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An observation approach in evaluation of ozone production to precursor changes during the COVID-19 lockdown

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HIGHLIGHTS

\begin{itemize}
  \item Both VOCs and NO\textsubscript{X} significantly declined during the COVID lockdown in Dongguan China.
  \item The largest reduction was observed from solvent use among various VOCs sources.
  \item A new approach was proposed to evaluate the response of ozone production to precursor changes.
\end{itemize}

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ABSTRACT

The increase of surface ozone during the Corona Virus Disease 2019 (COVID-19) lockdown in China has aroused great concern. In this study, we combine 1.5 years of measurements for ozone, volatile organic compounds (VOCs), and nitrogen oxide (NO\textsubscript{X}) at four sites to investigate the effect of COVID-19 lockdown on surface ozone in Dongguan, an industrial city in southern China. We show that the average concentrations of NO\textsubscript{X} and VOCs decreased by 70\%-77\% and 54\%-68\% during the lockdown compared to pre-lockdown, respectively. Based on the source apportionment of VOCs, the contribution of industrial solvent use reduced significantly (86\%-94\%) during the lockdown, and climbed back slowly along with the re-opening of the industry after lockdown. A slight increase in mean ozone concentration (3\%-14\%) was observed during the lockdown. The rise of ozone was the combined effect of substantial increase at night (58\%-91\%) and small reduction in the daytime (1\%-17\%). These conflicting observations in ozone response between day and night to emission change call for a more detailed approach to diagnostic ozone production response with precursor changes, rather than directly comparing absolute concentrations. We propose that the ratio of daily Ox (i.e. ozone + NO\textsubscript{2}) enhancement to solar radiation can provide a diagnostic parameter for ozone production response during the lockdown period. Smaller ratio of daily Ox (ozone + NO\textsubscript{2}) enhancement to solar radiation during the lockdown were observed from the long-term measurements in Dongguan, suggesting significantly weakened photochemistry during the lockdown successfully reduces local ozone production. Our proposed approach can provide an evaluation of ozone production response to precursor changes from restrictions of social activities during COVID-19 epidemic and also other regional air quality abatement measures (e.g. public mega-events) around the globe.

1. Introduction

To directly suppress the widespread of Corona Virus Disease 2019 (COVID-19), the Chinese government started a lockdown in the city of Wuhan since 23\textsuperscript{rd} January 2020, followed by a nationwide lockdown in most cities of China (Wang et al., 2020). The strict lockdown regulations...
led to an almost complete shut-down of industrial operation and traffic flow, resulting in an emission reduction of the precursors of secondary pollutants (Ding et al., 2020; Lee et al., 2020; Pei et al., 2020; Venter et al., 2020). The nationwide lockdown due to COVID-19 provides a unique opportunity to analyze the response of air quality to emission reduction strategies.

Recently, numerous studies have been reported to evaluate the effect of emission reduction strategies on the improvement of air quality during the COVID-19 lockdown (Le et al., 2020; Liu et al., 2020; Shi and Brasseur, 2020; Sun et al., 2020; Zangari et al., 2020). For the primary air pollutants (NOX, CO, CO2, SO2, primary PM2.5), the obvious decline was found in most cities during the lockdown (Liu et al., 2020a, 2020b, 2020c; Yuan et al., 2020). Despite improved air quality has been seen in most cities of China during the lockdown (He et al., 2020; Liu et al., 2020), pollution episodes of particulate matter (PM) were not avoided in North China Plain region and Yangtze River Delta due to unfavorable meteorological conditions and enhanced secondary formation (Chang et al., 2020; Huang et al., 2020; Le et al., 2020). Besides PM, an increase in daily mean ozone concentration was also reported in China during the lockdown due to lower ozone titration by NO as the result of NOX emission reduction (Sicard et al., 2020; Yuan et al., 2020; Zhao et al., 2020).

NOX and volatile organic compounds (VOCs) play vital roles in the ozone formation process and ozone production can be either VOCs-limited or NOX-limited (Kleiman, 1994). According to ozone–NOX-VOC sensitivities, the governments of most cities made emission reduction strategies of NOX and VOCs to control ozone concentrations. In the VOC-limited region, NOX emission reduction can promote photochemical ozone formation due to the non-linear relationships between ozone and its precursors, whilst a substantial reduction in NOX emissions results in reduced NO titration that increases ozone concentration was observed during the lockdown (Kroll et al., 2020; Qin et al., 2004; Sicard et al., 2020; Tan et al., 2009). Several studies also pointed out that the change of ozone relative to days before COVID-19 lockdown have significantly stronger than the changes between weekday and weekend, a common change pattern of human activities used for evaluation in atmospheric chemistry community (Pei et al., 2020; Sicard et al., 2020). Therefore, understanding the response of ozone to the emission reduction of precursors is a critical component to interpret COVID-19 impact on air quality (Kroll et al., 2020). However, multi-site online measurements of VOC in one city during the lockdown period has not been provided in the literature, which would be helpful in analyzing the response of ozone production to precursor changes.

