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Research article

Worsening ozone air pollution with reduced NO_x and VOCs in the Pearl River Delta region in autumn 2019: Implications for national control policy in China

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ABSTRACT

Ozone (O_3) pollution has emerged as a major air quality issue in China. Here we emphasize the great challenges in controlling O_3 pollution by analyzing the recent experience of the Pearl River Delta (PRD) in southern China in reducing the autumn O_3 peaks. Despite significant reductions in the concentration of O_3 precursors, i.e., nitrogen oxides (NO_x) and volatile organic compounds (VOCs), regional O_3 pollution in the PRD was largely worse in autumn 2019 than in autumn 2018. We found that the supra-regional and regional background concentrations of O_3 increased significantly in the PRD in autumn 2019 due to increased concentrations of O_3 in the vast surrounding areas. We also observed slight increases in the concentrations of PRD-regionally and Guangzhou-locally produced O_3. A chemical box-model analysis confirmed a slight increase in the in-situ production of O_3 and revealed that increased biogenic VOCs (BVOCs) and decreased NO_x levels negated the effect of significant decrease in the anthropogenic VOCs. Taken together, these aspects exacerbated O_3 pollution in the PRD region in autumn 2019 relative to autumn 2018. The findings from this study highlight the strong interactions of O_3 pollution over multiple regions and the need for collaborative inter-regional efforts to control O_3 pollution. The experience of PRD also underlines the key role of BVOCs and the importance of science-based strategies to decrease VOCs and NO_x.

1. Introduction

Ground-level ozone (O_3) air pollution has attracted great public concern due to its adverse effects on the economy, human health and plant growth (Ahmad et al., 2013; Li et al., 2018; Lippmann, 1991; Selin et al., 2009), and has become a widespread environmental problem in China in recent decades (Xue et al., 2014a,b). The implementation of the "Air Pollution Prevention and Control Action Plan" in the 2013–2017 period by the China State Council led to significant decreases in the concentrations of nitrogen oxides (NO_x, a major precursor of O_3) and other routinely monitored pollutants, such as sulfur dioxide (SO_2), carbon monoxide (CO) and particulate matters (including $PM_{2.5}$ and PM_{10}) (He et al., 2017; Li et al., 2019b; Zeng et al., 2019). However, the concentration of O_3 is an exception, as levels of this pollutant have increased over many areas of China (Gaudel et al., 2018; Lu et al., 2020; Sun et al., 2016; Wang et al., 2017). As a result, O_3 pollution has been targeted as one of core challenges in air quality management in China. In particular, the management of emissions of volatile organic compounds (VOCs), another important class of O_3 precursors, will be attached more importance nationally in the near future.

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The Pearl River Delta (PRD) region is one of the most developed regions in China, and has suffered from severe air pollution due to its rapid urbanization and industrialization. However, the PRD has controlled air pollution better than other major air pollution prevention regions in China. For example, the PRD has implemented stringent $PM_{2.5}$-control measures and, in 2015, became first of three air pollution hot spots (i.e., the PRD, the Beijing-Tianjin-Hebei region, and the Yangtze River Delta region) to meet the $PM_{2.5}$ Class II Chinese National Ambient Air Quality Standard (i.e., annual average of 35 $\mu g/m^3$), and has met this standard for the past 5 years. Despite these achievements, O_3 pollution has worsened in the PRD region, and O_3 has replaced $PM_{2.5}$ as the top air pollutant in the warm seasons (April–September) (China National Environmental Monitoring Station, 2017).

There are two challenges to mitigating O_3 pollution in the PRD and many other regions (Atkinson, 2000; Li et al., 2012; Sillman, 1999). The first challenge involves developing cost-effective measures to control NO_x and VOC emissions, which is further complicated by the highly non-linear relationship between O_3 and its precursors (NO_x and VOCs) (Tang et al., 2012). The second challenge involves understanding the dynamic transport of tropospheric O_3 pollution, as O_3 has a relatively long lifetime and can be transported to the areas adjacent from where it is formed. For O_3 pollution control, the understanding of dynamic transport over different scales and scientific control of different precursors are the primary focus.

To alleviate the severe O_3 pollution in the PRD, the Guangdong provincial government in 2017 established a series of regulations to strengthen control of VOC emissions (referred to as the "Autumn O_3 Peak Reduction Campaigns"). This series represents an extension of regulations to control VOC and NO_x emissions that proposed reductions in the total anthropogenic emissions of VOCs and NO_x by 18% and 3% from 2015 to 2020, respectively. During these campaigns, the government refined VOC emission management to target key industries and major anthropogenic sources. The entire PRD was involved, and Guangzhou, Shenzhen, Foshan, and Dongguan were listed as the demonstration cities for the campaign. The measurements confirmed significant decreases in anthropogenic NO_x and VOC emissions during the "Autumn O_3 Peak Reduction Campaigns" in 2019 (see Section 3.1). However, O_3 pollution remains a problem in the PRD and the vast surrounding areas over southern China (Ozone Pollution Control Professional Committee, 2020). The experience of "Autumn O_3 Peak Reduction Campaigns" in autumn 2019 in the PRD region presents a unique opportunity to examine how O_3 pollution can worsen against the backdrops of significant reductions of NO_x and VOCs in the PRD but rapid increase of O_3 concentrations in the surrounding areas.

