ABSTRACT: Despite the large reduction in anthropogenic activities due to the outbreak of COVID-19, air quality in China has witnessed little improvement and featured great regional disparities. Here, by combining observational data and simulations, this work aims to understand the diverse air quality response in two city clusters, Yangtze River Delta region (YRD) and Pearl River Delta region (PRD), China. Though there was a noticeable drop in primary pollutants in both the regions, differently, the maximum daily 8 h average ozone (O3) soared by 20.6−76.8% in YRD but decreased by 15.5−28.1% in PRD. In YRD, nitrogen oxide (NOx) reductions enhanced O3 accumulation and hence increased secondary aerosol formation. Such an increment in secondary organic and inorganic aerosols under stationary weather reached up to 36.4 and 10.2%, respectively, which was further intensified by regional transport. PRD was quite the opposite. The emission reductions benefited PRD air quality, while regional transport corresponded to an increase of 17.3 and 9.3% in secondary organic and inorganic aerosols, respectively. Apart from meteorology, the discrepancy in O3−VOCs−NOx relationships determined the different O3 responses, indicating that future emission control shall be regionally specific, instead of one-size-fits-all cut. Overall, the importance of regionally coordinated and balanced control strategy for multiple pollutants is highly emphasized.

1. INTRODUCTION
The outbreak of COVID (The 2019 novel coronavirus) has severely impacted human daily life, not only by causing mortality but also impeding commercial activities and economical production.1−6 China, the first epicenter of the COVID-19 pandemic, had enforced nationwide lockdown measures against the virus incidence and spread.7,8 A series of strict controlling measures were undertaken, for example, by issuing stay-at-home orders, shutting down non-essential factories, restricting public transportation, closing catering and entertainment industries, postponing the reopening of schools, and so forth. The side effect of such a lockdown is a noticeable drop of anthropogenic emissions, providing a unique window to explore the potential of emission control and the consequent air quality response. Such an unprecedented “experiment” is of great significance, especially in those countries that are suffering from poor air quality like China. Many studies have examined the impact of this abrupt COVID-19 shutdown on air quality. Compared with the period before the lockdown, tropospheric nitrogen oxide (NOx) emissions decreased by 30−60%, based on satellite data.9−11 Ambient concentrations of sulfur dioxide (SO2) and carbon monoxide (CO) dropped by 52.5 and 36.2%, respectively, in central China, according to Xu et al.12 Similarly, the levels of surface PM2.5 (particulate matter with diameter less than or equal to 2.5 μm) also declined by approximately 35%.13 Wang et al.14 attributed this decrease of PM2.5 in Beijing−Tianjin−Hebei to the suspension of transportation and industry. In contrast, O3 was found to increase in most areas in China.15−17 Similar findings were also found elsewhere in the world, that is in India,18 Iraq,19 and Austria.20 Regarding to the O3 increase in Northern China and Central China, Huang et al.7 revealed that the increment of O3 enhanced the atmospheric oxidizing capacity, and in turn provided a favorable condition for the formation of the secondary particulate matter. This finding was corroborated by Le et al.20 since haze events were still observed even though
and PM2.5 were routinely measured by Thermo Instruments. 

The major components of PM2.5 [organic carbon (OC), elementary carbon (EC), sulfate, nitrate, and ammonium] and meteorological parameters (surface winds, temperature, and solar radiation) were collected from January 1 to February 17, 2020. Detailed information on the measurements, that is, monitoring instruments, data coverage, and access method is summarized in Table S1. Briefly, the ambient concentrations of O₃, NO₂, and PM₂.₅ were routinely measured by Thermo Instruments (TEI 49i, 42i, and model 5030 SHARP). Water-soluble inorganic ions in PM₂.₅ were monitored by MARGA (Monitor for Aerosols and Gases in Ambient Air). OC and EC were detected by an OC/EC analyzer (RT-4). All the instruments are routinely calibrated for different durations. Meteorological data, including wind speed/direction and temperature, were obtained from China Meteorological Administration. Moreover, air quality monitoring network, founded by the Ministry of Ecology and Environment of China, was also used to provide spatial information (i.e., O₃, NO₂, and PM₂.₅). There were 225 sites in YRD, and 90 sites in PRD (Figure S1). The averaged results were regarded as the overall condition for a given region. In addition to in situ measurements, the fifth generation of the European Centre for Medium-Range Weather Forecasts atmospheric reanalysis (ERAS) data were used to provide meteorological parameters, including sea level pressure, 2 m temperature, 10 m wind, boundary lay height, UVB (ultraviolet radiation b), and precipitation at 0.25° × 0.25° grid (https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5, last access on January 2021).

