. foliar injury and yield reduction) (Brunekreef and Holgate 2002; Feng et al. 2015). As a typical secondary air pollutant, $O_3$ in the troposphere is produced mainly from two sources: (1) intrusion from stratospheric air with higher $O_3$ concentrations; and (2) photochemical reactions of precursors including nitrogen oxides ($NO_x$) and volatile organic compounds (VOCs) (Sillman 1999), which play dominant roles in ground-level $O_3$ formation. Due to considerable emissions of $O_3$ precursors from human activities and unfavorable climate conditions (e.g. stagnant hot weather), $O_3$ pollution episodes with non-attainment days occurred frequently worldwide (Kleinman et al. 2005; Tong et al. 2017; Geddes et al. 2021). Besides, obvious increasing trends (1 ~ 5 ppb/year) of surface $O_3$ have been observed among the developed urban areas of North America, Europe and Asia over past decades, which has posed great threats to the health of local people and regional economic development (Xie et al. 2019; Finch and Palmer 2020; Mitchell et al. 2021; Mousavinezhad et al. 2021).

Research on surface $O_3$ pollution began in the early 1970s, and since then great efforts have been made on investigating the chemical mechanism of $O_3$ production and loss (Guicherit and Vandop 1977; Bojkov 1986; Sillman 1999; Jacob 2000; Vingarzan 2004; Wang et al. 2017). In the troposphere, VOCs could be oxidized by the hydroxyl radical (OH) to produce peroxy radicals, which could further react with nitrogen monoxide (NO) to produce nitrogen dioxide ($NO_2$). Photolysis of $NO_2$ at wavelengths ≤ 424 nm could result in the production of atomic oxygen (O), which finally prompts the $O_3$ formation via the combination reaction of atomic oxygen and molecular oxygen ($O_2$). As a widely confirmed common feature, surface $O_3$ formation is nonlinearly dependent on its precursors (i.e. VOCs and $NO_x$). The relative contents of VOCs and $NO_x$ in the troposphere control the surface $O_3$ formation regime, with $O_3$ variation being sensitive to the less abundant air pollutants (Sillman and He 2002; Jin and Holloway 2015; Wang et al. 2017). In general, surface $O_3$ is likely to be $NO_x$-sensitive in remote and rural areas, where the air masses are aged with less freshly emitted air pollutants and low $NO_x$/VOCs ratios (Spirig et al. 2002; Wang et al. 2010; Ding et al. 2013). On the contrary, it tends to be VOCs-sensitive ($NO_x$-saturated) in urban and populous areas, where the $NO_x$/VOCs ratios are usually high and surface $O_3$ will decrease with reduced VOCs emissions but increase with reduced $NO_x$ emissions. In polluted urban areas with much higher levels of NO, $O_3$ production could be strongly suppressed by NO ($NO + O_3 \rightarrow NO_2 + O_2$). This situation is usually referred to as the $NO_x$-titration regime, which is also characterized by increased $O_3$ with decreased $NO_x$ (Wang et al. 2017). The $NO_x$-titration chemistry mainly occurs in regions characterized by net loss of surface $O_3$, especially during winter and nighttime. This is different from VOCs-sensitive chemistry, which occurs in regions characterized by photochemical $O_3$ production (Sillman 1999). In addition to the spatial variation, the $O_3$ sensitivity to precursors was also found to change obviously with time owing to varied atmospheric chemical conditions. For example, Mazzuca et al. (2016) found that $O_3$ production tended to be VOCs-sensitive in the morning and $NO_x$-sensitive in the afternoon in Houston, America; Wang et al. (2019) found that $O_3$ formation in eastern China had changed from VOCs-sensitive to the mixed regimes of VOCs-sensitive and $NO_x$-sensitive from 2012 to 2016, due to significant $NO_x$ reductions (> 25%). Accurate determination of the dominant precursors for $O_3$ formation is difficult given the tempo-spatial variability of $O_3$ sensitivity, but it is of great significance to the development of effective $O_3$ reduction strategies.
Besides the influence of precursors, meteorology can also play an important role in the variation of surface O₃ by affecting precursor concentrations and photochemical conditions. Generally, surface O₃ level is positively correlated with solar radiation and air temperature, and negatively correlated with relative humidity and wind speed (Abdul-Wahab et al. 2005; Pudasainee et al. 2006). Elevated O₃ concentrations are often observed on days with strong solar radiation, high air temperature and low wind speeds, which favor the photochemical production and accumulation of O₃ (Toh et al. 2013; Han et al. 2020). Meteorological effects on surface O₃ could cover or even reverse the effects of precursors. For example, a recent investigation found that the long-term trend of surface O₃ in eastern and central China could change from decreasing to increasing after removing the meteorological effects (Lin et al. 2021). Therefore, both meteorology and precursors should be taken into consideration before an accurate conclusion being reached on the O₃ pollution mechanism and efficient pollution control measures.