In this study, the PRD and Guangzhou (the demonstration city and regional center in the PRD) were taken as examples to determine the reasons for increased O_3 pollution despite decreases in the concentrations of NO_x and VOCs. The comprehensive measurement data during 2018–2019 were extracted for detailed analysis. First, we confirm that the emissions of O_3 precursors (mainly NO_x and anthropogenic VOCs (AVOCs)) were better controlled but O_3 pollution still aggravated in Guangzhou and the PRD in autumn 2019 compared to autumn 2018. Then, we reveal that increases in the regional background concentrations of O_3 and local O_3 production led to worse O_3 pollution in Guangzhou, and conduct a series of sensitivity model tests to further quantify the effects of changes in O_3 precursors on local O_3 production. Finally, we determine the optimum AVOCs/NO_x reduction ratios to effectively alleviate local O_3 formation and give recommendations for the future control of O_3 pollution.

2. Materials and methods

2.1. Measurement data

Two observational data sets were used for the analyses in this study. Briefly, ground-level O_3 concentration data were obtained from 1605 air-quality monitoring stations (AQMSs), which cover most areas of China, and the collected data reflect the inter-annual variations in national O_3 pollution (see Fig. 1(a)). The measurement data in autumn 2018 and autumn 2019 was extracted to evaluate the effectiveness of the "Autumn O_3 Peak Reduction Campaigns in 2019", which aimed to alleviate the severe O_3 pollution in autumn season, when O_3 exhibited maximum concentrations due to the polluted continental air masses coupled with dry and sunny weather conditions after the retreat of the summer marine air masses (Wang et al., 2009; Zheng et al., 2010). The O_3 inter-annual variation analysis was based on hourly and maximum daily 8-h average (MDA8) O_3 concentrations, and the O_3 non-attainment day was defined as a day when the MDA8 O_3 concentration exceeded the Chinese National Ambient Air Quality Standard, i.e., 75 ppbv (Class II). To eliminate the effect of nitric oxide (NO) titration, we also calculated the total oxidant ($O_x = O_3 + NO_2$) and MDA8 O_x concentrations, and subjected them to similar analyses. In addition, the air-quality data obtained from 56 AQMSs in the PRD and 11 AQMSs in Guangzhou were analyzed to reveal the inter-annual variations over the study areas and to determine the regional background concentrations and locally produced concentrations of O_3.

Another data set was obtained from biennial comprehensive observations conducted in urban Guangzhou. Guangzhou is a megacity, the capital of Guangdong province, and the regional center of PRD (see Fig. 1 for the location of Guangzhou). Guangzhou plays a leading role in air pollution control in the PRD region, and was a demonstration city during the "Autumn O_3 Peak Reduction Campaigns". Therefore, changes in the concentrations of NO_x and AVOCs over urban Guangzhou could well reflect the effects of NO_x and AVOC control over the PRD. Field observations were conducted at an urban site (see Fig. 1(b) for its location) from January 1, 2018 to December 31, 2019, and data from autumn (September–November) 2018 and 2019 were extracted for the analysis. The comprehensive dataset comprises the concentrations of trace gases (O_3, SO_2, NO, NO_2, CO) and VOCs (57 compounds) and various meteorological parameters (temperature, relative humidity, and pressure). These data were subjected to a detailed chemical box modeling analysis to quantify the effects of changes in O_3 precursors on photochemical O_3 formation. Detailed information on the sampling site, measurement techniques, and quality assurance and control procedures are documented in the SI.

2.2. Estimation of regional background concentrations and local production of O_3

The Texas Commission on Environmental Quality (TCEQ) method was used to evaluate the contributions of background concentrations of O_3 over different scales to the observed levels of O_3 in Guangzhou and the PRD (Berlin et al., 2013; Kemball-Cook et al., 2009; Langford et al., 2009; Nielsen-Gammon et al., 2005). We modified the TCEQ method to contain three nested domains (Guangzhou, PRD, and outside the PRD), based on the following logic. The afore-mentioned 11 AQMSs in Guangzhou (56 AQMSs in the PRD) provide good coverage of Guangzhou (the PRD). At the local scale, as wind directions change, we can assume that at least one of the 11 AQMSs is always located upwind of Guangzhou. Therefore, the data collected at that site (or sites) are reasonably representative of the regional background concentrations of O_3 that are transported into Guangzhou, while other sites are additionally affected by their local photochemistry to varying extents. At a larger spatial (i.e., regional) scale, the 56 AQMSs within the PRD region can be used to estimate the supra-regional background concentrations of O_3 that are transported into the PRD and the concentrations of O_3 that are produced over the PRD.

Hence, the contributions of supra-regional (outside PRD) background, regionally (PRD) produced, and locally (Guangzhou) produced concentrations of O_3 to the observed concentrations of O_3 in Guangzhou can be estimated on a daily basis using the following equations (E1–E3), and then averaged for the entire autumn seasons of 2018 and 2019:

