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1 Diverse response of surface ozone to COVID-19 lockdown in China

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13 Abstract: Ozone (O_3) is a key oxidant and pollutant in the lower atmosphere. Significant increases in surface O_3 have been 14 reported in many cities during the COVID-19 lockdown. Here we conduct comprehensive observation and modeling analyses of surface O₃ across China for periods before and during the lockdown. We find that daytime O₃ decreased in the subtropical 15 south, in contrast to increases in most other regions. Meteorological changes and emission reductions both contributed to the 16 17 O₃ changes, with a larger impact from the former especially in central China. The plunge in nitrogen oxide (NO_x) emission 18 contributed to O₃ increases in populated regions, whereas the reduction in volatile organic compounds (VOC) contributed to 19 O_3 decreases across the country. Due to a decreasing level of NO_3 saturation from north to south, the emission reduction in 20 NO_x (46%) and VOC (32%) contributed to net O₃ increases in north China; the opposite effects of NO_x decrease (49%) and 21 VOC decrease (24%) balanced out in central China, whereas the comparable decreases (45-55%) in the two precursors 22 contributed to net O₃ declines in south China. Our study highlights the complex dependence of O₃ on its precursors and the 23 importance of meteorology in the short-term O₃ variability.

- 24 Keywords: Surface ozone, meteorological condition, emission reduction, COVID-19
- 25

26 1 Introduction

27

28	of deaths(WHO, 2020). China, where the first case of COVID-19 was reported in the city of Wuhan, imposed country-wide
29	measures from 23 January to 13 February 2020 to prevent the spread of the disease, including social distancing, teleworking,
30	and closure of non-essential businesses (Chinazzi et al., 2020; Li et al., 2020). These restrictions drastically reduced
31	anthropogenic activities, resulting in a sharp decrease in emissions of air pollutants (Doumbia et al., 2020; Huang et al., 2020;
32	Wang et al., 2020a).
33	The huge and large-scale emission reductions during the COVID-19 lockdown can be treated as a natural outdoor experiment
34	to improve our understanding of the air pollutant's response to emission control. According to satellite and surface observations,
35	compared with the period before the lockdown, nitrogen dioxide (NO2) concentrations decreased by over 50% in China during
36	the lockdown period(Bauwens et al., 2020; Liu et al., 2020; Shi and Brasseur, 2020; Zhang et al., 2020). The concentrations
37	of other pollutants, including SO ₂ , particulate matter with an aerodynamic diameter less than 2.5 µm (PM _{2.5}), particulate matter
38	with an aerodynamic diameter less than 10 µm (PM10), and carbon monoxide (CO), also declined in a large area of
39	China(Miyazaki et al., 2020; Wang et al., 2020b). However, surface ozone (O ₃) concentrations in northern and central China
40	increased by over 100%(Lian et al., 2020; Shi and Brasseur, 2020). Similar O3 increases have been reported in southern Europe,
41	India, and Brazil despite the large decrease in other pollutants(Sharma et al., 2020; Sicard et al., 2020; Siciliano et al., 2020).
42	However, the underlying factors driving the O ₃ changes during the city lockdowns remain unclear.
43	Surface O ₃ is produced by photochemical reactions of ozone precursors, NO _x , volatile organic compounds (VOCs), and carbon

The outbreak of coronavirus disease 2019 (COVID-19) has severely threatened public health worldwide, leading to millions

44 monoxide (CO) and can also be transported from higher levels of the atmosphere and from outside regions (Akimoto et al., 45 2015; Liu and Wang, 2020a; Roelofs and Lelieveld, 1997). It is well known that O_3 has a non-linear dependence on its 46 precursors and that NO_x can either decrease or increase O_3 depending on the relative abundance of NO_x to VOCs (Atkinson, 47 2000; Wang et al., 2017a). In general, the O_3 production in urban areas with high $NO_x/VOCs$ ratios is VOCs limited, and 48 reducing NO_x emissions can increase O_3 due to decreased titration of O_3 and radicals. In addition to the two precursors,

49	particulate matters can influence ozone by altering the solar irradiance and chemical reactions on aerosol surfaces (Li et al.,
50	2019b; Liu and Wang, 2020b; Stadtler et al., 2018). Meteorological factors affect surface ozone by changing transport pattern,
51	wet and dry depositions, chemical reaction rates, and natural emissions (Liu and Wang, 2020a; Lu et al., 2019).
52	The responses of ozone (and other air pollutants) to short-term emission reductions have been previously studied for a number
53	of public and political events in China, such as the Beijing Summer Olympic Games (August 2008), the Asia-Pacific Economic
54	Cooperation (APEC) meeting in Beijing (November 2014), and the G20 summit in Hangzhou (September 2016). During these
55	events, various emission-reducing measures were implemented in the cities concerned and their surrounding areas. Whereas
56	atmospheric concentrations of primary air pollutants (NOx, CO, primary PM, and SO2) in the concerned cities generally
57	decreased in response to the temporary control measures, the O3 concentrations showed mixed responses. O3 decreased after
58	emission reductions for some events (Huang et al., 2017; Wang et al., 2017b) but increased in others (Wang et al., 2010; Wang
59	et al., 2015; Wu et al., 2019). The different O ₃ responses have been qualitatively attributed to differences in the meteorological
60	conditions (including regional transport of air masses) and to different control measures implemented by the local governments.
61	Compared with the previously studied situations, the COVID-19 lockdown is unique in that emissions decreased across the
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70 2 Materials and Methods

2.1 Surface measurement data

We obtained the observed concentrations of surface O₃ and other pollutants (PM_{2.5}, PM₁₀, SO₂, CO, NO₂) at 1643 stations from the China National Environmental Monitoring Center (<u>http://106.37.208.233:20035/</u>). Data quality control was conducted for the measurement data in accordance with previous studies(Lu et al., 2018; Song et al., 2017). Fig. 1 shows the locations of these environmental monitoring sites.

