Observation-Based Modeling of O₃–Precursor Relationships in Nanjing, China

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Abstract: Surface ozone (O₃) is formed through a series of photochemical reactions of nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in the atmosphere. In recent years, ozone concentrations in Nanjing have been increasing. To effectively mitigate ozone pollution, it is essential to understand the relationship between O₃ and its precursors. In this study, the observation-based model (OBM) coupled with the Master Chemical Mechanism (MCM), was applied to investigate the O₃-NO_x-VOCs relationship in the Nanjing Metropolitan Area of China in summer 2015. The OBM model well reproduces the levels and diurnal variations of O₃ with a high index of agreement value. Analysis of ozone formation potential (OFP) indicates that although observed alkanes are the most abundant VOCs (46.8%), their contributions to OFP are relatively small due to lower maximum incremental reactivity (MIR). Aromatics contribute the most to OFP (61.6%), followed by alkenes (18%). These two groups dominate in the top ten VOCs of OFP. In particular, m/p-Xylene shows a significant contribution to OFP with the highest OFP value over 30 $\mu g/m^3$. The relative incremental reactivity (RIR) results demonstrate that the reduction of anthropogenic VOCs (AVOCs) is the most efficient way to mitigate local O₃ pollution in Nanjing. Specifically, m/p-Xylene emissions should be reduced at it shows the highest RIR among all the AVOCs. Based on the results of OBM, a cutting ratio of AVOCs to NO_x of more than 0.46 is proposed to implement efficient control measurements in Nanjing for the study period.

Keywords: O₃ pollution, OBM, OFP, RIR.

1. INTRODUCTION

The troposphere ozone (O_3) is a secondary pollutant produced by the complex photochemical reactions of nitrogen oxides (NO_x) and volatile organic compounds (VOCs) emitted into the atmosphere. Due to its high oxidative capacity, O3 exhibits adverse impacts on human health (Brauer et al., 1997; Yang et al., 2012) and climate change (Stich et al., 2007; Unger et al., 2006) under high levels. In China, although the haze pollution has improved by the implementation of strict emission reduction measures (Zhang et al., 2015; Ma et al., 2019), the O₃ level has gradually increased (Lu et al., 2018; Li et al., 2017), especially in summer. High O_3 has become another issue in air pollution prevention and control in the fast-developing regions and urban areas in China (Xue et al., 2014). It is crucial to understand the relationship between O3 and the precursors for making efficient mitigation strategies.

The relationship between O_3 and its precursors can be reflected using the empirical kinetic modeling approach (EKMA) (Tan *et al.*, 2018; Jia *et al.*, 2016), which also indicates the sensitivities of O_3 to the

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changes of emitted NO_x and VOCs. Generally, O₃ formation is limited by VOCs in the urban areas (Guo *et al.*, 2013; Geng *et al.*, 2007; An *et al.*, 2015; Xu *et al.*, 2017) and by NO_x in rural site or background regions (Xu *et al.*, 2015). However, He *et al.* (2019) reported that the O₃ formation at a rural site is under the VOC-limited regime during the photochemical season in the western Pearl River Delta (PRD) region. Besides, Lyu *et al.* (2019) found that the dominating regime of O₃ formation changes from VOCs controlled in the non-pollution episode to transitional regime in the pollution episode.

The contributions of VOCs to O₃ in China have also been widely documented. Louie et al. (2013) found that aromatics and alkenes totally account for around 80% of the ozone formation potential (OFP) and the contributions volatile of oxygenated organic compounds (OVOCs) should be considered in the PRD region. Wang et al. (2016) suggested that the majority VOCs of the total OFP are alkenes (87.5%) and aromatics (83.0%), such as propene, toluene, m/p-Xylene, butane, o-xylene, and ethylbenzene, in the suburban area of Beijing. Lyu et al. (2016) reported that ethane (34.3%) and toluene (31.5%) have significant contributions to O₃ production at an urban site in Wuhan. Overall, aromatics and alkenes are the key VOCs for O₃ formation in the urban environment of China.

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Nanjing, the capital of Jiangsu Province, is a megacity located in the Yangtze River Delta (YRD) region. Driven by rapid economic development and industrialization, O₃ pollution has become severe in recent years in this region, with much higher O₃ level in May to September. The 90th percentile of the maximum daily 8-hour average (MDA8) O₃ concentration was 154 µg/m³ in YRD in 2014 (available at https://www.mee.gov.cn/hjzl/sthjzk/zghjzkgb/, last access: 29 July 2020). In particular, Nanjing has exceeded O₃ Grade II standard for two consecutive years since 2014, and had more than 40 pollution days due to un-attainment O₃ concentration in 2015, with a peak concentration of 114 ppb in August. Therefore, it is essential to analyze the O₃ formation and explore the relations between O_3 and its precursors in Nanjing.

In this study, the measured hourly concentrations of O_3 and its precursors (*i.e.*, VOCs and NO_x) were used to study the relationships between O_3 and the precursors in summer 2015. The contributions of different VOC groups to OFP are identified and then the most important VOC species for O_3 production are determined. Moreover, an observation-based model (OBM) coupled with a near-explicit photochemical mechanism was used to investigate the sensitivity of O_3 to its precursors and the dominant factor for O_3 abatement. Finally, the reasonable policy for emission reduction of important precursors is proposed.