In this study, based on continuous measurements from January 2019 to April 2020, we analyzed the variations in ozone, VOCs, and NOX before, during, and after the COVID-19 lockdown in Dongguan, Guangdong province, which is an air pollution hot spot in Pearl River Delta (PRD) of China (Barletta et al., 2008; Wang et al., 2015). Factor analysis was applied to identify and quantify changes of source contributions to VOCs during different periods. Finally, the photochemical formation of ozone response to the emission reduction of precursors is discussed.

2. Materials and methods
2.1. General description on monitoring site and data

Dongguan is a highly industrialized city, locating in the core of the Pearl River Delta (PRD) region of China (Fig. S1). Here, we use measurement data from four ambient monitoring stations, namely Supersite, Wanjiang, Dalingshan, and Houjie in Dongguan. Both Supersite and Wanjiang site represent the pollution in the downtown area of the city, whereas Dalingshan and Houjie sites are located in the industrial and residential area.

Hourly concentrations of VOCs, NOX, ozone, and meteorological parameters were measured at local air monitoring stations from 1st January 2019 to 30th April 2020. VOCs were monitored by online gas chromatograph-mass spectrometer (GC-MS/8666, Chromatotec) instruments, providing hourly concentrations of 56 VOC species including acetylene, 29 alkanes, 10 alkenes, and 16 aromatics. Noted that only aromatics were measured at Houjie site during the COVID-19 lockdown (from 1st January to 25th February 2020), due to the lack of access and maintenance supply to the measurement site during this period. Concentrations of six criteria air pollutants (O3, NOX, CO, SO2, and PM2.5) and meteorological parameters (including solar radiation) were provided by the Bureau of Ecology and Environment of Dongguan, China. We divided the dataset into four different periods: (1) Jan–May 2019 (from 14th January to 12th May 2019); (2) before lockdown (from 1st January to 23rd January 2020); (3) during lockdown (from 24th January to 10th February 2020); (4) after lockdown (from 11th February to 30th April 2020).

Four GC-FID/MS systems were calibrated quarterly by photochemical assessment monitoring stations (PAMS) standard gases (either from Scott or Linde, USA). The calibration results for measured VOCs were linear with correlation coefficient ($r^2$) greater than 0.98. Besides, the single-point calibration experiments were also conducted every week by using PAMS gas standards at mixing ratio of 5 ppbv, obtaining signal variations lower than 20% for most species were. The detection limits for various VOC species varied from 0.02 to 0.35 ppbv.

2.2. General description on PMF model

Source apportionment of VOCs was performed using positive matrix factorization (PMF) method (Paatero, 1997; Paatero and Tapper, 1994). PMF has been widely used for source analysis of VOCs, and its principle and applications have been detailed in many papers (Bon et al., 2011; Gaimoz et al., 2011; McCarthy et al., 2013). In this study, EPA PMF 5.0 was used for PMF calculations. A total of 31 VOC compounds from 1st January to 30th April 2020 (Table S1) were used as input of PMF model, which account for more than 82% of the TVOCs (56 species). Here, for VOC species whose concentration was equal to or less than method detection limit (MDL), the uncertainty of this species was set to be 5/6 of the MDL (Eq. (1)). However, for species whose concentration was higher than MDL, the uncertainty was defined as Eq. (2) (Gary Norris, 2014). MDL is obtained based on the monthly calibration results of the instrument.

$$Unc = \frac{5}{6} \times MDL$$

$$Unc = \sqrt{(Error\ Fraction \times x_i)^2 + (0.5 \times MDL)^2}$$

Where Unc is the uncertainty of VOC species $i$; $x_i$ is the concentration of species $i$; Error Fraction is set to 0.2.

3. Results and discussion

3.1. Variations of VOCs and NOX

Emissions of primary pollutants significantly decreased during the annually 7-day Chinese Spring Festival in China (Wang et al., 2017a), while the low concentration period of pollutants could be prolonged in 2020 because of limited social activities during the COVID-19 lockdown. Following the method in previous studies (Liu et al., 2020a), we defined the first day of Chinese Lunar Year as the origin (day 0) due to the irregular dates of the Lunar Year in China, to directly reflect the change of air pollutants at different periods.