In addition, satellite-derived data, including formaldehyde (HCHO) column densities, NO₂ column densities, and nighttime index were also adopted. Daily formaldehyde (HCHO) and NO₂ column densities were employed to distinguish O₃ formation sensitivity; both HCHO and NO₂ column densities were obtained from the Tropospheric Monitoring Instrument (TROPOMI) level-2 retrievals. TROPOMI is the satellite instrument on board the Copernicus Sentinel-5 Precursor (SSP) satellite with an overpass time of around 13:30 LST (local standard time) and a spatial resolution of 3.5 km latitude × 5.5 km longitude. The calibration, quality control, and processing algorithms were provided on the TROPOMI website (www.tropomi.eu/data-products, last access on January 21 2021). The nighttime light index data were obtained from the Version 4 DMSP-OSL (Defense Meteorological Satellite Program-Operational Line Scan System) by National Oceanic and Atmospheric Administration (NOAA)/National Geophysical Data Center (NGDC). The index ranged from 0 (background) to 63 (saturated), with a grid resolution of 1 km × 1 km. The anthropogenic lights were closely linked to human activities and can be straightforward to map urban areas. The grid cells with nighttime light index above 50 were regarded as urban.

the overall PM₂.₅ concentrations were decreased in northern China.

Most of the above-mentioned studies mainly focused on either one site or one region, and few highlighted the varied responses between different sites or regions. It is worth noting that the formation mechanism in a certain site or area may not represent the whole situation across the country. Exploring and comparing the diverse mechanisms in different regions help us to improve our knowledge and benefit the regional joint air pollution mitigation process. Here, focusing on the two largest city clusters in China, YRD (Yangtze River Delta region, in eastern central China) and PRD (Pearl River Delta region, in eastern southern China), this study conducts site-to-site and region-to-region comparisons by using comprehensive field measurements and model simulations. The individual role of meteorological condition and emission reduction due to the COVID lockdown is comprehensively investigated. Meanwhile, the relative importance of local emission control and regional transport is also analyzed to better understand the diverse air quality response to lockdown controls in YRD and PRD regions.

2. MATERIALS AND METHODS

2.1. Data Source. Air quality data from two in situ observation stations, one from the Nanjing University SORPES (Station for Observing Regional Processes of the Earth System) site in YRD and the other from Guangzhou Environmental Monitoring Center site (GEMC) in PRD, were collected for analysis (Figure 1a). SORPES is a regional background station, since it is located along the downwind of the North China Plain but upwind of downtown Nanjing (with a distance of ~20 km) whereas GEMC is a typical urban site located right at the center of PRD. A comprehensive set of data including continuous measurement of O₃, NO₂, PM₂.₅, major components of PM₂.₅ [organic carbon (OC), elementary carbon (EC), sulfate, nitrate, and ammonium] and meteorological parameters (surface winds, temperature, and solar radiation) was collected from January 1 to February 17, 2020. Detailed information on the measurements, that is, monitoring instruments, data coverage, and access method is summarized in Table S1. Briefly, the ambient concentrations of O₃, NO₂, and PM₂.₅ were routinely measured by Thermo Instruments (TEI 49i, 42i, and model 5030 SHARP). Water-soluble inorganic ions in PM₂.₅ were monitored by MARGA (Monitor for Aerosols and Gases in Ambient Air). OC and EC were detected by an OC/EC analyzer (RT-4). All the instruments are routinely calibrated for different durations. Meteorological data, including wind speed/direction and temperature, were obtained from China Meteorological Administration. Moreover, air quality monitoring network, founded by the Ministry of Ecology and Environment of China, was also used to provide spatial information (i.e., O₃, NO₂, and PM₂.₅). There were 225 sites in YRD, and 90 sites in PRD (Figure S1). The averaged results were regarded as the overall condition for a given region. In addition to in situ measurements, the fifth generation of the European Centre for Medium-Range Weather Forecasts atmospheric reanalysis (ERAS) data were used to provide meteorological parameters, including sea level pressure, 2 m temperature, 10 m wind, boundary lay height, UVB (ultraviolet radiation b), and precipitation at 0.25° × 0.25° grid (https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5, last access on January 2021).