As O₃ levels are usually high under intense sunlight and high air temperature, most studies have focused on O₃ pollution in relatively warm seasons with limited researches being carried out in winter. However, high O₃ episodes could also occur during wintertime due to unfavorable climate conditions (e.g. temperature inversion) in the cold season (Edwards et al. 2014; Mansfield and Lyman 2021). With the intensification of global warming and the enforcement of pollution control policy (e.g. NOₓ emission reduction for winter haze mitigation), unanticipated upward trend of winter O₃ concentration has been observed recently in Central Europe and East Asia (Feng et al. 2020; Gebhardt et al. 2021; Li et al. 2021a, b). During the wintertime, both human immunity and air quality usually get worse with the temperature drop (Dowell 2001; Wang et al. 2014). Elevated wintertime O₃, although with relatively low ambient levels, might still pose risks to human health via both direct oxidative stress (Di et al. 2017) and indirect health effects from boosting the formation of secondary particulate matter in winter (Huang et al. 2021). Therefore, a better understanding of winter O₃ formation is significant to air quality improvement and protection of human health.

In late December of 2019, a novel coronavirus disease (COVID-19) was identified, which has spread fast to become a global pandemic. In order to slow down its infection, most countries have taken social distancing measures with strict restriction on industrial production and transportation. As a consequence of the lockdown, anthropogenic emissions of primary air pollutants (e.g. NOₓ and VOCs) as well as their atmospheric concentrations decreased significantly (Li et al. 2020; Venter et al. 2020; Doumbia et al. 2021). This unexpected global event provided a natural experiment to investigate the response of surface O₃ to emission reductions of its precursors, with a great number of researches being carried out on this subject since its outbreak. As to the related studies in China, more than 200 scientific papers on O₃ pollution during the pandemic have been published till now, with most of them focusing on the O₃ variation characteristics and regimes of specific locations and regions. For example, using comprehensive measurements and modeling techniques, Le et al. (2020) found that the reduced NOₓ during pandemic lockdown resulted in significant O₃ enhancement in urban areas of Wuhan, Beijing, Shanghai and Guangzhou and the Beijing-Tianjin-Hebei (BTH) region of China, due to the nonlinear production chemistry and titration of O₃ in winter; Li et al. (2020) and Huang et al. (2021) found an obvious increase in surface O₃ of eastern China in response to the sharply NOₓ decrease during the COVID-19 lockdown, which in turn facilitated the formation of secondary particulate matter; Xu et al. (2020) investigated the impact of COVID-19 event on air quality of central China, and reported obvious increases by 12.7%, 14.3%, and 11.6% in January, February, and March, respectively, for the wintertime O₃. Based on the satellite
remote sensing technique, Ghahremanloo et al. (2021) found that the significant increases in surface O$_3$ in East China from February 2019 to February 2020 are likely the result of significantly decreased NO$_x$ concentrations and less NO$_x$ saturation during the daytime. In comparison with the extensive studies on city or regional scales, very limited researches about the impact of COVID-19 pandemic on surface O$_3$ have been done on the national scale of China. For example, based on model simulations, Zhao et al. (2020) examined the temporal–spatial variations of six air pollutants including O$_3$ during the COVID-19 lockdown, with significant O$_3$ increase being found caused by the nonmeteorological factors. In contrast, Liu et al. (2021) found that both meteorological changes and emission reductions contributed to the O$_3$ increase during the COVID-19 lockdown, with O$_3$ changes depending on the levels of NO$_x$ and VOC reductions in different regions of China. In this study, the changes of nationwide surface O$_3$ in winter during the COVID-19 lockdown were analyzed, and the major influencing factors on surface O$_3$ over different regions of China were evaluated. In comparison with the previous studies, which depended mainly on modeling techniques, this study was entirely based on observations and statistical analyses. A simple procedure was firstly developed in this research for removing the meteorological effects when determining the dominant precursor for O$_3$ formation specific to each monitoring station (detailed in the Methods Section). Based on these analyses in the context of the specific event of COVID-19 pandemic, it is expected to provide useful insights into the formation mechanism and reduction measures on surface O$_3$ of China.