Different species concentrations are normalized to the mean of from ~28 to ~14 days relative to Lunar New Year, then daily variations in the 7-day moving averages of each species are shown. As shown in Fig. 1, a large reduction of NOX is observed two weeks before the Spring Festival and reached a minimum during the Spring Festival holidays in both
The NOx curs almost every year in other urban areas of China (Liu et al., 2020a). 2019 (02/04–02/10) and 2020 (01/24–01/30). A similar situation occurred in the Spring Festival in 2019. However, the recovery rate of NOx emission recovered gradually to a normal level two weeks after the Spring Festival in 2019. However, the recovery rate of NOx emissions in 2020 was significantly slower than that in 2019 roughly from the Spring Festival holiday to mid-April. During the COVID-19 lockdown, the average concentrations of NOx decreased to 5–7 ppbv at various sites, reduced by 70%–77% compared to concentrations before the lockdown period (Fig. S3).

The variation of total VOCs (TVOCs) shows a similar trend as NOx, consisting of a sharp reduction around the Spring Festival holiday and a slow recovery from mid-February to mid-April as shown in Fig. 1. We observed significantly lower concentrations (decreased by 54–68%) of TVOCs during the lockdown than before lockdown, as shown in Fig. S3. However, the variations of different VOC groups during the lockdown are not the same (Fig. 1). The concentration of aromatics during the lockdown decreased by 86%, whereas the reduction of alkanes and alkenes concentrations were only 56% and 41%, respectively. The different behaviors among VOC groups indicate that the change for the emissions of various VOCs sources was different. The fraction of aromatics in total VOCs during the lockdown dramatically decreased from 27% (before the lockdown) to about 8%, and gradually grew back after lockdown (up to 29%). Besides, the larger reduction of higher reactive species also led to a greater decrease of TVOCs reactivity (74%) than the decrease of TVOCs concentration (Fig. 1). This is also reflected by the lowest ratio between the reactivity of TVOCs and the concentration of TVOCs during the lockdown at all measurement sites (Fig. S4). It is interesting to note that the larger decrease of VOC reactivity than VOC concentration is analogous to the idea of reactivity-based emission reduction for control of ozone pollution (Luecken and Mebust, 2008; Russell et al., 1995).

Five sources were identified by comparing PMF factor profiles with reported source profiles as shown in Fig. 2, including vehicle exhaust, evaporation, industry, shoe industry, and furniture and plastic industry. A detailed discussion on the source profile in each factor are provided in SI. Here, the third, the fourth, and the fifth factor are all related to solvent use, and thus we combine the three factors to represent contribution from solvent use. As the result, three distinct emission sources will be discussed in this study, namely vehicle exhaust, solvent use, and evaporation. The contribution of the solvent-use source to TVOCs reduced significantly (86–94%) during the lockdown compared to the period before lockdown (Fig. 3 using Supersite as the example). After the lockdown, the contribution of the solvent use source to TVOCs gradually increased along with the re-operation of various industrial activities. It is interesting to observe that the VOC concentrations contributed by vehicular emissions did not change significantly (25–27%) during the lockdown period. This may be related to higher tendency to use private transportation (including private cars and taxi, both mainly gasoline in Dongguan) as people practice social distancing (Huang et al., 2020a).

The different reduction percentages for different VOC species during the lockdown can be generally explained by the change of the contribution from both solvent use and vehicle exhaust. As shown in Fig. 3, the species related to solvent use (e.g. toluene, ethylbenzene, xylene, C5~C8 branched alkanes, and cycloalkanes) had reduced largely during the lockdown. It indicates that the reduction magnitudes of VOC species during the lockdown mainly depend on the decrease of contribution from solvent use. Similar results are obtained for Wanjiang and Dalingshan sites (Fig. S6 and 7).

As mentioned above, the contribution of vehicle exhaust to TVOCs did not decrease significantly during the lockdown. However, the...
concentration of NO\textsubscript{X} declined substantially during the lockdown. As diesel vehicles emit more NO\textsubscript{X} than gasoline vehicles (Zhong et al., 2018), the change of diesel trucks during the lockdown may explain the variation of NO\textsubscript{X}. The operating rate of diesel trucks in both 2019 and 2020 (the fraction of diesel trucks that running over 20 km each day, https://report.amap.com) is also shown in Fig. 1. The similar variations of NO\textsubscript{X} level and diesel trucks operating rate suggests that emissions from diesel trucks were the main source for NO\textsubscript{X} in Dongguan. In addition to on-road trucks, the emissions from non-road diesel vehicles should also decrease significantly, as construction and other related activities were halted during the Lunar New Year and the lockdown period. The large contributions of NO\textsubscript{X} from diesel trucks agree with the results of previous emission inventory in the PRD of China (Zhong et al., 2018).