In addition, satellite-derived data, including formaldehyde (HCHO) column densities, NO₂ column densities, and nighttime index were also adopted. Daily formaldehyde (HCHO) and NO₂ column densities were employed to distinguish O₃ formation sensitivity; both HCHO and NO₂ column densities were obtained from the Tropospheric Monitoring Instrument (TROPOMI) level-2 retrievals. TROPOMI is the satellite instrument on board the Copernicus Sentinel-5 Precursor (SSP) satellite with an overpass time of around 13:30 LST (local standard time) and a spatial resolution of 3.5 km latitude × 5.5 km longitude. The calibration, quality control, and processing algorithms were provided on the TROPOMI website (www.tropomi.eu/data-products, last access on January 21 2021). The nighttime light index data were obtained from the Version 4 DMSP-OSL (Defense Meteorological Satellite Program-Operational Line Scan System) by National Oceanic and Atmospheric Administration (NOAA)/National Geophysical Data Center (NGDC). The index ranged from 0 (background) to 63 (saturated), with a grid resolution of 1 km × 1 km. The anthropogenic lights were closely linked to human activities and can be straightforward to map urban areas. The grid cells with nighttime light index above 50 were regarded as urban.
and the rest were taken as suburbs (Figure S1). Similar methods were extensively adopted by existing studies. In this study, all the observational data and simulation results were divided into two periods to investigate the impact of COVID lockdown, namely, the period before the lockdown (defined as PRE, from January 1 to January 24) and the period during the lockdown (defined as LOCK, from January 26 to February 17). In fact, the lockdown period covered the Chinese Spring Festival (from January 25 to January 31), which also contributed to emission reduction during the LOCK to some extent. Data on the Lunar New Year’s Day (January 25) were deducted in order to avoid the influence of intensive fireworks emissions. Besides, all the data were processed after deducting rainy days, based on the ERA5-derived precipitations. In general, all the data were well-controlled, and previous studies have demonstrated the good performance in air quality analyses.

2.2. Lagrangian Dispersion Modeling. Backward Lagrangian particulate dispersion modeling (LPDM) was carried out in order to identify the potential source region for the air masses measured at the observation stations. The LPDM was conducted using hybrid single-particulate Lagrangian-integrated trajectory model (HYSLITTY) driven by the ARL format Global Data Assimilation System (GDAS) data. With a time resolution of 1 h, 3000 particulates were released at 100 m a.s.l (above sea level) over the site and traced backward for 48 h. The position of particulates was calculated both vertically and horizontally after the consideration of mean wind and a turbulence transport component. The footprint “retroplume”, namely, the spatial residence time of particulates, which reflects the distribution of the surface probability or the residence time of the simulated air mass, was used to diagnose the contribution from potential source regions. LPDM simulations help us to distinguish whether the observational data is dominantly influenced by local emissions or regional transport.