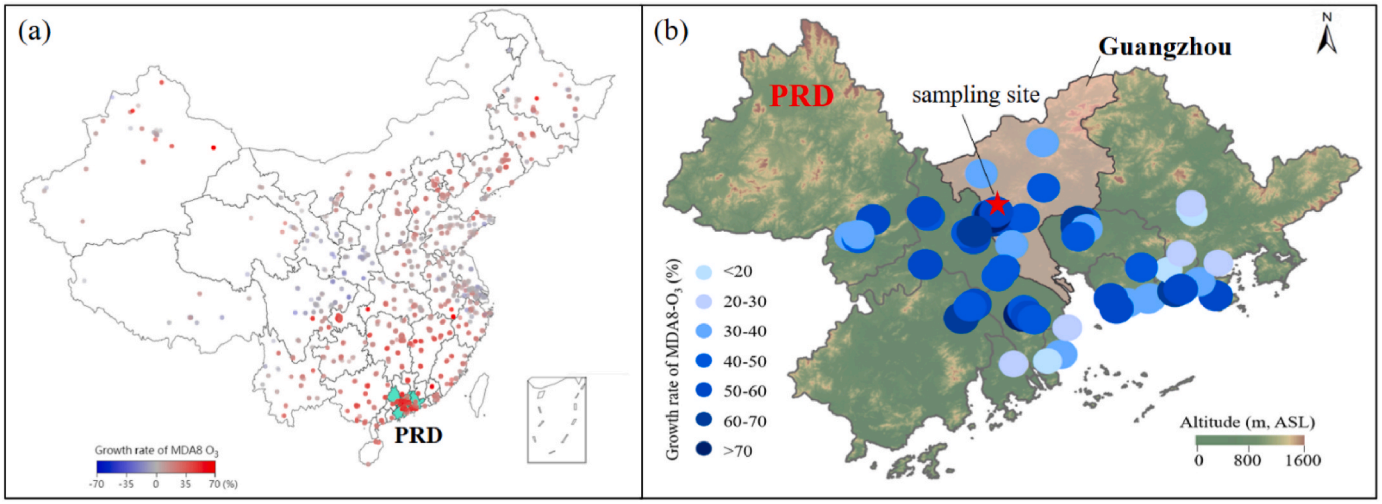

Fig. 1. The maps showing the inter-annual variations of autumn average maximum daily 8-h average ozone (MDA8 O_3) in (a) China and (b) the Pearl River Delta (PRD) region from 2018 to 2019. Also shown are the locations of (b) Guangzhou in the PRD and the sampling site in Guangzhou. The O_3 data were obtained from the Air-Quality Monitoring Stations from the public website of the China Ministry of Ecology and Environment.

$$C_{Supra-regional} = Min_{O_{X-PRD}} \quad \text{(E1)}$$

$$C_{Regional} = Min_{O_{X-Guangzhou}} - Min_{O_{X-PRD}} \quad \text{(E2)}$$

$$C_{Local} = Max_{O_{X-Guangzhou}} - Min_{O_{X-Guangzhou}} \quad \text{(E3)}$$

where $C_{Supra\text{-}regional}$, $C_{Regional}$, and C_{Local} represent the contributions of the supra-regional background, regionally produced, and locally produced concentrations of O_3, respectively. Min_{Ox} and Max_{Ox} represent the lowest and the highest MDA8 O_x concentrations recorded by the available AQMSs over a specific region (the PRD or Guangzhou), respectively.

Note that the local meteorology might lead to uncertainties in the TCEQ-estimated background O_3 concentrations (Berlin et al., 2013). To reduce the uncertainties, a least-squares linear regression analysis has been performed on TCEQ results, and the detailed information were documented in the SI.

2.3. Chemical box model

A chemical box model built on two different chemical mechanisms, namely the Master Chemical Mechanism (MCM v3.3.1; a near-explicit chemical mechanism) and the Regional Atmospheric Chemical Mechanism (RACM v2; a lumped chemical mechanism) (Goliff et al., 2013; Jenkin et al., 2003; Saunders et al., 2003), was adopted in the present study. Briefly, the MCM box model was used to quantify the net in-situ rate of O_x production and to evaluate the effects of decreases in O_3 precursor concentrations on O_3 formation, and the RACM2 box model was used to establish O_3 isopleth diagrams that describe the nonlinear responses of O_3 concentrations to decreases in O_3 precursors. Our previous studies showed that both mechanisms agreed well with O_3 simulations (Zong et al., 2018), and the RACM2 was adopted for developing O_3 isopleth mainly due to its more efficient computation. Physical processes such as solar radiation, dry deposition, and planetary boundary layer evolution are also considered in the chemical box model. A detailed description of the model setup is provided elsewhere (Xue et al., 2016; Zhang et al., 2019).

Within the MCM model, the net rate of O_x production was calculated as the difference between the rate of O_x production ($P(O_x)$) and rate of O_x loss ($L(O_x)$), which were quantified using Equations (S1)–(S2) in the SI. The observed O_3, CO, SO_2, NO, NO_2, and C_2–C_{12} non-methane hydrocarbon (NMHC) concentrations, relative humidity, temperature, and pressure data were averaged or interpolated into a 1-h time resolution to constrain the model. Observed nitrous acid (HONO) concentration data were unavailable and therefore estimated by linear regression with NO_2 (see details in the SI). Sensitivity tests showed that a 1.5-fold change in the concentration of HONO would exert a relatively minor effect on the net rate of O_x production, and would not change our major conclusions (see SI Fig. S2). The model was initiated at 00:00 local time (LT) on September 1, and the integration had a step of 1 h and duration of the entire autumn seasons (September–November) of 2018 and 2019. The model was pre-run for 3 days prior to each simulation to approach a steady state to stabilize the unconstrained species. A series of sensitivity tests were also conducted to evaluate the effects of changes in O_3 precursor concentrations on the net in-situ rate of O_x production.

To establish the O_3 isopleth diagrams, the RACM2 box model was constrained in the base run by the autumn-average diurnal profiles of SO_2, CO, NO, NO_2 and C_2–C_{12} NMHC concentrations, temperature, relative humidity, and pressure. Then, a series of emission-reduction simulations were conducted using X times (X = 0–1, with a bin of 0.05) the base concentrations of AVOCs and NO_x. To obtain a more reliable modeling result, the initial concentrations of oxygenated VOCs (OVOCs) and radicals (OH and HO_2) were prescribed according to the 2-day pre-run results. O_3 was initialized using the data observed at 06:00 h LT (the initial time of model integration), and its chemistry and concentrations were simulated using the constraints of other relevant species in the following integration. The model was initiated at 06:00 h LT, and the integration had a step of 1 h and duration of 24 h. The model-simulated maximum concentrations of O_3 under the base scenarios and 399 emission-reduction scenarios were extracted to describe the non-linear response of O_3 to decreases in the concentrations of O_3 precursors (similar to but slightly modified from the so-called EKMA plot).