76	The country-wide measures to control the spread of COVID-19 were implemented starting from 23 January 2020 (the exact
77	date varies for different cities), just before the Chinese New Year. All enterprises remained closed until no earlier than 13
78	February, except those required for essential public services, epidemic prevention and control, and residential life needs. We
79	focused on the period during the COVID-19 lockdown from 23 January to 12 February 2020 (hereafter referred to as the CLD
80	period), 3 weeks in total. We derived the changes in O ₃ and other pollutants by comparing the CLD period with the 3 weeks
81	before the COVID-19 outbreak, from 2 to 22 January 2020 (hereafter referred to as the pre-CLD period). We focused on three
82	typical regions in China (Fig. 1): north China (NC, 35-41.5°N, 113-119°E, including Beijing, Tianjin, Hebei, and western
83	Shandong), central China (CC, 28.8-33°N, 108-117°E, including Hubei province, where Wuhan is situated, and the surrounding
84	regions), and south China (SC, 21.5-24°N, 111-116°E, including the Pearl River Delta and the surrounding regions). The NC
85	region is situated in the North China Plain, which is known to suffer from severe haze in winter; CC was the original epicenter
86	of the COVID-19 outbreak in China and is an important economic hub for the central regions of China; the Pearl River Delta,
87	where the megacities of Guangzhou and Shenzhen are situated, is the most developed region in southern China.

88 2.2 Numerical modeling

The CMAQ model (Community Multiscale Air Quality model, v5.2.1) was applied to simulate the O₃ mixing ratios over China from 2 January to 12 February 2020. The WRF model (Weather Research and Forecasting model, v3.5.1) was driven by the dataset of the National Center for Environmental Prediction (NCEP) FNL Operational Model Global Tropospheric Analyses with a horizontal resolution of $1^{\circ} \times 1^{\circ}$ and provided meteorological inputs for the CMAQ model. The CMAQ target domain covered the continental China at a horizontal resolution of 36 km × 36 km. SAPRC07TIC (Carter, 2010; Hutzell et al., 2012) and AERO6i (Murphy et al., 2017; Pye et al., 2017) were adopted as the gas-phase chemical mechanism and aerosol

95	mechanism, respectively. The CMAQ model has been improved with updated heterogeneous reactions to better predict the O ₃
96	concentration; details can be found in Liu and Wang (2020a). Although the WRF-CMAQ model was run in offline mode, the
97	CMAQ model employs an in-line method that uses the concentrations of particles and O ₃ predicted within a simulation to
98	calculate the solar radiation and photolysis rates (Binkowski et al., 2007). As a result, the effect of aerosol on O3 concentrations
99	via changing the photolysis rates were also considered in the simulation. The chemical boundary conditions were provided by
100	the results of Whole Atmosphere Community Climate Model (WACCM, https://www.acom.ucar.edu/waccm/download). The
101	anthropogenic emissions in China were obtained from the Multi-resolution Emission Inventory for China (MEIC) in 2017
102	(http://www.meicmodel.org) with scaling factors to the year 2020 (Table S1, see text in Supplementary Information), which
103	were estimated based on the Three-Year Action Plan (2018-2020) issued by the government and the changes in the multi-
104	pollutant emissions of different sectors in recent years (Zheng et al., 2018). The emission adjustments during the lockdown
105	period are based on recent publications (see text in Supplementary Information). Emissions from the other countries were
106	derived from the MIX emission inventory (Li et al., 2017). International shipping emissions were taken from the Hemispheric
107	Transport Atmospheric Pollution (HTAP) emission version 2.2 dataset for 2010(Janssens-Maenhout et al., 2015). Biogenic
108	emissions were calculated by the Model of Emissions of Gas and Aerosols from Nature (MEGAN) version 2.1(Guenther et al.,
109	2012) with meteorological inputs from the WRF model.
110	Two experiments were conducted to investigate the impacts of meteorological changes and emission reductions on O3 during
111	the CLD period. The first (baseline) used the same anthropogenic emissions for the pre-CLD and CLD periods, and the second
112	(Reduction case) used emission reductions of 70%, 40% and 30% for transportation, industry and power generation,
113	respectively, and a 10% increase of residential emission during the CLD period. These emission reductions for the whole
114	country were estimated according to the previous literature(Doumbia et al., 2020; Huang et al., 2020; Wang et al., 2020a).
115	Comparing these two model simulations, the O3 changes during the CLD period relative to the pre-CLD period for the
116	Reduction Case were considered to be entirely due to the meteorological changes and emission reductions. The impacts of the
117	meteorological changes (including the changes in chemical boundary conditions) were quantified by subtracting the O3 mixing
118	ratios of the pre-CLD period from those of the CLD period for the baseline experiment, while the impacts of emission reduction

- 119 were estimated by comparing the O₃ mixing ratio during the CLD period between the Reduction Case and the baseline
- 120 experiment. Furthermore, we individually reduced the emissions of nitrogen oxide (NO_x), VOCs, CO, PM (particulate matter,
- 121 including PM₁₀, PM_{2.5}, black carbon, and organic carbon), and SO₂ during the CLD period to elucidate the response of O₃ to
- each pollutant reduction.
- 123 The performance of the CMAQ model in simulating the O₃, NO₂, PM_{2.5}, SO₂, and CO concentrations for the Reduction Case
- 124 was evaluated (Fig. S1 and Table S2), showing reasonable agreements with the respective surface observations. Details of the
- 125 emission estimation and the model evaluation are presented in Supplementary Information.