2.3. WRF-Chem Simulation. A coupled online model, Weather Research and Forecasting model with Chemistry (WRF-Chem, version 3.7), was used to quantify the relative impact of emission reductions and meteorology and also to investigate the responses of atmospheric oxidizing capacities. Table S2 summarizes the physical/chemical settings for WRF-Chem. Briefly, the model domain covered East China and its surrounding areas, centered at 35.0°N, 110.0°E with a grid resolution of 20×20 km. National Centers for Environmental Prediction (NCEP) global final analysis data (FNL) was used as the initial and lateral boundary conditions of meteorological variables with a 1°×1° spatial resolution. Four-dimensional data assimilation (FDDA) was used as the grid analysis to improve the meteorology simulation. The Noah land surface scheme, along with the MMS Monin–Obukhov scheme was chosen for the land-atmosphere exchange study. The planetary boundary layer (PBL) was reproduced by the Yonsei University PBL (YSU) scheme. Carbon Bond Mechanism Version Z (CBMZ) and Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) were used for gas-phase and aerosol chemical simulations, respectively. The simulations were conducted based on two sets of emission inventories, including an up-to-date emissions inventory, and a business-as-usual emission inventory. Detailed information of the emission scenario settings are provided in Section 1 of the Supporting Information. After a spin-up of 7 days, the simulation started from January 1, 2020 to February 17, 2020, whereas January 1–24 was before the lockdown (PRE) and January 26 to February 17 was during the lockdown (LOCK). Our previous work has validated the performance of the WRF-Chem model. Table S3 summarizes the statistical results between the simulated and the observed O3, NO2, and O3 in YRD and PRD. The good agreement between the observations and the simulations demonstrate that the WRF-Chem model simulation in this work is capable of well reproducing the spatial and temporal variations of the main air pollutants.

3. RESULTS AND DISCUSSION

3.1. Opposite Response of Secondary Pollution in Two Regions. Figure 1b shows temporal variations of NO2, PM2.5, and O3 during the PRE and the LOCK periods at

Figure 2. Averaged diurnal variations of meteorological parameters, that is, wind, temperature (TEMP) and UVB, and air quality data, that is, NO2, PM2.5, and O3, during the PRE and the LOCK in Guangzhou and Nanjing, respectively.
SOPRES and GEMC. Both sites showed a significant drop of NOx and PM2.5 in the LOCK period as compared to those in the PRE period. Observational data showed that NO2 and PM2.5 declined by 55.1 and 41.6% in SOPRES, and 64.7 and 33.8% in GEMC, respectively. However, the average maximum daily average 8 h O3 (MDA8h O3) rose by 35.6% in SOPRES, whereas it decreased by 21.3% in GEMC. Not confined to these two stations, such a phenomenon also held true regionally in YRD (MDA8h O3 increased by 20.6–76.8%) and PRD (MDA8h O3 decreased by 15.5–28.1%) according to the air quality monitoring network (Figure S2). These results are indicative of the diverse response of O3 among the two regions.