2 Materials and methods

2.1 Study period and monitoring data

This study focused on the O$_3$ variation from December 25, 2019 to February 22, 2020, which is divided into two time periods, i.e. one month before (December 25, 2019~January 23, 2020) and one month after (January 24~February 22, 2020) the initiation of the first-level public health emergency response to COVID-19 pandemic in China. Hourly data of surface O$_3$ and NO$_2$ at 1641 ground stations from 31 provinces were obtained from the China National Environmental Monitoring Center (http://106.37.208.233:20035/). Meteorological data including air temperature (TEMP), relative humidity (RH), pressure (P), wind direction (WD), wind speed (WS) and rainfall were acquired from National Oceanic and Atmospheric Administration (NOAA)’s National Climate Data Center archive (http://www.ncdc.noaa.gov/oa/ncdc.html).

In this study, the daily average concentrations of NO$_2$ (DAV NO$_2$) and the maximum daily 8-h average concentration of O$_3$ (MDA8h O$_3$) were calculated for statistical analyses. As NO$_x$ (= NO + NO$_2$) was not the conventional pollution index and was unavailable to this study, NO$_2$ was used as a surrogate marker of NO$_x$ given the close positive relationship between these two indices (Clapp and Jenkin 2001). In order to address any potential problem concerning data quality, a filtering procedure was performed as follows (Yin et al. 2019; Mousavinezhad et al. 2021): (1) all the non-positive values were eliminated; (2) the data with values greater than P$_{75}$ + 1.5 × (P$_{75}$−P$_{25}$) or less than P$_{25}$−1.5 × (P$_{75}$−P$_{25}$) (P$_{25}$ and P$_{75}$ represent the 25th and 75th percentiles, respectively) were removed as outliers; (3) only the daily statistical values were retained if more than 70% of its hourly values were valid; (4) only the data of a station were used if more than 70% of its daily statistical values were valid. After applying this criterion on data quality control, the 1641 stations were finally reduced to 1451 for further analysis in this study.
2.2 Data analyses

2.2.1 Statistical analysis

Independent-samples t test and non-parametric tests (Kolmogorov–Smirnov Z) on two independent samples were used to evaluate whether there are statistically significant differences between the means of air pollutants for the periods before and after the initiation of public health emergency response to COVID-19 pandemic. Results were given as the average (avg) ± the standard deviation (sd), with the differences being considered as significant if \( p < 0.05 \). Spearman correlation analysis was used to check the relationships between surface \( O_3 \) and other variables (\( NO_2 \) and meteorological parameters). All data statistics were performed using the statistical package SPSS 19.0 (SPSS Inc., Chicago, IL, USA).