126 **3 Results**

127 **3.1 Observed O3 changes in different parts of China**

128 Figures 2 and 3 present the changes in observed concentrations of O_3 and other pollutants during the CLD period compared 129 with pre-CLD. The concentrations of most pollutants (SO₂, CO, PM_{2.5}, PM₁₀) that partially or fully originated from the direct 130 emissions declined in China during the lockdown. NO₂, a precursor of O₃, decreased by about 50% across the entire continental 131 China, and by similar amounts in all regions (Fig. 3b). However, the O₃ mixing ratio exhibited varying changes in different 132 regions (Fig. 2a). In NC and CC, the daily average O_3 increased significantly, by 112% and 73%, respectively (Fig. 2b); in 133 contrast, it remained almost unchanged in SC. The O₃ changes also varied between daytime (8:00-20:00 LST) and nighttime 134 (20:00-8:00 LST). During daytime, the O₃ increase in most parts of China was smaller than the daily average (Fig. 2c), 92% 135 and 71% in the NC and CC regions (Fig. 2d). In the SC region, most stations displayed a decrease in O₃ during daytime, 136 leading to a regional average O₃ drop of 14%. During nighttime, the O₃ mixing ratio increased significantly across China (Fig. 137 2e), by 154%, 77%, and 18% in NC, CC, and SC, respectively (Fig. 2f). These results reveal the diverse response of O_3 during 138 the lockdown in different regions, especially for the daytime. The changes in the O_x (NO₂+O₃) concentration (Fig. 3a), which 139 takes into account the NO titration, also varied in different regions. The daytime average Ox levels increased by 4% in NC and 140 by 11% in CC, and decreased by 29% in SC. These results suggest that the NO titration effect was not the only cause of the O₃

141 increase in northern and central China, as O_x would have decreased with sharply reduced NO_x emissions.

142 **3.2** Contribution of meteorological changes and emission reductions to O₃

143	Ground-level O ₃ is influenced by both chemical reactions of O ₃ precursors and meteorology. In this study, we used the WRF-
144	CMAQ model to separate the impacts of meteorological changes and emission reductions on the changes in O ₃ across China
145	(Fig. S2), which reveals significant contributions of both meteoroglgy (over most of contrinental China) and emissions (mainly
146	in populated areas of eastern China). Fig. 4 shows the more detailed results for the NC, CC, and SC regions for both daytime
147	and nighttime and Fig. S3 presents the meteorological impact and emission impact in terms of percent change. The observed
148	O3 changes in these regions were reasonably captured by the simulations. For the daytime average, the O3 increase in NC was
149	attributed to the comparable contributions from both meteorological changes (58%) and emission reductions (42%) (Fig. 4a).
150	In CC (Fig. 4b), the meteorological change (98%) was the primary cause of the O ₃ increase, whereas the contribution of
151	emission reduction was much lower (2%). In SC (Fig. 4c), the meteorological changes (73%) and emission reductions (27%)
152	both contributed to the O3 decrease. During nighttime, the emission reduction increased O3 in all three regions (including SC),
153	and its impact was stronger; the effect of meteorological changes weakened at night (Fig. S3).

154 **3.3 Impacts of meteorological changes on O**₃

155 The impacts of meteorological changes on O₃ for the NC, CC, and SC regions can be explained by the changes in the weather pattern and specific meteorological factors. In winter, continental China is generally controlled by a cold high-pressure system 156 157 (Fig. 5). During our study period, the center of this high-pressure system was located in northern China, moving southward 158 from the pre-CLD to the CLD period, with weakening strength. The high-pressure system therefore became increasingly 159 dominant in southern China, and the strengthened southward winds brought colder air masses from the north (Fig. 6c), which 160 decreased the temperature locally (Fig. 6a). In contrast, in central and northern China, the winds shifted to a more northward 161 direction, transporting warmer air masses from the south (Fig. 6c), which increased the temperature (Fig. 6a). During daytime, 162 the decrease (increase) in temperature in the SC region (CC and NC regions) weakened (enhanced) the surface O₃ chemical

- 163 production. Biogenic emission is an important source of VOCs and thereby contributes to O₃ formation in China (Wu et al.,
- 164 2020). The temperature changes led to an increase (decrease) of biogenic emissions in the CC (SC) region (Fig. S4). Thus, the
- 165 temperature changes increased (decreased) O₃ in the CC (SC) region by influencing chemical reaction rates directly (Fu et al.,
- 166 2015; Steiner et al., 2010) and altering biogenic emissions indirectly (Im et al., 2011; Liu and Wang, 2020a).
- 167 The changes in the weather pattern also resulted in less clouds and precipitation in northern and central China, but more clouds 168 and precipitation in southern China (Fig. 6e and f). Clouds can reduce the amount of solar radiation reaching the surface and 169 thus the chemical production of O₃(Lelieveld and Crutzen, 1990), while precipitation can also reduce O₃ through the 170 scavenging of its precursors(Seinfeld and Pandis, 2006; Shan et al., 2008). The cloud and precipitation patterns therefore 171 contributed to O₃ increases in CC and NC and decreases in SC. Furthermore, in NC and CC, the significant increase in the 172 planetary boundary layer height during the lockdown (Fig. 6d) might promote the transport of O_3 from the upper air to the 173 surface, contributing to the O₃ increase in these regions (He et al., 2017; Sun et al., 2009). The increase (decrease) in specific 174 humidity in NC and CC (SC) might also have contributed to the decrease (increase) in O₃ mixing ratios in those regions (Li et 175 al., 2019c; Ma et al., 2019) (Fig. 6b). During nighttime, the changes in meteorological factors were similar to those in daytime 176 (Fig. S5) but exerted smaller impacts on O_3 changes due to the decreasing effects of temperature and cloud cover (negligible 177 biogenic emissions and solar radiation).
- 178 **3.4 Response of O₃ to emission reductions**