We further investigated the variations in meteorological conditions. In spatial, a cold high-pressure dominated northern/central China during the PRE, and the cold invasion extended to southern China affecting PRD during the LOCK (Figure S3). Generally, the temperature and boundary height in YRD increased during the LOCK as the cold front passed by, and the opposite was witnessed by PRD due to the cold invasion. The diurnal characteristics of typical meteorological parameters are presented in Figure 2. In Guangzhou, north winds prevailed during both PRE and LOCK periods (Figures 2 and S3), and the average wind speed in the LOCK was 2.5 m/s, higher than that of PRE (1.2 m/s). The temperature in the LOCK was much lower than that in PRE, while the diurnal pattern of UVB was comparable in both the periods, with the peak value of LOCK slightly lower than that of PRE. In Nanjing, different meteorological variations were found. Though north winds dominated during PRE and LOCK periods, the wind speed in the LOCK period was rather small, 0.6 m/s, when compared to 1.1 m/s in PRE, indicating a stagnant condition during the lockdown period. The temperature was increased (by 1.3 °C on an average) with the maxima reaching 3 °C. Similar to that in Guangzhou, UVB was comparable during the two periods in Nanjing. Previous studies indicated that meteorological conditions played important roles in the secondary aerosol and O3 formation.29,30 For example, relatively low wind speed, high temperature, high solar radiation, and low planetary boundary layer height (PBLH) were the favorable meteorological conditions for the formation of secondary pollutants. Our analysis implied that meteorological conditions were more conducive to photochemical formation during the LOCK than during the PRE in Nanjing, but were more unfavorable in Guangzhou. The diurnal cycle of NO2 and PM2.5 agreed with the short-term decline of anthropogenic activities, with the concentrations reduced by 19.2 ppb and 11.3 μg/m3 in Guangzhou and 25.7 ppb and 27.8 μg/m3 in Nanjing, respectively. The noticeable drop of NO2 and PM2.5 at both sites indicated the positive effect of emission reductions. However, different responses of O3 were found at the two sites. In Guangzhou, daytime O3 decreased, while nighttime O3 increased in the LOCK period. The O3 reduction in daytime, on the one hand, might be attributed to the reduction of precursors, as Huang et al., reported that NOx emissions were reduced by 50% and VOC emissions were reduced by 46% during the LOCK period in PRD. And on the other hand, the unfavorable meteorological conditions were likely to be another reason for the daytime O3 decrease. The increment of nighttime O3 was possibly due to the weakened effect of NOx titration since NOx emissions were significantly reduced.31,32 In Nanjing, the magnitude of the overall O3 concentration was increased throughout the day during the LOCK period. This increment was consistent with previous observations, as reducing NOx emissions in NOx-saturated regions would lead to O3 increment.15,16,33 It should be pointed out that, in spite of the response of O3 to its precursors, our analysis showed that meteorological conditions in Nanjing seemed to be more conducive for O3 formation during the LOCK period with lower wind speeds and higher temperatures than those of the PRE. Thus, the relative impact of meteorology and emissions need to be further quantified. In addition, the different responses of O3 in Guangzhou and Nanjing indicated again the diverse mechanisms in O3 formation at different regions, and hence, we need to be careful while implementing emission reduction strategies.

Since NOx emissions are mainly emitted by transportation, industrial production, and power plants, and are usually concentrated in downtown,34,35 the effects of reductions might be different between urban and suburban areas. Therefore, we examined the daytime (12:00–17:00) and nighttime (0:00–5:00) responses of O3 in urban and suburban areas within YRD and PRD (Figure 3a,b). In YRD, both urban and suburban areas witnessed O3 increase throughout the day. However, in PRD, daytime O3 decreased in both urban and suburban areas, while nighttime O3 increased (by 1.4 ppb) in the urban areas, but decreased (by 1.5 ppb) in the suburban areas. To illustrate the diverse responses in the daytime, we introduced the ratio of column HCHO/NO2 as an indicator to distinguish different O3 formation sensitivity regimes (Figure 3c). In this study, the HCHO/NO2 ratio below 1 was considered as NOx-saturated (VOC-limited) regime; the HCHO/NO2 ratio above 2 reflected the NOx-limited regime; and the HCHO/NO2 ratio between 1 and 2 indicated the mixed-limited regime.36,37 The diagnosis of O3 formation sensitivity regimes from satellite retrievals, and the WRF-Chem simulations agreed well with each other (Figure S4). It was found that the YRD-urban areas were NOx-saturated, while the YRD-suburbs were mixed-limited, and therefore, O3 would be enhanced with the reduction of NOx, due to the lack of hydrogen oxide (HOx) radicals.38 In contrast, the PRD-urban areas were within the mixed-limited regime, and the PRD-suburbs were character-
ized by the NOx-limited regime, which meant that the decrease of precursor emissions inhibited O₃ formation during daytime. With regards to the nighttime increase in PRD-urban areas, it was likely due to the weakened effect of NO titration (NO + O₃ → NO₂ + O₂), whereas the nighttime O₃ drop in PRD-suburbs was due to the dominant roles of emission reduction in NOx-limited regimes.