3. Results and discussion

3.1. Variations in the autumn concentrations of O_3 and O_3 precursors between 2018 and 2019

We compared the concentrations of NO_x and VOCs in urban Guangzhou between autumn 2018 and autumn 2019 to evaluate the effectiveness of air pollution control over the PRD region (shown in Fig. 2 (a) & (b)). Compared to 2018, NO_x concentrations in 2019 were significantly lower (by 7%; $p < 0.01$) during the daytime (7:00–18:00 h LT) but slightly higher (by 4%; $p = 0.26$) during the nighttime (19:00–6:00 h LT). A similar inter-annual variation in the pattern of NO_2 concentrations was also revealed by data from 56 AQMSs over the PRD

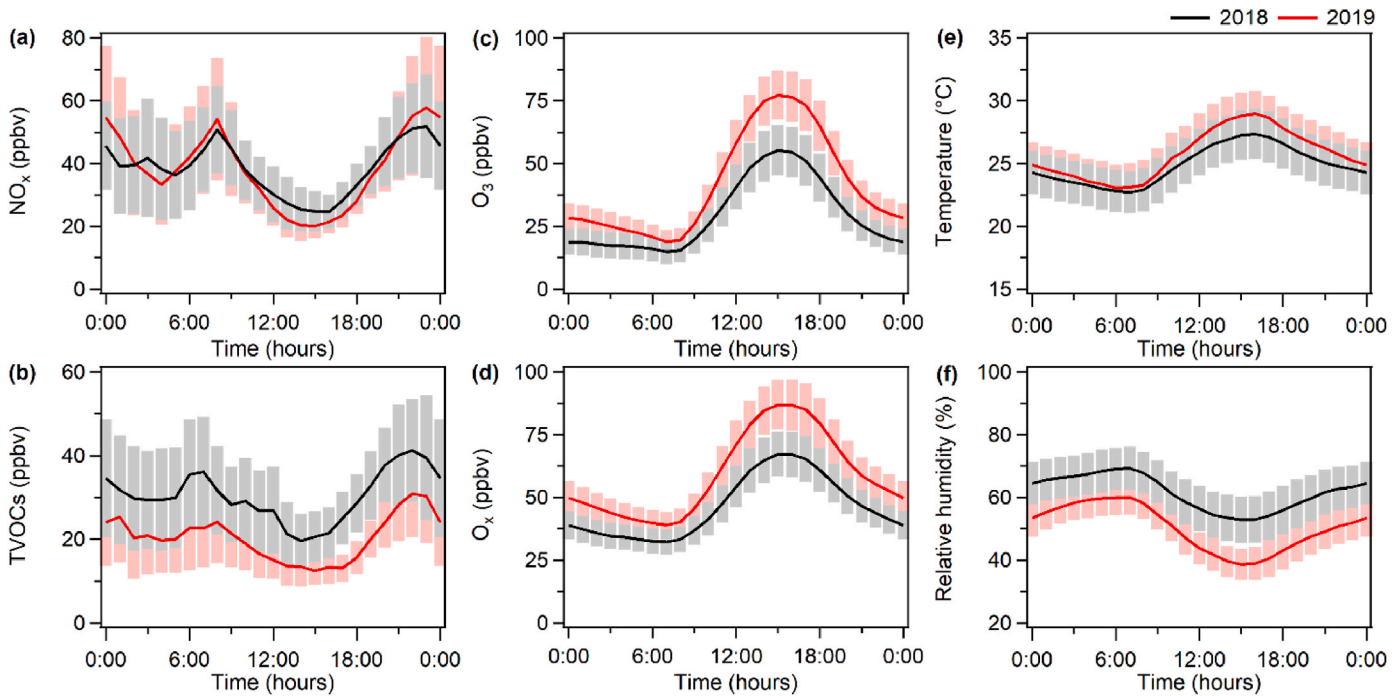

Fig. 2. Average diurnal variations in the concentrations of (a) nitrogen oxides (NO_x), (b) total volatile organic compounds (TVOCs), (c) ozone (O_3), and (d) O_x ($O_x = O_3 + NO_2$), and meteorological parameters ((e) temperature and (f) relative humidity) in the Pearl River Delta (PRD) in autumn 2018 and autumn 2019. The error bars indicate a half-standard deviation of the mean. The concentrations of O_3 and O_x were obtained from 56 air-quality monitoring stations in the PRD, the concentrations of NO_x and VOCs, and the meteorological parameters (temperature and relative humidity) were measured at an urban site in Guangzhou, the capital city of Guangdong Province and the center of the PRD region.

(see SI Fig. S4). In comparison, remarkable decreases in both the daytime and nighttime concentrations of AVOCs were observed in 2019, compared with 2018. Specifically, the AVOC concentrations and their OH reactivities and O_3 formation potentials (OFP) decreased by 28%, 10%, and 20%, respectively (see Table. S1). Note that the OFP was calculated based on the localized maximum incremental reactivity (MIR) scales for Guangzhou (Zhang et al., 2021). A one-way analysis of variance revealed that all of these changes were statistically significant. A further inspection of the sub-group variations in AVOC concentrations revealed that the concentrations of alkenes (isoprene was excluded), aromatics, alkanes, and alkynes decreased by 28%, 27%, 29%, and 20%, respectively. In comparison, the concentrations of biogenic VOCs (BVOCs) (i.e., isoprene) significantly increased by 33%, which should be attributed to the higher leaf temperature and stronger photosynthetically active radiation (indicated by higher ambient temperatures) in autumn 2019 (Sabillon and Cremades, 2001; Chang et al., 2009). Similarly, decreases in AVOCs and increases in BVOCs were also observed in Shenzhen (another demonstration city of the "Autumn O_3 Peak Reduction Campaigns") during 2018–2019. Overall, the inter-annual variations in the concentrations of primary pollutants (especially AVOCs) demonstrate the success of the emission-reduction programs implemented in Guangzhou (as a demonstration city of the "Autumn O_3 Peak Reduction Campaigns") and the PRD, and provide an example for future VOC emission control programs in other regions of China.