2.2.2 Removing the meteorological effects

The variations of \( O_3 \) with its precursors, which is indicative of the \( O_3 \) formation regimes (e.g. VOCs-sensitive or \( NO_x \)-sensitive), could be masked by its responses to the variations of meteorological variables (Lin et al. 2021). In order to remove the meteorological effects and accurately determine the \( O_3 \) formation regimes for each monitoring site, a simple statistical procedure was developed and applied to the dataset. Firstly, the major meteorological variables which are significantly correlated with surface \( O_3 \) were selected based on the correlation analysis. Secondly, for each of the dominant meteorological variables and the two air quality variables (MDA8h \( O_3 \) and DAV \( NO_2 \)), the differences between the daily values for each of the 30 days during the COVID-19 lockdown and those for each of the 30 days during the pre-lockdown were calculated. Therefore, a total of 900 differences were obtained for each variable of each monitoring site. Thirdly, only the differences of MDA8h \( O_3 \) and DAV \( NO_2 \) were adopted for further analysis on \( O_3 \) formation regimes if the corresponding absolute differences for the dominant meteorological variables were lower than certain critical values. In terms of this study, the critical values for the major meteorological variables, i.e. temperature, relative humidity and wind speed, were selected to be 1 °C, 5% and 1 m/s, respectively, based on the following reasons: (1) The ranges of variation for daily average temperature, relative humidity and wind speed from pre-lockdown to COVID-19 lockdown are 0 ~ 30°C, 0 ~ 68% and 0 ~ 6 m/s, respectively, during the study period. Given these big changes in meteorological variables on the nationwide scale, the relatively small meteorological fluctuations, which are defined by the selected critical values, were assumed to be unable to alter the general trend of variation of surface \( O_3 \) with its precursors, especially when the precursor emissions plummeted due to the enforcement of strict lockdown measures; (2) There are enough data values left for each station for further analyses on \( O_3 \) formation regime after applying the selected critical values to removing the meteorological effects on \( O_3 \) variations. Finally, the average differences for MDA8h \( O_3 \) and DAV \( NO_2 \), which are independent of the influence of major meteorological factors, were calculated for each monitoring site.
3 Results and discussion

3.1 Temporal variations of surface $O_3$ and $NO_2$ in response to the COVID-19 lockdown

The time series of geographically (1451 stations) averaged MDA8h $O_3$ and DAV $NO_2$ before and during the COVID-19 lockdown in China were shown in Fig. 1, which indicated obviously increased concentration of surface $O_3$ but decreased concentration of $NO_2$ during the lockdown. On the national scale of China, the average value of $NO_2$ during the COVID-19 lockdown was significantly ($p < 0.001$) lower than that during the pre-lockdown period (Table 1 and Fig. 2), with the decrease rate of 54.03%. This

Table 1 The statistics (avg ± std) of $O_3$, $NO_2$ and meteorological variables before and during the COVID-19 lockdown in China

|                      | Pre-lockdown | COVID-19 lockdown | DIFF* | Change rate (%) |
|----------------------|--------------|-------------------|-------|-----------------|
| MDA8h $O_3$ (μg/m³) | 49.05±12.90  | 64.22±8.35        | 15.17±13.86 | 39.43±38.80    |
| DAV $NO_2$ (μg/m³)  | 38.73±14.50  | 17.85±8.56        | -20.89±8.44 | -54.03±11.38   |
| TEMP (°C)           | 2.89±9.18    | 4.15±7.56         | 1.26±2.09   | 96.84±446.38   |
| RH (%)              | 71.43±11.73  | 66.11±12.95       | -5.32±5.53  | -7.73±9.49     |
| P (hPa)             | 1025.29±3.95 | 1025.77±3.34      | 0.47±1.22   | 0.05±0.12      |
| WS (m/s)            | 2.26±0.45    | 2.46±0.51         | 0.20±0.23   | 9.37±10.76     |
| Rainfall (mm)       | 66.78±74.38  | 90.76±97.47       | 23.98±88.45 | 312.27±942.98  |