2. METHODS

2.1. Observation Data

The field measurements of O_3 and VOCs were launched in the urban site of Naniing (32.04°N,118.78°E) (as shown in Figure 1). 56 VOCs were measured at the sampling site, including 29 alkanes, 16 aromatics, 10 alkenes and acetylene, with the time resolution of 1 hour from June 12 to 15, June 30 to July 5, and August 26 to September 5 in 2015. 4 trace gases (i.e., O₃, NO, NO₂, CO) were also continuously monitored at this urban site. In this study, the high O₃ days were defined as the days when MDA8 O_3 concentrations exceed 160 μ g/m³ (~ 80ppb) according to the ambient air quality standards (GB 3095-2012) released by Ministry of Ecology and Environment of the People's Republic of China. As a result, there are three days (from August 28 to 30) selected during the whole measurement period.

2.2. Model Application

An OBM coupled with the Master Chemical Mechanism version 3.3.1 (MCMv3.3.1, available at



Figure 1: The location of the urban site in Nanjing (the yellow star denotes the urban sampling site).

http://mcm.leeds.ac.uk/MCM/, last access: 25 July 2020) was applied to simulate O_3 formation and to investigate the sensitivity of O₃ formation to precursor concentrations at a given urban site. Unlike some lumped or condensed mechanism, such as SAPRC and carbon bond mechanism, MCM is a near-explicit photochemical mechanism, which involves 5829 species and 17221 reactions, to represent the multistep oxidation of VOCs in the atmosphere. OBM is the combination of a box model and observations of primary emitted species (NO_x, VOCs, CO, etc.), *i.e.* the model reset the concentrations of these species to the observed real-time value at each time step. To improve the accuracy of model performances, all the hourly observed data were linearly interpolated into 1 minute. The meteorological parameters were extracted from WRF outputs at Nanjing. Wet and dry deposition processes were not considered in the model.

OBM determines the O_3 -precursors relationships by calculating the relative incremental reactivity (RIR), which represents percentage change of O_3 production with that of precursors. The RIR for precursor X at site "S" is given by

$$\mathsf{RIR}(\mathsf{X}) = \frac{\left[P_{O_3}^{S}(X) - P_{O_3}^{S}(X - \Delta X)\right] / P_{O_3}^{S}(X)}{\Delta S(X) / S(X)}$$
(1)

where S(X) is the concentration of X (in ppb) at the site; and \triangle S(X) is the concentration change of X caused by a hypothetical change in S(X) (20% in this study); $P_{O_3}^{S}(X)$ and $P_{O_3}^{S}(X - \Delta X)$ represent O₃ production in this site conducted by the base case (unchanged observed concentrations of all O₃ precursors) and sensitive case (20% change in precursors concentrations), respectively. The positive RIR value of a given precursor suggests that once the VOC emission is controlled, the O_3 production will decrease. And the averaged RIR can be calculated by the following equation:

$$\overline{RIR}(X) = \frac{\sum_{1}^{N} [RIR(X) P_{O_3}^{S}(X)]}{\sum_{1}^{N} P_{O_3}^{S}(X)}$$
(2)

where N is the number of simulated days.

2.3. Model Evaluation

The index of agreement (IOA) was used to quantitatively evaluate the level of model performance. The equation is as follows:

IOA = 1-
$$\frac{\sum_{i=1}^{N} (O_i - S_i)^2}{\sum_{i=1}^{N} (|O_i - \bar{O}| + |S_i - \bar{O}|)^2}$$
(3)

where S_i and O_i are simulated and observed concentrations, respectively, \overline{O} is the mean of

observed values, and N is the number of valid samples. According to the equation expression, the closer the IOA is to 1, the better the agreement between simulated results and observations is.

Figure **2** shows the comparison of simulated and observed O_3 concentrations in Nanjing during 2015 summertime. Overall, the model shows a good agreement with observations, which well reproduces the O_3 concentrations and diurnal trends. The IOA value is 0.86, furthermore suggesting that the model results are valid in O_3 simulation owing to the acceptable performance of the OBM. Moreover, the IOA values all exceed 0.96 on high O_3 days, implying that high O_3 concentrations are mainly produced from local photochemical reactions.

3. RESULTS AND DISCUSSION

3.1. General Statistics

Figure **3**(**a**) shows the fractions of different VOC groups (*i.e.*, Alkanes, aromatics, alkenes and



Figure 2: Time series of simulated (red line) and observed (black dots) O₃ concentrations in Nanjing during 2015 summertime.