Figure 4 compares the major components of PM₂.₅ at both sites. In this study, we split OC to POC (primary organic carbons) and SOC (secondary organic carbons) based on an EC (elemental carbon)-tracer method, and their quantity were calculated using the following equations.

\[
P_{OC} = \frac{OC_{pri}}{EC_{pri}} \times EC + OC_{non-comb} \quad (1)
\]

\[
SOC = OC - POC \quad (2)
\]

where \(OC_{pri}\) and \(EC_{pri}\) are the primary OC and EC, respectively, acquired from the pairs of OC and EC with their ratios within the 10% lowest, and \(OC_{non-comb}\) represented the OC not affected by combustion activities. The corresponding values were calculated from the slope and intercept of the linear regression between \(OC_{pri}\) and \(EC_{pri}\) respectively. In Figure 4, the proportion of SNA (sulfate, nitrate, and ammonium) accounted for 78.7 and 86.3% during the PRE in Guangzhou and Nanjing, respectively, and the corresponding proportion reduced to 61.1 and 80.3% during the LOCK, respectively. Indeed, the mass concentrations of sulfate, nitrate, and ammonium were reduced by 59.8, 80.7, and 60.0% in Guangzhou and 30.1, 56.6, and 48.3% in Nanjing, respectively (Figure S2). As the major components of PM₂.₅, SNA decreased substantially, which could explain the reductions of PM₂.₅. For carbonaceous aerosols, although the changes in the percentage of POC and EC in PM₂.₅ were negligible in both Guangzhou and Nanjing, decrease (\(p < 0.05\)) in the mass concentrations were significant in both the cities (Figure S5). In contrast, the proportion of SOC increased from 10.1 to 25.3% in Guangzhou and from 5.0 to 12.1% in Nanjing. Like O₃, SOC are also secondary products and reflected the degree of atmospheric oxidizing capacity to some extent. In Nanjing, the rise of SOC agreed with the pattern of O₃, which might be attributed to the increase of atmospheric oxidizing capacity due to NOx reduction. Contrarily, in Guangzhou, the variation of SOC was opposite with that of O₃. Considering that the wind speed was higher during the LOCK (\(p < 0.05\)), regional transport might play an important role. Thus, it is very

![Figure 4](image)

**Figure 4.** Comparison of proportions of different PM₂.₅ components during the PRE and LOCK periods in Guangzhou and Nanjing. The outer and inner rings present the PRE and the LOCK periods, respectively.

![Figure 5](image)

**Figure 5.** (a) Relative contribution of meteorology (METE) to absolute changes in O₃ between the LOCK and the PRE periods. (b) Same as (a) but for emission reduction (EMISS); (c) dominant factor between meteorology and emission reduction. The label C is the averaged difference in O₃ concentrations between the LOCK and the PRE periods; M > E indicates meteorology dominated, and E > M refers to emission reduction dominated.
important to distinguish between the impacts of meteorology and emission reductions, which is further investigated in following sections.