*DIFF: Significant differences ($p<0.001$) for different variables between pre-lockdown and COVID-19 lockdown
decline in the concentration of O₃ precursor was mainly caused by the reductions in primary pollutant emissions due to the sharp restriction in transportation and industrial production during the pandemic in China (Doumbia et al. 2021; Zhang et al. 2021). On the contrary, the average MDA8h O₃ (64.22 μg/m³) during the COVID-19 lockdown was significantly (p < 0.001) higher than that (49.05 μg/m³) before the lockdown, with the increase rate of 39.43%. This increase in O₃ level was also accompanied with an hour delay (from 15:00 to 16:00) for the peak time of hourly O₃ concentration (Fig. 2). As reported by Strode et al. (2019), the changing emissions of precursor gases could significantly affect the diurnal cycle of surface O₃. During the COVID-19 lockdown in China, the diurnal human activities and precursor emissions on the national scale were generally postponed, which can be partly indicated by the delay (from 14:00 to 15:00) of diurnal valley values of NO₂ from pre-lockdown to COVID-19 lockdown (Fig. 2). This might account for the one hour delay of the diurnal O₃ peak occurrence. During the wintertime, the general NOₓ levels are usually high in China due to increased emissions from domestic heating and adverse diffusion conditions with lowered atmospheric boundary layer (Lin 2012). Under this situation, O₃ production is usually sensitive to VOCs and is suppressed by NOₓ. The significant O₃ suppression by NOₓ in cold seasons has been confirmed over most areas of China (Liu et al. 2010; Lu et al. 2019; Yang et al. 2019), which means that the O₃ production is inversely related to NOₓ emission during the wintertime. This relationship might largely account for the significant increase in MDA8h O₃ during the lockdown (Table 1), when significant decreases of NOₓ was observed in China. Besides the precursor influence on the overall O₃ level in China, meteorological variables might also play a role in surface O₃ variations during the study period. During the COVID-19 lockdown, significant increase and decrease were observed in air temperature and relative humidity, respectively (Table 1 and Fig. 1), which had probably promoted the O₃ rise during the lockdown. This positive influence
of meteorology on surface O$_3$ in China during the pandemic has also been confirmed by Gaubert et al. (2021) and Yin et al. (2021).

3.2 Changes in the spatial distributions of O$_3$ and NO$_2$ due to the COVID-19 lockdown

For the 1451 ground stations analyzed in this study, a total of 1450 (99.93%) stations exhibited decreased NO$_2$ in response to the COVID-19 lockdown (Fig. 3). The changes in NO$_2$ for most of the stations were within -40 ~ -5 μg/m$^3$ (Fig. 4). Bigger drops in

![Spatial distributions of averaged MDA8h O$_3$ and DAV NO$_2$ during the periods of pre-lockdown (a) and COVID-19 lockdown (b), and the differences (c) between the two periods](image)

![Variations of station numbers with the differences of MDA8h O$_3$ and DAV NO$_2$ between pre-lockdown and COVID-19 lockdown](image)
NO₂ (<-30 μg/m³) were found in Central East China, where higher levels of NO₂ were observed during the pre-lockdown period (Fig. 3). As to the changes of surface O₃, a total of 1256 stations (86.56%), which mainly located in the northern and central areas of China, showed elevated levels in MDA8h O₃, with the changes for most stations in the range of 0 ~ 40 μg/m³ (Figs. 3 and 4). The stations with bigger increases (> 30 μg/m³) in surface O₃ mostly located in Central East China, which is generally in accordance with the significant decreases in its precursors.

Based on the O₃ ambient air quality standards (AAQS) for human health and ecological environment protection in China (MDA8h O₃ of Grade I: 100 μg/m³, and Grade II: 160 μg/m³), the number of stations with at least one non-attainment day (i.e. pollution episode) over AAQS Grade I has increased from 260 (17.91%) during the pre-lockdown to 807 (55.58%) during the COVID-19 lockdown (Table 2). While for the AAQS Grade II, the number of exceeding stations has decreased from 39 (2.69%) during the pre-lockdown to 22 (1.52%) during the COVID-19 lockdown. During the period of pre-lockdown, most of the stations with non-attainment days in surface O₃ mainly located in the coastal regions of southeast China (Fig. 5a). In contrast, the stations with O₃ ever the AAQS spread all over China during the COVID-19 lockdown (Fig. 5b). These results indicated that: (1) O₃ pollution episodes with adverse health effects could happen in a large area of China during the COVID-19 lockdown of wintertime, which has also been reported in previous study by Li et al. (2021a); (2) The spatial scope of the winter O₃ pollution in China had expanded greatly from the period of pre-lockdown to that of the COVID-19 lockdown.