Despite these significant decreases in the concentrations of AVOCs and NO_x, the concentrations of O_3 significantly increased in autumn 2019 compared with 2018 over most areas of the PRD, reaching a maximum increase of 70% (see Fig. 1(b)). As shown in Fig. 2, the concentrations of O_3 in the PRD increased by an average of 41% ± 21% ($p < 0.01$) during the daytime and 44% ± 19% ($p < 0.01$) during the nighttime. In addition, the concentrations of O_x increased consistently and significantly during the daytime (by 29% ± 22%; $p < 0.01$) and the nighttime (by 26% ± 20%; $p < 0.01$). We also determined the inter-annual variations in the number of O_3 non-attainment (exceedance) days on which the MDA8 O_3 exceeded 75 ppbv (see SI Fig. S5). In autumn 2019, there were a significant number of O_3 non-attainment days (41%; 37 days), compared to only a few such days in autumn 2018 (9%; 8 days). The higher concentrations of O_3 (O_x) and more non-attainment days indicate worse O_3 pollution in the PRD in autumn 2019 than in autumn 2018. The mechanism by which these distinct inter-annual variations in O_3 precursors (especially AVOCs) affected O_3 pollution remains unclear. It appears that the significant decreases in the O_3 precursor abundance in autumn 2019 did not work to alleviate O_3 pollution in the PRD, although the underlying cause remains unclear.

The inter-annual variations of other related factors were examined in Guangzhou, namely the $PM_{2.5}$ concentrations and meteorological parameters. Compared with autumn 2018, the $PM_{2.5}$ concentrations significantly increased in 2019 (by 10%; $p < 0.01$, based on $PM_{2.5}$ data obtained from 11 AQMSs in Guangzhou). However, this increase can be excluded as a possible reason for the increased O_3 concentrations, due to the negative interactions between O_3 and $PM_{2.5}$ as reported by Li et al. (2019a). In addition, higher ambient temperature (by 4%; $p < 0.01$) and lower relative humidity (by 18%; $p < 0.01$) were recorded in autumn 2019 than in autumn 2018 (see Fig. 2); these meteorological conditions favor O_3 formation and may partly account for the worsened O_3 pollution in autumn 2019. The synoptic weather conditions during autumn 2019 certainly contributed to increases in O_3 pollution in the PRD and across southern China, compared with 2018, and significant increases in O_3 concentrations were indeed recorded at almost all of the AQMSs in these areas (see Fig. 1). However, these small or moderate variations in meteorological parameters should be insufficient to fully explain the significant increases in the O_3 concentration over the PRD in autumn 2019.

3.2. *Increased regional-background and locally-produced O_3 in 2019*

To identify the reasons for the significant increase in O_3

concentrations in autumn 2019, the contributions of background O_3 concentrations over different scales were quantified to the observed O_3 concentrations in Guangzhou and the PRD region (Fig. 3(a)). As shown, the regional background O_x that was transported into Guangzhou comprised dominant contributions to the ambient concentrations of O_x in Guangzhou in both 2018 (65% ± 12%) and 2019 (71% ± 9%). Similarly, the supra-regional background O_x transported into the PRD made important contributions to the ambient concentrations of O_x in the PRD in both 2018 (53% ± 13%) and 2019 (61% ± 12%). Compared with 2018, both the supra-regional and regional background O_x concentrations increased in 2019 in terms of their absolute values (by 18 ppbv and 19 ppbv, respectively) and relative contributions (by 8% and 6%, respectively). This evidence directly demonstrates that the increasing supra-regional background O_3 concentrations contributed to the increased O_3 pollution over the PRD, which then indirectly contributed to the increased O_3 pollution over Guangzhou in 2019 via the inward transport of increased regional background O_3 from the PRD. These results are consistent with those reported by Xue et al. (2014b) and Yang et al. (2019), who identified increased regional background O_3 concentrations as a key contributor to the severe O_3 pollution in the PRD and Hong Kong in southern China. In addition, the increased contributions from regional background are consistent with the severe O_3 pollution over the vast area of eastern China, which has continually worsened in recent decades (Liu and Wang, 2020a, b; Lu et al., 2018). Driven by the synoptic conditions, the widespread exacerbation of O_3 pollution over southern China in autumn 2019 (Fig. 1) should account for increases in the supra-regional background concentrations of O_3 in the PRD region.

The inter-annual variations in the concentrations of regionally produced O_3 over the PRD and locally produced O_3 in Guangzhou were also presented. Compared with 2018, the regionally and locally produced O_x concentrations didn't decrease and even slightly increased (by 1 ppbv) in 2019. In contrast, the concentrations of O_3 precursors, especially AVOCs, decreased significantly. It appears that the significant reductions in AVOCs and NO_x were inefficient for controlling local O_3 formation in autumn 2019. For comparison, the net in-situ rates of O_x production during the entire autumn season in urban Guangzhou was calculated using the MCM box model (see Fig. 3(b)). The results obtained using two independent methods are generally consistent, as the model-calculated daytime rate of O_x production in 2019 (13.80 ± 8.11 ppbv h^{-1}) was slightly higher than that in 2018 (13.52 ± 8.54 ppbv h^{-1}). Although the increases in the regionally and locally produced concentrations of O_3 are statistically insignificant and relatively small, compared with the changes in the supra-regional and regional background O_3 concentrations, it is important and unclear why significant decreases in the concentrations of NO_x and AVOCs in urban Guangzhou (the PRD) led to increases or non-decreases in the locally (regionally) produced O_3 concentrations. Analogously, it is not apparent why the local photochemical production of O_3 did not decrease in response to the large reductions in NO_x and AVOCs.