We further investigated the impact of multi-pollutant reductions on the O₃ changes. Because transportation and industrial activities were reduced significantly during the lockdown and they are the major sources of NO_x (>80%) and VOCs (>60%) (Fig. 7), the estimated reductions of NO_x and VOC emissions were more significant than those for CO, particulate matter (PM), and SO₂ (Fig. 8a, c, e). The NO_x emission reductions were 46%, 49%, and 55% in the NC, CC, and SC regions, respectively, while the respective reductions for VOC emissions were 32%, 24%, and 45%. The relationship between O₃ and the emissions of its precursors is non-linear. We used the ratio of production rates between H₂O₂ and HNO₃ (P_{H2O2/PHNO3}) (Gaubert et al., 2021; Tonnesen and Dennis, 2000) to identify the O₃ formation regime in China for the periods before and during the lockdown 186 (Fig. 9). $P_{H202}/P_{HN03} < 0.06$ is VOC-limited region; $P_{H202}/P_{HN03} \ge 0.2$ is NO_x-limited region, and $0.06 \le P_{H202}/P_{HN03} < 0.2$ is 187 the transition zone. For the pre-CLD period, during daytime, the VOC-limited (or NO_x-saturated) regions included North 188 China Plain and other urban areas, while NO_x-limited regions located in southern China and other rural areas (Fig. 9a). During 189 nighttime, most regions are VOC-limited (Fig. 9c).

190 The O_3 formation regime determines the response of O_3 to the NO_x reduction during the CLD period. During daytime, NO_x 191 reduction increased O₃ in NO_x-saturated regions, but decreased it in NO_x-limited regions (Fig. 10b). We also found that 192 although the daytime O_3 formation regime in most regions shifted from the VOC-limited regime to the NO_x-limited regime 193 after the emission reductions during the CLD period, the daytime O₃ formation in the North China Plain was still controlled 194 by the VOC level (Fig. 9b), which suggests that the NO_x level is still high in this region. During nighttime, the reduction of 195 NO_x emission contributed increased O₃ due to the NO titration effect in a large areas (Fig. 10c). The reduction of VOC emission 196 decreased O₃ across China (Fig. 10d-f). As an O₃ precursor, the reduction of CO emission also contributed to a small decrease 197 in the O₃ mixing ratio (Fig. 10g-i); in contrast, the PM and SO₂ emissions reductions increased O₃ (Fig. 10j-o) through the weakening of aerosol effects (Li et al., 2019a; Liu and Wang, 2020b), but their impacts were much smaller and were 198 199 insignificant (< 1 ppbv) due to the smaller reductions, compared with NO_x and VOCs.

200 The response of O_3 to the emission reductions in different regions depended on the levels of NO_x and VOC reductions. For the 201 daytime average, in the NO_x-saturated NC region, the O₃ increase by the NO_x reduction counteracted the O₃ decrease by the 202 VOC emission reduction, leading to the decrease in increased O₃ production rates (Fig. 11) and a substantial net O₃ increase 203 (Fig. 8b). In CC, the contributions of the NO_x and VOC reductions were comparable in magnitude, and their opposing impacts 204 resulted in only a slight change in O_3 (Fig. 8d). In the NO_x-limited SC region, the impact of the NO_x reduction on O_3 was 205 smaller than that of the reduction of VOCs, leading to the decrease in O_3 production rates (Fig. 11), and a net decrease in O_3 206 (Fig. 8f). During nighttime, the effect of the VOC reduction was weakened due to the lower rate of degradation of VOCs by 207 radicals compared with daytime, and the O₃ level increased in all three regions due to decreases in the NO titration effect (Fig. 208 11). The impacts of emission reductions on whole-day average O₃ were similar to those during daytime.

209 The above modeling results show that the contribution of NO_x reductions (by 46%–55%) to the rise of O_3 decreased from NC

210	to CC and to SC, reflecting the decreasing level of NO _x saturation from north to south. In contrast, the impact of the estimated
211	VOC reduction on O_3 increased from north to south, which can in part be attributed to the regional variation of VOC reductions.
212	In the SC region, transportation and industry are the predominant sources of VOCs (97%, compared with 85% and 60% in NC
213	and CC, respectively) (Fig. 7). During the CLD period, the reduction of VOC emission in SC (45%) was significant and
214	comparable with the NO _x reduction (55%). In contrast, the VOC reductions in the NC (32%) and CC (24%) regions were much
215	lower (Fig. 8a, c) and could not offset the impact of NO _x reduction on O ₃ . The residential sector (mainly household coal
216	burning) is an important source of VOC emission in the NC and CC regions, whereas its contribution is smaller in SC. The
217	residential emissions increased during the CLD period because many migrant workers came back for the Chinese New Year
218	holiday and were stranded there due to the lockdown (Wang et al., 2020a; Wang et al., 2020b).
219	4. Conclusion
220	The first country-wide lockdown during the COVID-19 outbreak in China drastically reduced transportation and industrial
221	activities, leading to sharp declines in air pollutant emissions from these sectors. Surface O ₃ in urban areas of China responded
222	differently in the northern (increase) and southern regions (decrease) compared to the three-week period before the lockdown,
223	which can be explained by changes in meteorology and differences in the O3 chemistry regimes and the magnitudes of
224	precursor reductions in these regions. The model simulated contributions of meteorology to daytime O ₃ changes were larger
225	or comparable to most regions. The extent of VOC reduction, which suppressed O ₃ formation, was insufficient to offset the
226	large NO titration effect during daytime in northern China, and that larger reductions of VOCs (e.g., from residential sectors)
227	would have been needed to reduce the O ₃ concentration in the northern and central China. The rising O ₃ concentration in
228	northern China during the COVID-19 lockdown and in recent winters should receive greater attention because O3 boosts the
229	atmospheric oxidative capacity and therefore production of secondary aerosols (Fu et al., 2020; Huang et al., 2020; Zhu et al.,
230	2020), which are important components of winter haze in northern China. Our findings in China are relevant to untangling the
231	underlying factors driving the O3 changes in other parts of the world during their COVID-19 lockdowns.