3.2. Spatial Disparity Caused by Meteorology and Emission Reduction. Here, the relative impact of emission and meteorology was quantified by WRF-Chem simulation (Figure 5). The impact of meteorology was an integrated result of all meteorological parameters, including the temperature, wind, PBLH, solar radiation, and so on. The detailed method to distinguish the individual impact of meteorology and emission reductions is elaborated in Section 1 of Supporting Information. As illustrated in Figure 5, in YRD and central China, meteorology variation promoted O3 formation with the maxima reaching $\sim 10$ ppb, whereas its role in PRD was mostly negative (approximately -5 ppb). On the other hand, emission reduction due to the lockdown increased O3 concentrations in YRD and central China, while it led to O3 decrease in most parts of PRD (Figure 5b). This could be explained by the regional disparities of the O3−NOx−VOC regime as presented in Figures 3c and S4. The diagnosis from both model results and satellite observations showed that most areas of YRD and central China were NOx-saturated, where cutting NOx emissions would increase O3. In PRD, most rural areas were NOx-limited, and most developed city clusters were mix-limited, with only a few being VOC-limited. The discrepancies of O3 formation sensitivity between YRD and PRD highlighted the diverse chemical sensitivities in O3 formation. Overall, by using a simple weighting method (introduced in Section 1 of Supporting Information), we found that most central areas of YRD (i.e., Nanjing, Suzhou, and Shanghai) were more affected by emission reductions (E > M, Figure 5c), whereas the northern and the southern parts were more affected by meteorology (M > E). Both the impact of emission reduction and meteorology led to the increment of O3 in YRD (C > 0). Differently in PRD, both emission reduction and meteorology resulted in the decrease of O3 (C < 0), and the effects of emission reduction were more than those of meteorology in most areas of Guangdong (E > M), with the exception of a few areas in the northeast (M > E).

3.3. Importance of Regional-Specific and Coordinated Emission Control. In addition to O3, WRF-Chem simulated oxidants, that is, NO3 radical and gas-phase H2SO4, were also induced to characterize atmospheric oxidizing capacity. Figure 6a,b compares spatial changes of NO3 and H2SO4 between the PRE and the LOCK, respectively. Enhancements of both NO3 and H2SO4 were witnessed in most areas of YRD (except in several urban areas). The different responses of NO3 and H2SO4 implied different relationships between oxidation products and their precursors in YRD and PRD. Therefore, a further study was to explore the sensitivity to different NOx reduction rates (from 10 to 90%); the non-linear responses of O3, NO3, and H2SO4 to NOx emissions are revealed in Figure 6c,d. In YRD, a continuous increase in O3, NO3, and H2SO4 was found by cutting NOx emission before reaching the tipping point, namely, $\sim 60−70\%$ NOx reductions. In fact, NOx emissions were reduced by approximately 49% during the LOCK period. With regard to PRD, the tipping point was closer ($\sim 40\%$ NOx reductions). There was around 50% NOx emission reduction during the lockdown, according to the emission estimation, which could explain the reduction of the oxidants in PRD. Notably, even though the regional average response of H2SO4 in PRD was reduced, an increase of H2SO4 over some urban areas was still found. This was because these urban areas were still under VOC-limited (shown in...
Figure S4), and the sulfate increment was a result of the higher atmospheric oxidizing capacity due to NOx reduction.

Responses of O3 to its precursors, NOx and VOCs, in both YRD and PRD are presented in Figure 7. The O3 isopleth was derived from hundreds of simulations with various NOx and VOCs emissions. The NOx-saturated, mixed-limited, and NOx-limited regimes corresponded to the maximum 1 h O3 concentration for corresponding precursors and were separated by ridge lines.37 It is worth noting that the relatively high O3 levels were associated with the mixed-limited regimes in both YRD and PRD, and the mixed-limited area was characterized by relatively high O3 levels on the left (with less VOCs reductions) and relatively low O3 levels on the right (with more VOCs reductions). In YRD, the situation of the PRE was a typical NOx-saturated regime, and cutting NOx emissions would inevitably enter it into the high-O3 area, that is, the mixed-limited regime. Indeed, the situation during the LOCK was close to the mixed-limited areas with higher O3 mixing ratios. In PRD, the situation of the PRE was at the border between NOx-saturated regime and mixed-limited regime, and nearly 50% reductions in both NOx and VOCs emissions led the LOCK in the "right" area of the mixed-limited regime with lower O3 mixing ratios.