To investigate these aspects, a series of sensitivity modeling tests were conducted to quantify the effects of variations in the concentrations of individual major O_3 precursors (i.e., NO_x and VOCs) on the in-situ O_x production (see Fig. 4). The autumn 2018 was selected as a baseline and sensitivity tests were conducted using the target precursor concentrations constrained by the data in autumn 2019 while the other parameters were fixed. We found that the average daily maximum net rate of O_x production was 21.1 ± 8.1 ppbv h^{-1} in autumn 2018, and the effects of decreases of NO_x and VOCs are clearly double-edged. Notably, the decrease in AVOCs in 2019 significantly lowered the daily maximum net rates of O_x production (−4.57 ppbv h^{-1} or -21.7%; $p < 0.01$). This confirms that controlling AVOC emissions can effectively reduce the local production of O_3. Further inspection shows that of the major AVOC groups, decrease in aromatic compounds led to the largest decrease in the daily maximum net rates of O_x production (−2.37 ppbv h^{-1} or -11.3%; $p < 0.05$), followed by decreases in alkanes (−2.32 ppbv h^{-1} or -11.0%; $p < 0.05$) and alkenes (−0.48 ppbv h^{-1} or -2.3%; $p = 0.69$). The relatively small effects of decreases in alkenes on the O_x production relative to those of aromatics and alkanes are attributable to the higher concentrations of long-chain alkenes in 2019 than in 2018.

In comparison, a higher concentration level of isoprene led to greater local production of O_3, as demonstrated by a 1.24 ppbv h^{-1} (5.9%) increase in the daily maximum net O_x production rate in 2019 relative to

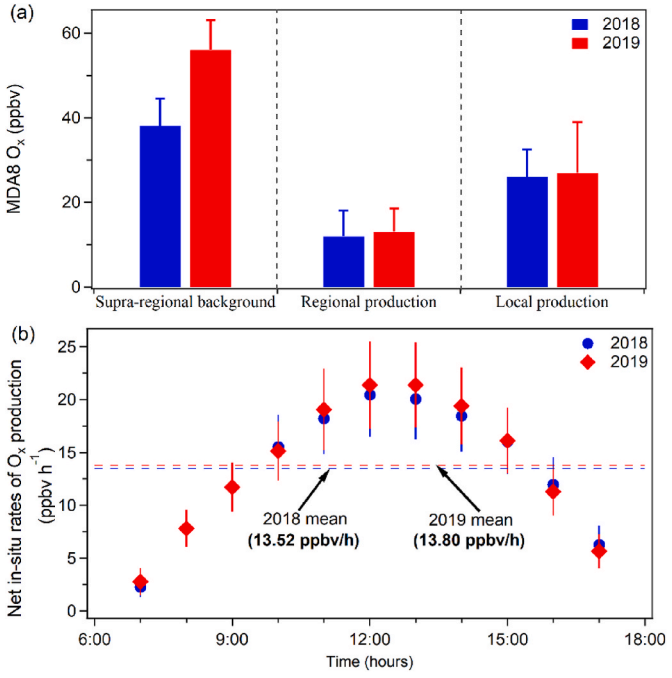

Fig. 3. (a) Contributions of the supra-regional background concentrations of O_x, Pearl River Delta (PRD)-regionally produced concentrations of O_x, and locally produced concentrations of O_x to the observed concentrations of O_x in Guangzhou; (b) Average diurnal variations in the modeled net in-situ rates of O_x production at an urban site in Guangzhou. The error bars indicate a half-standard deviation of the mean.

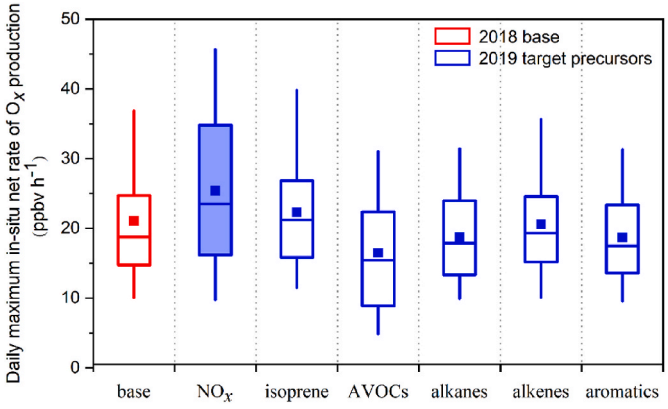

Fig. 4. Daily maximum net in-situ rates of O_x production calculated using the Master Chemical Mechanism box model at an urban site in Guangzhou in autumn. The red box represents the base case results in 2018, and the blue boxes represent the sensitivity test results with the target precursor (NO_x, isoprene, AVOCs, alkanes, alkenes, and aromatics) concentrations constrained by observational data from 2019, while other parameters are fixed. The box plot provides the 5th, 25th, 50th, 75th, and 95th quantiles of the results. The solid bar represents "NO_x disbenefits" (i.e., when decreased concentrations of NO_x lead to increased concentrations of O_3). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

2018. Although NO_x emissions were decreased (by 7% during the daytime) in 2019, the daily maximum net rate of O_x production was significantly increased in 2019 (by 4.35 ppbv h^{-1} or 20.7%; $p < 0.01$) due to a reduction in the NO titration effect. Evidently, the inadequate decrease in the emissions of NO_x and increase in the emissions of BVOCs enhanced the in-situ O_3 production, which negated the benefits of the significant decrease in the emissions of AVOCs. For comparison, similar sensitivity studies were also conducted using autumn 2019 as a baseline, and the obtained results are generally consistent with those of the above sensitivity tests (see SI Fig. S6).