232 Data availability

- 233 The codes and data used in this study are available upon request from Yiming Liu (liuym88@mail.sysu.edu.cn) and Tao
- 234 Wang (cetwang@polyu.edu.hk).

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- 238 University for providing the emission inventory.

239 Author contributions

- 240 T.W. initiated the research. Y.M.L. and T.W. designed the research framework. C.G., and T.D. estimated the emission
- 241 changes. Y.M.L. performed model simulations and drew the figures. T.W. and Y.M.L. analyzed the results. T.W. and
- 242 Y.M.L wrote the paper with the contributions from all the authors.

243 **Competing interests**

244 The authors declare that they have no conflict of interest.

245 Materials & Correspondence

246 Correspondence and requests for materials should be addressed to T.W. or Y.M.L.

247 Additional information

248 Supplementary information is available for this paper.

249 **Reference**

- 250 Akimoto, H., Mori, Y., Sasaki, K., Nakanishi, H., Ohizumi, T., Itano, Y., 2015. Analysis of monitoring data of ground-level
- 251 ozone in Japan for long-term trend during 1990-2010: Causes of temporal and spatial variation. Atmos. Environ. 102,
- 252 **302-310**.
- 253 Atkinson, R., 2000. Atmospheric chemistry of VOCs and NOx. Atmos. Environ. 34, 2063-2101.
- Bauwens, M., Compernolle, S., Stavrakou, T., Müller, J.-F., van Gent, J., Eskes, H., Levelt, P.F., van der A, R., Veefkind, J.P.,
- 255 Vlietinck, J., Yu, H., Zehner, C., 2020. Impact of Coronavirus Outbreak on NO₂ Pollution Assessed Using TROPOMI and
- 256 OMI Observations. Geophys Res Lett. 47, e2020GL087978.
- Binkowski, F.S., Arunachalam, S., Adelman, Z., Pinto, J.P., 2007. Examining Photolysis Rates with a Prototype Online
 Photolysis Module in CMAQ. J. Appl. Meteorol. Climatol. 46, 1252-1256.
- 259 Carter, W.P.L., 2010. Development of the SAPRC-07 chemical mechanism. Atmos. Environ. 44, 5324-5335.
- 260 Chinazzi, M., Davis, J.T., Ajelli, M., Gioannini, C., Litvinova, M., Merler, S., Pastore y Piontti, A., Mu, K., Rossi, L., Sun, K.,
- Viboud, C., Xiong, X., Yu, H., Halloran, M.E., Longini, I.M., Vespignani, A., 2020. The effect of travel restrictions on the
 spread of the 2019 novel coronavirus (COVID-19) outbreak. Science. 368, 395-400.
- 263 Doumbia, T., Granier, C., Elguindi, N., Bouarar, I., Darras, S., Brasseur, G., Gaubert, B., Liu, Y.M., Shi, X.Q., Stavrakou, J.,
- Tilmes, S., Wang, T., 2020. Changes in global air pollutant emissions during the Covid-19 pandemic: a dataset for atmospheric chemistry modeling. submitted to Earth. Sys. Sci. Data.
- Fu, T.M., Zheng, Y.Q., Paulot, F., Mao, J.Q., Yantosca, R.M., 2015. Positive but variable sensitivity of August surface ozone
- to large-scale warming in the southeast United States. Nat Clim Change. 5, 454-458.
- 268 Fu, X., Wang, T., Gao, J., Wang, P., Liu, Y., Wang, S., Zhao, B., Xue, L., 2020. Persistent Heavy Winter Nitrate Pollution
- 269 Driven by Increased Photochemical Oxidants in Northern China. Environ Sci Technol. 54, 3881-3889.
- 270 Gaubert, B., Bouarar, I., Doumbia, T., Liu, Y., Stavrakou, T., Deroubaix, A., Darras, S., Elguindi, N., Granier, C., Lacey, F.,
- 271 Müller, J.-F., Shi, X., Tilmes, S., Wang, T., Brasseur, G.P., 2021. Global Changes in Secondary Atmospheric Pollutants
- During the 2020 COVID-19 Pandemic. Journal of Geophysical Research: Atmospheres. 126, e2020JD034213.
- 273 Guenther, A.B., Jiang, X., Heald, C.L., Sakulyanontvittaya, T., Duhl, T., Emmons, L.K., Wang, X., 2012. The Model of