As aforementioned, the O3−VOCs−NOx sensitive regime is characterized by great regional disparities. The calculations of potential source region based on LPDM analysis in Figure S6 show that YRD and PRD were affected by both local and northerly air flows. To better understand the roles of local emission reduction and regional transport, we further analyze the air quality responses under locally- and regionally-dominant conditions. Specifically, for every hour, observational data recorded at the two stations were diagnosed as "local" when the 72 h retroplumes were within the administrative border of Guangdong province and the YRD region of Guangzhou and Nanjing (Figure 1a). It was identified as "regional" air masses when the 72 h retroplumes overstepped...
the border of the “local”. Secondary pollutants, that is, O₃, SOC, and SNA were recollected according to the classification of air flows. The spatial distribution of “local” and “regional” air masses is provided in Figure S6.

In Guangzhou, the concentrations of O₃, SOC, and SNA were higher in the PRE than those during the LOCK when the air flow was local, indicating that the significant reduction of anthropogenic emissions benefit local air quality in PRD (Figure 7). On the contrary, opposite results were found when PRD was affected by regional transport (mainly from central China). The concentrations of O₃, SOC, and SNA increased by 126, 17.3, and 9.3%, respectively, during the LOCK, since the upwind central China featured O₃ increase due to NOx reduction. Thus, the regional transport partly offset the impact of PRD emission reductions and contributed to the increase of the secondary pollution. In Nanjing, concentrations of O₃, SOC, and SNA increased by 94.5, 36.4, and 10.2%, respectively, under locally dominant conditions. What is worse, the increment became more noticeable (increased by 56.4, 87.2, and 15.1%, respectively) when YRD was a worse, the increment became more noticeable (increased by 56.4, 87.2, and 15.1%, respectively, under locally dominant conditions. What is worse, the increment became more noticeable (increased by 56.4, 87.2, and 15.1%, respectively) when YRD was affected by regional transport (mainly from the North China Plain). This finding highlighted that regional transport could intensify the secondary pollution in the areas of downwind O₃-rebounded regions, which raise the alarm for the abrupt emission cut in those NOx-saturated areas.

4. IMPLICATION

Air pollution have drawn a lot of public attention in China over the last few years. Due to great efforts devoted to emission control, the haze pollution has been alleviated with declining PM2.5 concentrations. However, O₃ kept rising in recent years. Facing the complex air pollution, characterized by O₃ and secondary PM2.5 pollution, a large number of studies have been carried out, and among which, some proposed emission reduction scenarios by studying the responses of air pollutants to the hypothetical amount of primary emission reduction, and then provide scientific controlling suggestion for policy makers. Uniquely, the COVID-19 lockdown provided a real-world experiment with a nationwide anthropogenic emissions reduction, for studying the impact of transportation-dominant emission control on air quality.

Our data shows that PM₂.₅ and NO₂ decreased in both YRD and PRD due to the emission reduction, while O₃ rose in YRD but dropped in PRD. By addressing the importance of meteorology, we found that the short-term cut of anthropogenic emissions benefited PRD air quality, while the regional transport contributed to the increase of secondary pollution, that is, O₃ and secondary PM₂.₅. In YRD, NOx emission reductions enhanced atmospheric oxidizing capacity and led to the rise in O₃, SOC, and SNA. Moreover, the regional transport from the north (mainly from the North China Plain) further worsened the secondary pollution. Our modeling results revealed that YRD was typically NOx-saturated, and PRD was closer to the mixed-limited. The disparities in the O₃ sensitivity regimes partly explained the different responses of O₃ to the lockdown in PRD and YRD during the COVID-19 pandemic. Given the fact that the current O₃−VOCs−NOx relationships in the urban areas were mostly NOx-saturated or close to mixed-limited regimes, reducing NOx emissions would inevitably bring the chemical state closer to a mixed-limited situation with relatively high O₃ mixing ratios. Our suggestion is to take VOC emission as a joint effort in addition to NOx control, and more particular emphasis on VOC emission reductions would result in less O₃ within the mixed-limited regime (Figure 7). Consequently, emission control strategies should be adopted according to local conditions rather than being defined uniformly for the entire country (or an entire region). Regionally coordinated and balanced control strategy for multiple pollutants are highly recommended in the future.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c08383.
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Notes

The authors declare no competing financial interest.

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