**Table 2** Statistics on the stations exceeding the ambient air quality standards

| Grade | Pre-lockdown | COVID-19 lockdown |
|-------|--------------|-------------------|
| Grade I (100 μg/m³) | 260 | 807 |
| Percent of exceeding stations % | 17.91 | 55.58 |
| Average exceeding days | 3.67 | 2.99 |
| Range of exceeding days | 1 ~ 18 | 1 ~ 19 |
| Grade II (160 μg/m³) | 39 | 22 |
| Percent of exceeding stations % | 2.69 | 1.52 |
| Average exceeding days | 1.23 | 1.14 |
| Range of exceeding days | 1 ~ 2 | 1 ~ 2 |

**Fig. 5** Spatial distribution of stations with MDA8h O₃ exceeding AAQS for at least one day during the study periods of pre-lockdown (a) and COVID-19 lockdown (b)
It is worth noting that although most (86.56% in percent) stations had exhibited elevated O₃ levels during the lockdown, there is still a considerable amount (195) of stations showing obvious decreased O₃ concentrations (Fig. 3c), with most of them locating in the south region of China. In the meantime, the severe O₃ pollution episodes (> 160 μg/m³ of Grade II AAQS), which occurred mainly in the southern coastal region of China before the lockdown, has also been reduced during the COVID-19 lockdown (Table 2 and Fig. 5). Two possible reasons might account for this tempo-spatial difference in surface O₃ during the study period: (1) The NOₓ concentrations in winter are relatively low in the southern part of China (Fig. 3a), where no winter heating with large NOₓ emissions was applied due to the relatively high air temperature in winter (Fig. 6). Therefore, surface O₃ was unlikely to be in the NOₓ-titration regime in the southern China. As reported by previous researches, surface O₃ in southern coastal area of China (e.g. Pearl River Delta region) is in the VOCs-sensitive (Liu et al. 2010; Zou et al. 2015) or transitional regimes (Jin and Holloway 2015; Liu et al. 2021) in winter. This could lead to the O₃ decreases with the simultaneous emission reductions of both precursors during the COVID-19 lockdown (Liu et al. 2021); (2) As suggested by the recent studies of Hu et al. (2021a) and Ma et al. (2021), meteorological factors could greatly affect the tempo-spatial variations of surface O₃ in China. During the study period, most stations with decreased MDA8h O₃ during the COVID-19 lockdown located in the regions characterized by decreased temperature, increased relative humidity (RH) and rainfall (Fig. 6c). These meteorological changes, which were unfavorable to atmospheric O₃ formation, might comprehensively lead to the decrease in surface O₃ during the COVID-19 lockdown. Similar limiting effects on regional O₃ production from meteorological variables in China has also been reported by Lin et al. (2021).