- 274 Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for
- 275 modeling biogenic emissions. Geosci Model Dev. 5, 1471-1492.
- He, J., Gong, S., Yu, Y., Yu, L., Wu, L., Mao, H., Song, C., Zhao, S., Liu, H., Li, X., Li, R., 2017. Air pollution characteristics
- and their relation to meteorological conditions during 2014–2015 in major Chinese cities. Environ. Pollut. 223, 484-496.
- Huang, Q., Wang, T., Chen, P., Huang, X., Zhu, J., Zhuang, B., 2017. Impacts of emission reduction and meteorological
- conditions on air quality improvement during the 2014 Youth Olympic Games in Nanjing, China. Atmos. Chem. Phys.
 17, 13457-13471.
- 281 Huang, X., Ding, A., Gao, J., Zheng, B., Zhou, D., Qi, X., Tang, R., Wang, J., Ren, C., Nie, W., Chi, X., Xu, Z., Chen, L., Li,
- 282 Y., Che, F., Pang, N., Wang, H., Tong, D., Qin, W., Cheng, W., Liu, W., Fu, Q., Liu, B., Chai, F., Davis, S.J., Zhang, Q.,
- 283 He, K., 2020. Enhanced secondary pollution offset reduction of primary emissions during COVID-19 lockdown in China.
- 284 National Science Review.
- Hutzell, W.T., Luecken, D.J., Appel, K.W., Carter, W.P.L., 2012. Interpreting predictions from the SAPRC07 mechanism based
 on regional and continental simulations. Atmos. Environ. 46, 417-429.
- 287 Im, U., Markakis, K., Poupkou, A., Melas, D., Unal, A., Gerasopoulos, E., Daskalakis, N., Kindap, T., Kanakidou, M., 2011.
- The impact of temperature changes on summer time ozone and its precursors in the Eastern Mediterranean. Atmos Chem
 Phys. 11, 3847-3864.
- 290 Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G., Keating, T., Zhang, Q., Kurokawa,
- J., Wankmuller, R., van der Gon, H.D., Kuenen, J.J.P., Klimont, Z., Frost, G., Darras, S., Koffi, B., Li, M., 2015.
- 292 HTAP_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air
- 293 pollution. Atmos Chem Phys. 15, 11411-11432.
- Lelieveld, J., Crutzen, P.J., 1990. Influences of Cloud Photochemical Processes on Tropospheric Ozone. Nature. 343, 227-233.
- 295 Li, K., Jacob, D.J., Liao, H., Shen, L., Zhang, Q., Bates, K.H., 2019a. Anthropogenic drivers of 2013-2017 trends in summer
- surface ozone in China. P Natl Acad Sci USA. 116, 422-427.
- 297 Li, K., Jacob, D.J., Liao, H., Zhu, J., Shah, V., Shen, L., Bates, K.H., Zhang, Q., Zhai, S., 2019b. A two-pollutant strategy for

- improving ozone and particulate air quality in China. Nat Geosci. 12, 906-910.
- Li, M., Zhang, Q., Kurokawa, J., Woo, J.H., He, K.B., Lu, Z.F., Ohara, T., Song, Y., Streets, D.G., Carmichael, G.R., Cheng,
- 300 Y.F., Hong, C.P., Huo, H., Jiang, X.J., Kang, S.C., Liu, F., Su, H., Zheng, B., 2017. MIX: a mosaic Asian anthropogenic
- emission inventory under the international collaboration framework of the MICS-Asia and HTAP. Atmos Chem Phys. 17,
 935-963.
- Li, R., Pei, S., Chen, B., Song, Y., Zhang, T., Yang, W., Shaman, J., 2020. Substantial undocumented infection facilitates the
 rapid dissemination of novel coronavirus (SARS-CoV-2). Science. 368, 489-493.
- 305 Li, R., Wang, Z., Cui, L., Fu, H., Zhang, L., Kong, L., Chen, W., Chen, J., 2019c. Air pollution characteristics in China during
- 306 2015–2016: Spatiotemporal variations and key meteorological factors. Sci. Total Environ. 648, 902-915.
- 307 Lian, X., Huang, J., Huang, R., Liu, C., Wang, L., Zhang, T., 2020. Impact of city lockdown on the air quality of COVID-19-

308 hit of Wuhan city. Sci. Total Environ. 742, 140556.

- 309 Liu, F., Page, A., Strode, S.A., Yoshida, Y., Choi, S., Zheng, B., Lamsal, L.N., Li, C., Krotkov, N.A., Eskes, H., van der A, R.,
- 310 Veefkind, P., Levelt, P.F., Hauser, O.P., Joiner, J., 2020. Abrupt decline in tropospheric nitrogen dioxide over China after
- 311 the outbreak of COVID-19. Science Advances. 6, eabc2992.
- Liu, Y., Wang, T., 2020a. Worsening urban ozone pollution in China from 2013 to 2017 Part 1: The complex and varying
 roles of meteorology. Atmos. Chem. Phys. 20, 6305-6321.
- Liu, Y., Wang, T., 2020b. Worsening urban ozone pollution in China from 2013 to 2017 Part 2: The effects of emission
- changes and implications for multi-pollutant control. Atmos. Chem. Phys. 20, 6323-6337.
- 316 Lu, X., Hong, J.Y., Zhang, L., Cooper, O.R., Schultz, M.G., Xu, X.B., Wang, T., Gao, M., Zhao, Y.H., Zhang, Y.H., 2018.
- 317 Severe Surface Ozone Pollution in China: A Global Perspective. Environ Sci Tech Let. 5, 487-494.
- 318 Lu, X., Zhang, L., Chen, Y., Zhou, M., Zheng, B., Li, K., Liu, Y., Lin, J., Fu, T.M., Zhang, Q., 2019. Exploring 2016–2017
- surface ozone pollution over China: source contributions and meteorological influences. Atmos. Chem. Phys. 19, 83398361.
- 321 Ma, T., Duan, F.K., He, K.B., Qin, Y., Tong, D., Geng, G.N., Liu, X.Y., Li, H., Yang, S., Ye, S.Q., Xu, B.Y., Zhang, Q., Ma,