3.3. O_3-precursor relationships and implications for coordinated VOCs and NO_x reduction

To establish future directions for the cost-effective control of O_3 pollution, a series of model scenarios was designed to establish the responses of O_3 concentrations to coordinated decreases in the emissions (concentrations) of AVOCs/NO_x in urban Guangzhou (see Fig. 5). Urban Guangzhou is clearly under an NO_x-saturated and VOCs-limited O_3 formation regime in autumn, and therefore decreasing the concentrations of AVOCs is the most efficient approach for achieving short-term decreases in ambient O_3 levels. However, it is challenging to achieve long-term O_3 concentration objectives via a control strategy that is focused solely on decreasing AVOC concentrations, particularly as the PRD region has relatively intense BVOC emissions (see SI Fig. S7). Specifically, highly reactive BVOCs generally cannot be controlled by anthropogenic efforts, and increased concentrations of BVOCs could partly negate the effectiveness of significant decrease in AVOC emissions, thereby increasing the difficulty of controlling O_3.

To achieve the long-term control of O_3 pollution, the control policy

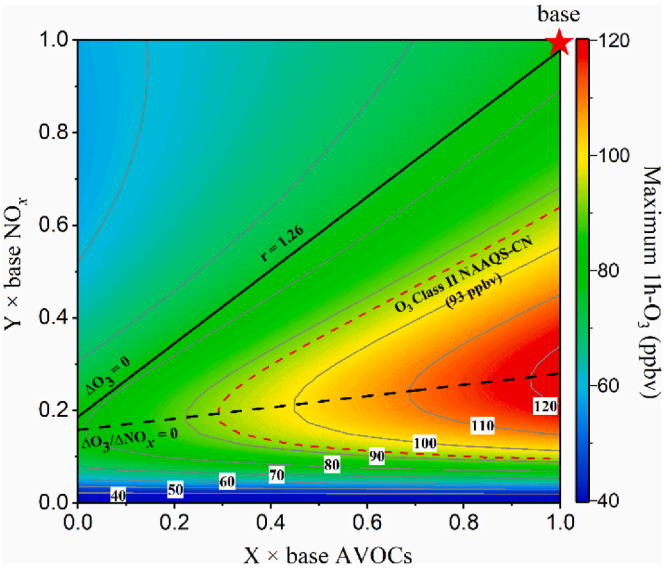

Fig. 5. Isopleths of the modeled maximum concentrations of ozone (O_3) as a function of X times the base concentrations of AVOCs and NO_x. The red pentagram represents the base (i.e., observed) concentrations of AVOCs and NO_x in autumn 2019. The black solid line represents the fitting line for a base case with an AVOC/NO_x reduction ratio of 1.26 [i.e., percentage change in concentration (ppbv)/percentage change in concentration (ppbv)], which exerts nearly zero effect on O_3 production (whereas the combined effects of an AVOC/NO_x reduction ratio >1.26 (i.e., above the fitting line) would lead to decreased O_3 production). The black dashed line represents the line fitting that demonstrates the nearly zero effect of decreases in the concentrations of NO_x on the production of O_3 (i.e., the boundary between the VOC-limited and mix-limited O_3 formation regime); the red dashed line represents the Class II Chinese National Ambient Air Quality Standard (NAAQS-CN) (i.e., 93 ppbv (hourly)). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

should target at transiting current O_3 formation regime from VOC-limited to NO_x-limited. For example, in urban Guangzhou, the desired O_3 attainment objective would be achieved if NO_x emissions were decreased by ≥ 84% (see the black dashed line in Fig. 5); however, this would be challenging to achieve using the existing immature technologies. We also found that an AVOC/NO_x concentration-reduction ratio >1.26 (the percentage-change in the concentration of AVOCs (ppbv)/percentage-change in the concentration of NO_x (ppbv); see the solid black line in Fig. 5) is a key requirement during a transition from high-to low-NO_x concentration conditions, as it prevents an exacerbation of O_3 pollution, and this ratio value is comparable to those determined by previous studies (He et al., 2019; Ou et al., 2016; Sun et al., 2019). Overall, these results underscore that scientifically based decreases in AVOC/NO_x emissions are required to prevent worsening O_3 pollution, and that large decreases in the emissions of NO_x are fundamental to realizing long-term decreases in the concentrations of O_3.

The relative increment reactivity (RIR) for major O_3 precursor groups were also calculated, and the results are shown in Fig. 6. RIR is defined as the ratio of the percentage decrease in the simulated net rate of O_x production to the percentage decrease in the concentration of the target precursor (10% in this case) (Cardelino and Chameides, 1995; Martien et al., 2003; Wang et al., 2018). A higher positive RIR value indicates that O_3 production is more sensitive to this precursor. AVOCs exhibited the highest positive RIRs (RIR = 1.22), and among AVOC groups, aromatics had the greatest influence (RIR = 0.70) on O_3 production. In contrast, the RIRs for the current NO_x concentrations were highly negative (RIR = −1.02). This finding suggests that decreasing the emissions of aromatic compounds would most effectively decrease the local formation of O_3 in urban Guangzhou (the PRD), whereas decreasing the emissions of NO_x by 10% would increase the formation of O_3.