3.3 Determination of the O₃ formation regimes for different monitoring sites

As has been discussed in the previous section, both increase and decrease in O₃ concentrations have been observed among the monitoring sites with the consistent decreased NO₂ during the COVID-19 lockdown (Fig. 3). This indicated that the O₃ sensitivities to its precursors might be different for the monitoring sites. Based on the correlation analysis, the dominant meteorological factors affecting winter O₃ were identified to be air temperature, relative humidity and wind speed, which showed significant correlations (p < 0.01) with surface O₃ (Table 3). Using the statistical procedure introduced in Sect. 2.2.2, the influences of these factors on O₃ variations were eliminated. As shown in Fig. 7a, after removing the major meteorological effects, a total of 1166 sites (Type I) showed increased O₃ concentrations with the decreases of NO₂ during the lockdown period. This indicated that the majority of the ground monitoring stations had the NOₓ-titration or VOCs-sensitive regime of O₃ formation, which is characterized by increased O₃ with decreased NOₓ (Sillman 1999). The levels of NOₓ saturation for these sites were probably high, which had significantly suppressed local O₃ formation during the pre-lockdown period. As reported in the recent studies on air quality changes during the COVID-19 pandemic (Doumbia et al. 2021; Gaubert et al. 2021; Hu et al. 2021b; Li et al. 2021a; Liu et al. 2021; Wang et al. 2021), besides the reduced NOₓ emissions, the VOC emissions as well as its ambient concentrations across China had also been reduced due to the countrywide restriction on both transportation and industrial activities. For the 1166 monitoring sites of Type I, the VOCs emission reductions during the lockdown, which could have led to the decrease in O₃ levels, were probably not large enough to offset the promotion effects on O₃ increase from NOₓ reduction. This predominant effect
of NO\textsubscript{x} reduction on O\textsubscript{3} increase has been widely confirmed by recent studies on air quality changes during the COVID-19 lockdown (Shi and Brasseur 2020; Liu et al. 2021; Yin et al. 2021), which suggests that the conventional emission reduction strategy (NO\textsubscript{x} reduction only) might be less sufficient to reduce surface O\textsubscript{3} concentrations in China.

Fig. 6 Spatial distributions of averaged meteorological variables during the periods of pre-lockdown (a) and COVID-19 lockdown (b), and the differences (c) between the two periods
In contrast with the widely observed O₃ rise in China, there is still a considerable amount of sites (223 in total, Type II) showing simultaneous decreases in both O₃ and NO₂ concentrations (Fig. 7a). The pre-lockdown NO₂ concentrations for the sites of Type II were generally lower than those for the sites of Type I (Fig. 7b). This indicated a relatively low level of NOₓ saturation for the minor group of sites, which could facilitate the formation of the non-NOₓ-titration regimes (i.e. NOₓ-sensitive or VOCs-sensitive) for surface O₃ production. As the simultaneous decreases in both NOₓ and VOCs emissions, which is the case during the COVID-19 lockdown, could comprehensively lead to the net O₃ declines no matter surface O₃ is sensitive to NOₓ, or to VOCs or in the transition state between both regimes, it is not easy to determine the precise O₃ formation regimes for the 223 sites. Based on the previous studies concerning O₃-NOₓ-VOCs chemistry (Sillman 1999; Sillman and He 2002; Peralta et al. 2021; Yang et al. 2021), it can be inferred that the NOₓ-sensitive regime was usually characterized by a higher ratio (>4) of concentration changes between O₃ and NOₓ (i.e. ΔO₃/ΔNOₓ), and insensitivity of O₃ to VOC changes alone. As the data of NOₓ (sum of NO and NO₂) for the monitoring sites were not available to this study, two extreme scenarios with much low (10%) and high (90%) NO₂/NOₓ ratios, which are typical of the ambient NO₂ fractions for the polluted urban sites and remote clean sites, respectively, were adopted for the calculations of NOₓ concentrations and the ΔO₃/ΔNOₓ ratios. As shown in Fig. 8, the ΔO₃/ΔNOₓ ratios for the polluted scenario were lower than 0.8 in all the sites, while those for the clean scenario were lower than 4 in 96.9% of all the sites. This indicated that the O₃ formation for most of the 223 sites (Type II) was probably in the VOCs-sensitive or the transition regime.

Across the whole country of China, the sites with VOCs-sensitive/transition regimes of O₃ formation (Type II: low level of NOₓ saturation) were scattered near the sites with

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### Table 3 Spearman correlation coefficients between surface O₃ and other environmental variables

|          | DAV NO₂ | TEMP | RH     | P     | WS    | WD    | Rainfall |
|----------|---------|------|--------|-------|-------|-------|----------|
| MDA8h O₃| -0.751**| 0.224*| -0.591**| 0.137 | 0.270*| 0.114 | -0.209   |