3.2. Variation of ozone and its response to precursor changes

Although both the concentrations of VOCs and NO\textsubscript{X} decreased significantly during the lockdown, the change of daily mean ozone concentration is not as obvious as its precursors. Comparing with the period before lockdown, the mean ozone concentration increased by 3%–14% during the lockdown (Fig. S3). However, this is not observed during the same period of 2019, though precursors were also declined in 2019. This is partial as the results of a strong ozone pollution episode before the Lunar New Year in 2019. The large variability of ozone concentrations is also observed after the lockdown. A similar phenomenon is also shown for the concentrations of O\textsubscript{X} (Fig. S8). These large variations of absolute concentrations of ozone and O\textsubscript{X} preclude a clear understanding for the response of ozone to precursor changes. Ozone

Fig. 2. Source profiles resolved from the PMF model from each source factor.

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levels can also be associated with pollution episodes when meteorology is favorable for ozone accumulation for several days (Parrish et al., 2011; Zoran et al., 2020). Daytime ozone is produced mainly from photochemical reactions and nighttime ozone is affected by transport and accumulation (Wang et al., 2017b). Therefore, we divided the ozone concentration data into the daytime (8:00–16:00) and nighttime (22:00–6:00) for further analysis. We found that nighttime ozone and daytime ozone showed the opposite trend, with 58–91% increase at night and 1–17% decrease in the daytime (Figs. S3 and S8). It is also reflected by the weaker diurnal profile of ozone during the lockdown (Fig. S9). The increase of ozone at night is found to be solely because of a smaller titration by NO during the lockdown period, as the concentration of OX remained at similar levels compared to other periods (Fig. S9).

Fig. 3. Abrupt decline in solvent use during the lockdown. (a-c) The proportion of the contribution of three sources to TVOCs before, during, and after the lockdown. (d) Time series of three sources contribution to TVOCs from 1st January to 30th April 2020. The shaded area indicates the period during the lockdown. (e) The line chart (Red and Blue) represents the mean percentage of non-vehicle source contribution to each VOC species during the periods before lockdown and during the lockdown. The black line chart represents the percentage of the reduction of each VOC species during the lockdown compared with the period before lockdown. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)
Previous discussions suggest that a better parameter other than absolute concentration should be used for evaluating ozone production (in terms of O$_X$) response to the change of precursors. Inspired by the diurnal profile analysis in Fig. S9, we introduce the daily enhancement of ozone (or O$_X$), i.e., $\Delta$O$_3$ or $\Delta$O$_X$, which are defined as the difference of the ozone (or O$_X$) concentration at 16:00 and 8:00 for each day (schematic diagram in Fig. S10). The daily enhancement largely reflects the change of ozone production by local photochemical processes, which are directly related to observed concentrations of ozone precursors at the measurement sites. The variations of $\Delta$O$_3$ and $\Delta$O$_X$ were also normalized to the mean of from –28 to –14 days relative to Lunar New Year as shown in Fig. 4. Though there is still considerable variability, the daily enhancement of ozone and O$_X$ during the lockdown were at relatively low levels (12–15 ppbv by average) for the whole period.

To evaluate the impacts of meteorological factors on O$_X$ enhancement, we explore the daily variations in the 7-day moving averages of meteorological factors at Supersite in Dongguan (Fig. S11). We observed decrease (18%) and increase (22%) of ambient temperature during the lockdown in 2020 and during the Spring Festival in 2019, respectively. However, enhancement of ozone and O$_X$ were both lowest during the two periods. As the result, we conclude that ambient temperature may not play a significant role in ozone production during the investigated periods. Actually, previous studies show that ambient temperature usually affect ozone mainly by changing VOCs emissions (LaFranchi et al., 2011; Pusede et al., 2015), but changes of VOCs emissions due to changed pattern of human activities in Dongguan should be much larger than temperature-induced changes. In addition, there was no significant change in wind speed and wind direction during the lockdown period in the Dongguan region (Fig. S12), which further suggested that the O$_X$ enhancement change during the lockdown was not attributed to the long-range transport. The mean values of different meteorological factors in different periods are also listed in Table S2.