3.4. Discussions: implications for future control of O_3 pollution in China

Current increases in the intensity and extent of O_3 air pollution in China have woken up the government and public, and the attainment of stipulated atmospheric O_3 concentrations has become a key target of air quality management in the upcoming 14th "Five-Year Plan" (2021–2025). Since 2013, the Chinese government has issued a series of emission control measures, such as "Air Pollution Prevention and Control Action Plan" (2013–2017) and "Three-Year Plan on Defending the Blue Sky" (2018–2021). As a consequence, the anthropogenic emissions of SO_2, NO_2 and CO have been reduced by 78%, 48% and 56% from 2013 to 2021, respectively, and the $PM_{2.5}$ concentrations have been reduced by 58% (Fig. S8), which were also demonstrated in previous

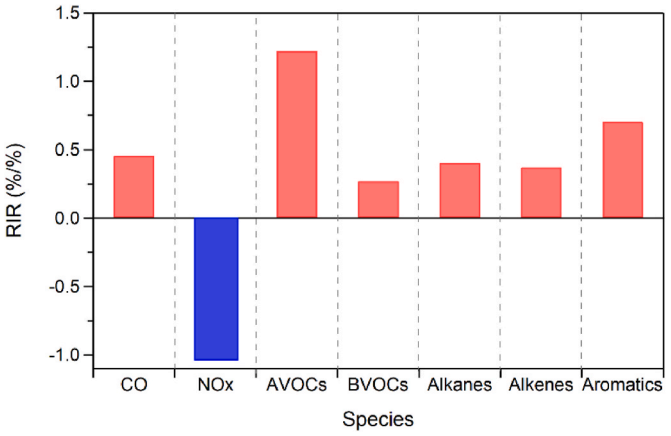

Fig. 6. The model-calculated relative increment reactivity (RIR) for ozone precursors (namely, CO, NO_x, AVOCs, and isoprene) and major AVOC groups (i.e., alkanes, alkenes, and aromatics).

studies (Cai et al., 2017; Maji et al., 2020; Zhao et al., 2021). However, these emission reduction measures have not been effective for mitigating O_3 pollution, as the O_3 concentrations still exhibited an upward trend in many regions of China (Lu et al., 2020; Wang et al., 2022). Reduction in NO_x emission is indeed the correct way to control photochemical O_3 pollution. However, extensive evidence indicates that the current strategy for controlling and decreasing NO_x emissions is insufficient for mitigating O_3 pollution, as O_3 formation persists in NO_x-saturated and VOC-limited regimes in a variety of urban areas (Sun et al., 2019). In comparison with efforts to control NO_x emissions, efforts to VOC emissions have been largely lagged in China, due to the complexity of emission sources and a lack of control technologies. Hence, the AVOC-focused control strategy has been adopted by the government as the hope to control O_3 pollution in the future step. In this context, the experience of O_3 pollution control in the PRD region, as presented in this study, provides a valuable lesson for the entire area of eastern China, which is currently struggling with worsening regional O_3 air pollution. Thus, the key implications of the O_3 pollution control in the PRD are as follows.

First, science-based coordinated control of AVOC and NO_x emissions is the key to lessen local O_3 production in short-term or medium-term stages. Under an NO_x-saturated O_3-formation regime, a small (or moderate) decrease in the concentrations of NO_x may negate the effectiveness of large decreases in the concentrations of AVOCs. The optimum AVOC/NO_x reduction ratio must therefore be regularly examined on a yearly (or seasonal) basis, and should be based on changes in synoptic weather conditions, progress in emission reduction, and the development of control technologies. Long-term O_3 attainment strategy will require large decreases in the emissions of NO_x, as this is the fundamental way to transform the current high-NO_x environment to a low-NO_x environment. Innovations in denitrification technology are highly needed to enable this transformation. Second, the emissions of BVOCs under unfavorable weather conditions are an important factor that perturbs attempts to control O_3 pollution, especially given the acceleration of afforestation and urban greening in recent decades in China. Given the high photochemical reactivity of BVOCs, a small increase in BVOCs may substantially influence the magnitude of the required decreases in emissions of AVOCs. Finally, it would be insufficient to resolve severe O_3 pollution by only local emission reductions in an individual region, as O_3 pollution is a regional environmental problem and can be transported over multiple regions. There is an urgent need for synchronous control on regional emissions over most areas of China towards national O_3 pollution mitigation, especially under unfavorable synoptic weather conditions.

4. Summary

We evaluated the effectiveness of the "Autumn O_3 Peak Reduction Campaign 2019" implemented in the PRD region, which aimed at mitigating O_3 pollution by largely cutting down anthropogenic emissions of VOCs and NO_x. This program was successful in reducing the ambient concentrations of AVOCs and NO_x, but failed to lessen regional O_3 pollution, which was even much worse in autumn 2019 compared to autumn 2018. Further inspections revealed that both the increased background and locally-produced O_3 contributed to the worsened O_3 pollution observed in the PRD in autumn 2019. At a large scale, the supra-regional and regional background O_3 concentrations significantly increased in the PRD due to the elevated O_3 concentrations from the vast surrounding areas. At a local scale, increased BVOCs and decreased NO_x negated the effect of significant decreases in AVOCs on in-situ O_3 formation. The unsuccessful experience of the PRD in reducing autumn O_3 peaks highlights the currently existing challenges of O_3 pollution control in China, and demonstrates the importance of science-based AVOCs/NO_x control strategy and synchronous control on regional emissions to the national O_3 pollution mitigation in China, which also has important implications for other countries suffering from serious O_3 air pollution.

Author statement

Min Zhao[1]: Conceptualization, Formal analysis, Writing – original draft. **Yingnan Zhang**[1]: Conceptualization, Formal analysis, Writing – review & editing. **Chenglei Pei**: Investigation, Resources. **Tianshu Chen**: Methodology, Software. **Jiangshan Mu**: Methodology. **Yuhong Liu**: Methodology. **Yujun Wang**: Investigation. **Wenxing Wang**: Supervision, Resources. **Likun Xue**: Conceptualization, Funding acquisition, Project administration, Writing – review & editing.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2022.116327.

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