- 322 Y.L., 2019. Air pollution characteristics and their relationship with emissions and meteorology in the Yangtze River Delta
- 323 region during 2014-2016. J. Environ. Sci. 83, 8-20.
- Miyazaki, K., Bowman, K., Sekiya, T., Jiang, Z., Chen, X., Eskes, H., Ru, M., Zhang, Y., Shindell, D., 2020. Air Quality
 Response in China Linked to the 2019 Novel Coronavirus (COVID-19) Lockdown. Geophys Res Lett. 47,
 e2020GL089252.
- 327 Murphy, B.N., Woody, M.C., Jimenez, J.L., Carlton, A.M.G., Hayes, P.L., Liu, S., Ng, N.L., Russell, L.M., Setyan, A., Xu, L.,
- 328 Young, J., Zaveri, R.A., Zhang, Q., Pye, H.O.T., 2017. Semivolatile POA and parameterized total combustion SOA in
- 329 CMAQv5.2: impacts on source strength and partitioning. Atmos. Chem. Phys. 17, 11107-11133.
- 330 Pye, H.O.T., Murphy, B.N., Xu, L., Ng, N.L., Carlton, A.G., Guo, H., Weber, R., Vasilakos, P., Appel, K.W., Budisulistiorini,
- 331 S.H., Surratt, J.D., Nenes, A., Hu, W., Jimenez, J.L., Isaacman-VanWertz, G., Misztal, P.K., Goldstein, A.H., 2017. On
- the implications of aerosol liquid water and phase separation for organic aerosol mass. Atmos. Chem. Phys. 17, 343-369.
- 333 Roelofs, G.-J., Lelieveld, J., 1997. Model study of the influence of cross-tropopause O₃ transports on tropospheric O₃ levels.
- Tellus B: Chemical and Physical Meteorology. 49, 38-55.
- Seinfeld, J.H., Pandis, S.N., 2006. Atmospheric Chemistry and Physics-from Air Pollution to Climate Change. John Wiley &
 Sons, New Jersey.
- Shan, W., Yin, Y., Zhang, J., Ding, Y., 2008. Observational study of surface ozone at an urban site in East China. Atmos Res.
 89, 252-261.
- Sharma, S., Zhang, M., Anshika, Gao, J., Zhang, H., Kota, S.H., 2020. Effect of restricted emissions during COVID-19 on air
 quality in India. Sci. Total Environ. 728, 138878.
- 341 Shi, X., Brasseur, G.P., 2020. The Response in Air Quality to the Reduction of Chinese Economic Activities During the
- COVID-19 Outbreak. Geophys Res Lett. 47, e2020GL088070.
- 343 Sicard, P., De Marco, A., Agathokleous, E., Feng, Z., Xu, X., Paoletti, E., Rodriguez, J.J.D., Calatayud, V., 2020. Amplified
- ozone pollution in cities during the COVID-19 lockdown. Sci. Total Environ. 735, 139542.
- 345 Siciliano, B., Dantas, G., da Silva, C.M., Arbilla, G., 2020. Increased ozone levels during the COVID-19 lockdown: Analysis

- 346 for the city of Rio de Janeiro, Brazil. Sci. Total Environ. 737, 139765.
- 347 Song, C.B., Wu, L., Xie, Y.C., He, J.J., Chen, X., Wang, T., Lin, Y.C., Jin, T.S., Wang, A.X., Liu, Y., Dai, Q.L., Liu, B.S., Wang,
- 348 Y.N., Mao, H.J., 2017. Air pollution in China: Status and spatiotemporal variations. Environ. Pollut. 227, 334-347.
- 349 Stadtler, S., Simpson, D., Schröder, S., Taraborrelli, D., Bott, A., Schultz, M., 2018. Ozone impacts of gas-aerosol uptake in
- 350 global chemistry transport models. Atmos. Chem. Phys. 18, 3147-3171.
- 351 Steiner, A.L., Davis, A.J., Sillman, S., Owen, R.C., Michalak, A.M., Fiore, A.M., 2010. Observed suppression of ozone
- formation at extremely high temperatures due to chemical and biophysical feedbacks. Proceedings of the National
 Academy of Sciences. 107, 19685-19690.
- 354 Sun, Y., Wang, Y., Zhang, C., 2009. Vertical observations and analysis of PM_{2.5}, O₃, and NO_x at Beijing and Tianjin from towers
- during summer and Autumn 2006. Advances in Atmospheric Sciences. 27, 123.
- 356 Tonnesen, G.S., Dennis, R.L., 2000. Analysis of radical propagation efficiency to assess ozone sensitivity to hydrocarbons and
- 357 NO_x : 1. Local indicators of instantaneous odd oxygen production sensitivity. J. Geophys. Res.-Atmos. 105, 9213-9225.
- 358 Wang, P., Chen, K., Zhu, S., Wang, P., Zhang, H., 2020a. Severe air pollution events not avoided by reduced anthropogenic

activities during COVID-19 outbreak. Resources, Conservation and Recycling. 158, 104814.

- 360 Wang, S., Zhao, M., Xing, J., Wu, Y., Zhou, Y., Lei, Y., He, K., Fu, L., Hao, J., 2010. Quantifying the Air Pollutants Emission
- Reduction during the 2008 Olympic Games in Beijing. Environ Sci Technol. 44, 2490-2496.
- 362 Wang, T., Xue, L.K., Brimblecombe, P., Lam, Y.F., Li, L., Zhang, L., 2017a. Ozone pollution in China: A review of
- 363 concentrations, meteorological influences, chemical precursors, and effects. Sci. Total Environ. 575, 1582-1596.
- Wang, Y., Yuan, Y., Wang, Q., Liu, C., Zhi, Q., Cao, J., 2020b. Changes in air quality related to the control of coronavirus in
- 365 China: Implications for traffic and industrial emissions. Sci. Total Environ. 731, 139133.
- 366 Wang, Z., Li, Y., Chen, T., Li, L., Liu, B., Zhang, D., Sun, F., Wei, Q., Jiang, L., Pan, L., 2015. Changes in atmospheric
- 367 composition during the 2014 APEC conference in Beijing. 120, 12695-12707.
- 368 Wang, Z.S., Li, Y.T., Zhang, D.W., Chen, T., Wei, Q., Sun, T.H., Wang, B.Y., Pan, J.X., Cui, J.X., Pi, S., 2017b. Analysis on
- air quality in Beijing during the military parade period in 2015. China Environ. Sci. 37, 1628-1636.