* indicates p<0.01; ** indicates p<0.001
NO\textsubscript{x}-titration/VOCs-sensitive regimes (Type I: high level of NO\textsubscript{x} saturation) (Fig. 7a), which means that the O\textsubscript{3} sensitivities to precursors could change even within small spatial regions. This might be accounted for by the spatial difference of precursor emissions related to the urban–rural development gradient, with different levels of O\textsubscript{3} precursors among different city areas (i.e. central, suburban and rural) (Geng et al. 2008; Wang et al. 2017). Previous studies based on ground monitoring data (e.g. Liu et al. 2021; Yang et al. 2021) and satellite retrievals (e.g. Jin and Holloway 2015) had also showed this mixed state of O\textsubscript{3} formation regimes around China. Although varied regimes of O\textsubscript{3} formation could be observed across China, the NO\textsubscript{x}-titration/VOCs-sensitive regimes (Type I) were predominant in northern and central China during the wintertime, while the VOCs-sensitive/transition regimes (Type II) were predominant in the southern China. The NO\textsubscript{x} concentrations were different across China with generally higher and lower levels in northern and southern regions, respectively, during the cold season (Fig. 3a). This could significantly affect the region-specific O\textsubscript{3} responses to precursor reductions during the COVID-19 lockdown, which might account for the spatial distribution differences for varied O\textsubscript{3} formation regimes.

### 4 Conclusions

Our results showed that the O\textsubscript{3} pollution with concentrations higher than the Chinese AAQS could occur widely in China in the recent winter, and the pollution state had been worsened by the emission reduction and meteorological changes during the COVID-19 lockdown. On the national scale of China, surface O\textsubscript{3} increased significantly with the drastic decrease of NO\textsubscript{2} concentration, which could be accounted for by the reduced suppression effects of NO\textsubscript{x} as well as the favorable meteorological conditions (e.g. increased air temperature) on O\textsubscript{3} formation during the lockdown period. As to the pollution states of different monitoring stations, with the consistent decrease in NO\textsubscript{x}, surface O\textsubscript{3} responded oppositely for the sites in different regions of China. In contrast with the O\textsubscript{3} increases in the northern and central China, obvious decreases in O\textsubscript{3} levels had been observed in south China. The low NO\textsubscript{2} concentrations in the southern part of China indicated a less or non-saturated state of NO\textsubscript{x}, which had contributed to the O\textsubscript{3} decrease with the substantial precursor reductions during the COVID-19 lockdown. Besides, the changes in meteorology (e.g. decreased temperature and increased relative humidity) in south China during the lockdown had further promoted surface O\textsubscript{3} declines.
After removing the major meteorological effects on O$_3$ variations, the site-specific formation regimes of wintertime O$_3$ in China were analyzed in this study. Most of the monitoring sites were characterized by the NO$_x$-titration/VOCs-sensitive regimes of O$_3$ formation, while a minority of the sites had the regimes toward the transition zone. Although the sites with different O$_3$ formation regimes were distributed in a mixed state across the country, the sites with the NO$_x$-titration/VOCs-sensitive regimes (high NO$_x$-saturation level) were dominant in the northern and central China, while those with the VOCs-sensitive/transition regimes (low NO$_x$-saturation level) were dominant in the southern China. This highlights the importance of implementing (1) VOCs emission controls throughout the country, and (2) the region-specific emission controls according to local NO$_x$ levels for effective O$_3$ mitigation. In northern and central China, VOCs emission reductions should be strengthened to achieve the net declines of surface O$_3$. Stringent regional joint-control within these regions might be an effective measure. While in southern China, where the NO$_x$ levels were generally low, cooperative controls on both NO$_x$ and VOCs emissions will be applicable. Given the increased winter O$_3$ in the recent year, which might causing adverse health effects both directly and indirectly by enhancing the atmospheric oxidizing capacity (Feng et al. 2020; Huang et al. 2021; Li et al. 2021a), more attention should be paid to the pollution regimes of winter O$_3$ in China, and the findings of our study could provide some useful insights into this issue.

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**Data availability** The datasets generated during and/or analyzed during the current study are available from the corresponding authors on reasonable request.

**Declarations**

**Competing interests** The authors have no relevant financial or non-financial interests to disclose.

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