We further show $\Delta$O$_X$ as a function of daily mean solar radiation ($\text{Rad}$) (Fig. 5), which is the average of the solar radiation between 8:00–16:00. Moderate correlation ($R = 0.33–0.63$) between $\Delta$O$_X$ and $\text{Rad}$ is obtained for different periods. Significantly lower slopes ($0.02 \pm 0.02$) are obtained during the lockdown period in 2020 and also the Lunar New Year in 2019 ($0.03 \pm 0.04$) compared with other periods ($0.12 \pm 0.01$). The smaller slope during the lockdown period in 2020
indicates that less ozone was produced at the same magnitude of solar radiation. We also plot the indicates that less ozone was produced at the same magnitude of solar radiation. Considering the changes of VOCs and NOx concentrations during the lockdown, we conclude that the reduction of emissions of VOCs and NOx during the lockdown period successfully decrease local ozone production. However, it should be noted that emission reduction during the lockdown period for VOCs and NOx may not be the most cost-effective in controlling ozone pollution, and the optimal emission reduction strategy for VOCs and NOx should be determined in the future.

Recently, several studies showed that the daily mean ozone concentrations increased during the lockdown in many cities (Lee et al., 2020; Sicard et al., 2020; Yuan et al., 2020). Our analysis in this study shows that daily mean ozone concentrations can be influenced by many meteorological conditions in chosen periods for investigation (Fig. S8). Here, we show that the diurnal profiles of ozone and O$_3$ along with correlation analysis between $\Delta$O$_3$ and $\Delta$Rad can unfold the response of ozone production to the reduction of precursors. As shown in this study and previous studies (Gong and Liao, 2019; Guo et al., 2016; Wang and Liao, 2020), ozone concentrations can be influenced by numerous factors, including precursor changes and meteorological conditions (e.g. accumulation and transport of ozone during episodes). Our approach deliberately removed influences of varying solar radiation from the analysis, the most important meteorological factors affecting ozone production. To fully disentangle all of the factors influencing ozone concentrations, a chemical transport model by exploring various modeling scenarios might provide the solution, given that emissions of VOCs and NOx, various meteorological factors can be well characterized in the model.

4. Conclusions

In this study, we analyze the response of surface ozone production to the emission reduction of NOx and VOCs during the COVID-19 lockdown in Dongguan, China. Our results show that the mean concentrations of NOx and VOCs during the lockdown decreased by 70–77% and 54–68%, respectively, compared to the period before lockdown. Based on the source apportionment results of PMF, the contribution from the solvent use source to VOCs declined significantly during the lockdown (86–95%). In contrast, the reduction of NOx was driven by the decrease in emissions from diesel trucks. Although daytime ozone decreased during the lockdown by 1%–17%, the significantly elevated nighttime ozone (58%–91%) led to an overall increase of the mean ozone concentrations (3%–14%). We attribute the nighttime increase of ozone concentration to the weakened titration effect from NO. These results indicate comparison of absolute ozone concentration in different periods fails to diagnostic the ozone response to precursor changes. We propose to use the slope between daily enhancement of O$_3$ ($\Delta$O$_3$) and solar radiation ($\Delta$Rad) to understand the change of local ozone production. The results show that the effect of photochemistry reached the weakest during the lockdown. It is implied that the ratio of $\Delta$O$_3$ to $\Delta$Rad can be used to evaluate the ozone production response on the improvement of air quality in the future, for both during the COVID-19 lockdown periods and other air quality mitigation measures (such as public mega-events including Olympic Games). The results of this study also support that the emission reduction of NOx and VOCs is a valid strategy to control the photochemistry effect in China.

Notes

The authors declare no competing financial interest.

CRediT authorship contribution statement

Jipeng Qi: Investigation, Formal analysis, Writing – original draft. Ziwei Mo: Investigation, Writing – review & editing. Bin Yuan: Conceptualization, Supervision, Investigation, Writing – review & editing. Shan Huang: Investigation, Writing – review & editing. Yibo Huangfu: Software, Writing – review & editing. Zelong Wang: Software. Xiaobing Li: Writing – review & editing. Suxia Yang: Investigation. Wenjie Wang: Writing – review & editing. Yiming Zhao: Data curation. Xuemei Wang: Resources, Supervision. Weiwen Wang: Project administration. Kexuan Liu: Resources. Min Shao: Resources, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/atomenv.2021.118618.

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