- 370 WHO, 2020. Coronavirus Disease (COVID-2019) Situation Reports. World Health Organization
- 371 Wu, K., Kang, P., Tie, X., Gu, S., Zhang, X., Wen, X., Kong, L., Wang, S., Chen, Y., Pan, W., Wang, Z., 2019. Evolution and
- Assessment of the Atmospheric Composition in Hangzhou and its Surrounding Areas during the G20 Summit. Aerosol
 Air Qual Res. 9, 2757-2769.
- Wu, K., Yang, X., Chen, D., Gu, S., Lu, Y., Jiang, Q., Wang, K., Ou, Y., Qian, Y., Shao, P., Lu, S., 2020. Estimation of biogenic
- 375 VOC emissions and their corresponding impact on ozone and secondary organic aerosol formation in China. Atmos Res.
 376 231, 104656.
- Zhang, R., Zhang, Y., Lin, H., Feng, X., Fu, T.-M., Wang, Y., 2020. NO_x Emission Reduction and Recovery during COVID19 in East China. Atmosphere 11, 433.
- 379 Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng,
- 380 Y., He, K., Zhang, Q., 2018. Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions.
- 381 Atmos. Chem. Phys. 18, 14095-14111.
- 382 Zhu, S., Poetzscher, J., Shen, J., Wang, S., Wang, P., Zhang, H.J.a.p.a., 2020. The seesaw impacts between reduced emissions
- and enhanced AOC on O₃ during the COVID-19.



386 Figure 1: Location of 1643 environmental monitoring stations (red "+" symbols) operated by the Ministory of Ecology and Environmental

387 Protection of China. The blue boxes denote the regions of north China, central China, and south China designated for further analysis.



Figure 2: Observed changes in O₃ mixing ratios across mainland China before and during the COVID-19 lockdown period. (a, c, e) The spatial distribution of O₃ changes for all-day average, daytime average, and nighttime average during the CLD period compared with the pre-CLD period. The black boxes in (a) show the locations of north China (NC, 184 sites), central China (CC, 108 sites), and south China (SC, 77 sites). (b) The variations of all-day average O₃ mixing ratios during the study period for the NC, CC, and SC regions. (d) The same with (b) but for daytime average O₃. (f) The same with (b) but for nighttime average O₃.

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397 Figure 3: Percentage change of (a) observed daytime average O_x (NO₂+O₃), whole-day average (b) NO₂, (c) CO, (d) SO₂, (e) PM_{2.5}, and (f)

³⁹⁸ PM₁₀ concentrations during the CLD period relative to the pre-CLD period.



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Figure 4: Changes in O₃ mixing ratios during the COVID-19 lockdown period and contributions from meteorological changes and emission reductions for three typical regions. (a) Observed and simulated changes in O₃ mixing ratios and the contributions from meteorological changes and emission reductions during the CLD period compared with the pre-CLD period in north China (NC). The O₃ changes for the all-day average, daytime average, and nighttime average are presented. (b) The same with (a) but for central China (CC). (c) The same with (a) but for south China (SC). The locations of these three regions are shown in Fig. 1. Note that the error bars mark the standard deviations within the region.



- 410 Figure 5: Averaged sea-level pressure during the pre-CLD and CLD periods. Data are from the National Center for Environmental Prediction
- 411 (NCEP) FNL Operational Model Global Tropospheric Analyses dataset.
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Figure 6: Model simulated changes in daytime temperature at 2 m height, specific humidity at 2 m height, wind field at 10 m height, planetary

416 boundary layer (PBL) height, cloud fraction, and precipitation during CLD period relative to pre-CLD period. In panel (c), the shaded color

417 and vector represent the wind speed and wind direction, respectively.



420 Figure 7: Percentage contribution to NO_x, VOCs, CO, PM, and SO₂ emissions from industrial (IND), power plant (POW), residential (RES),

421 and transportation (TRA) sectors in (a) north China, (b) central China, and (c) south China. Emission data are from 2017 MEIC

422 (<u>http://meicmodel.org</u>) with estimated scaling factors from 2017 to 2020.



Figure 8: The estimated reductions of multi-pollutant emissions due to the COVID-19 lockdown and their impacts on the O_3 changes for three regions. (a, c, e) The estimated reductions of NO_x , VOC, CO, PM, and SO₂ emissions during the CLD period compared with the pre-CLD period for north China, central China, and south China. (**b**, **d**, **f**) The impacts of different pollutant emission reductions due to the lockdown on O_3 changes for the three regions. The O_3 changes for all-day average, daytime average, and nighttime average are presented.

429 The error bars are the standard deviations.



Figure 9: Ozone formation regime in the daytime and nighttime before and during the lockdown periods estimated by the ratio of the production rates of hydrogen peroxide to nitric acid (P_{H2O2}/P_{HNO3}). VOC-limited region: $P_{H2O2}/P_{HNO3} < 0.06$; NO_x-limited region: P_{H2O2}/P_{HNO3} ≥ 0.2 , Transition zone: $0.06 \leq P_{H2O2}/P_{HNO3} < 0.2$. The production rates of H_2O_2 and HNO_3 were calculated by the integrated reaction rate (IRR) diagnose tool in the CMAQ model.



438 Figure 10: Model simulated changes in O₃ mixing ratios for all-day average, daytime average, and nighttime average due to the reductions





442 Figure 11: O₃ chemical production rates before and after the anthropogenic emission reductions and the changes during the COVID-19

443 lockdown period. The chemical production rates were calculated by the process analysis method in the CMAQ model.