Figure 3: The ratio of different VOC groups for the total observed VOCs (a) and their shares in OFP (b) during the whole episode.

acetylene) to the total during the whole episode. Alkanes are the most abundant VOC species accounting for nearly 50% at sampling site, followed by acetylene (26.5%), aromatics (19.2%) and alkenes (7.5%). Generally, alkanes have relatively low reactivity with OH radicals compared to other VOC groups so that they are not necessarily the predominant VOC group for O_3 production. To some extent, O_3 formation potential (OFP) characterizes the effects of VOC species on O₃ formation which depends on the observed VOC mass concentration and the corresponding maximum incremental reactivity (MIR). MIR quantifies the sensitivity of O3 to VOCs (g O3/g VOC) under high NO_x conditions. Table 1 lists MIR values of all VOCs used in this study. The contributions

Table 1:	The List of All VOCs and their MIR Valu	es
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of different VOC groups to OFP are shown in Figure **3**(**b**). Aromatics have a largest share of 62%, indicating that this kind of VOCs have a high potential to promote O_3 formation. In addition, alkenes are also important contributor (accounts for 18%) among all the groups in Nanjing. Although alkanes and acetylene are the top two groups in terms of concentration, their contributions to OFP are relatively small due to the lower MIR values.

Figure **4** shows the top ten VOCs of OFP including six aromatic species, three alkene species and acetylene. M/p-Xylene has a significant contribution to OFP with the highest value over 30 μ g/m³, followed by toluene (around 16 μ g/m³) and ethylene (around 8.5

VOCs	MIR ^ª	VOCs	MIR ^ª
Ethane	0.28	n-Dodecane	0.55
Propane	0.49	Toluene	4
Isobutane	1.23	n-Propylbenzene	2.03
n-Butane	1.15	iso-Propylbenzene	2.52
2,2-Dimethylbutane	1.17	Ethylbenzene	3.04
2,2,4-Trimethylpentane	1.26	m/p-Xylene	9.75
Isopentane	1.45	o-Xylene	7.64
Cyclopentane	2.39	m-Ethyltoluene	7.39
n-Pentane	1.31	p-Ethyltoluene	4.44
2,3-Dimethylbutane	0.97	o-Ethyltoluene	5.59
2-Methylpentane	1.5	1,3,5-Trimethylbenzene	11.76
3-Methylpentane	1.9	1,2,4-Trimethylbenzene	8.87
n-Hexane	1.24	1,2,3-Trimethylbenzene	11.97
n-Heptane	1.07	m-Diethylbenzene	7.1
Methylcyclopentane	2.19	p-Diethylbenzene	4.43
Dimethylpentane	1.55	Styrene	1.73
2,3,4-Trimethylpentane	1.03	Benzene	0.72
Cyclohexane	1.25	Ethylene	9
2,3-Dimethylpentane	1.34	Isoprene	10.61
2-Methylhexane	1.19	Propylene	11.66
3-Methylhexane	1.61	1-Butene	9.73
Methylcyclohexane	1.7	1-Pentene	7.21
2-Methylheptane	1.07	1-Hexene	5.49
3-Methylheptane	1.24	Cis-2-butene	14.24
n-Octane	0.9	Trans-2-butene	15.16
n-Nonane	0.78	Cis-2-Pentene	10.38
n-Decane	0.68	Trans-2-Pentene	10.56
n-Undecane	0.61	Acetylene	0.95

^a The MIR values are available at https://intra.engr.ucr.edu/~carter/SAPRC/ (last access: 3 July 2020).



Figure 4: The top ten VOCs of OFP during the whole episode.

 μ g/m³) during the entire episode. Therefore, aromatics and alkenes play a crucial role in O₃ formation. Besides, it is noteworthy that isoprene, mainly from biogenic source, is also counted among the top ten VOCs of OFP with relatively higher MIR among all VOCs.

The contributions of VOCs to O_3 in other cities have been investigated in recent studies. Wang et al. (2018) reported that although alkanes were the most abundant VOC groups (accounting for 58.67%), followed by aromatics (27.32%), aromatics were the most important contributor to O₃ production (accounting for 57.99%) in Zhoushan city. This is consistent with the results shown in Figure 3 that the contribution of aromatics to O_3 formation is 61.6%. Zhou et al. (2020) calculated ozone formation potential (OFP) of VOCs for Qingdao city and indicated that alkenes/alkynes and aromatics contributed the most together, reaching 76.9% of the total OFP. That is slightly lower than our results that the total contribution of alkenes, aromatics as well as alkynes to OFP was 83.4%. For the top VOCs of OFP, m/p-xylene, ethylene, propylene, toluene and o-xylene were listed among the top 10 VOCs of OFP in Wang et al. (2018) and Zhou et al. (2020), which are consistent with the results shown in Figure 4.

3.2. Relationship Between O₃ and its Precursors

Figure **5** shows the averaged RIR analysis of O_3 precursors on high O_3 days. The precursors were divided into four groups, namely anthropogenic VOC species (AHC), CO, ISOP (isoprene), and NO_x. The RIR value of AHC is significantly positive reaching 0.33, followed by CO and ISOP (0.09 and 0.06,

respectively). In contrast, the RIR of NO_x is negative (about -0.19), indicating that the reduction in NO_x emissions is not conducive for O₃ mitigation but aggravating pollution in Nanjing. As a result, O₃ formation is controlled by VOCs in Nanjing during the O₃ pollution episode and the emission reduction of anthropogenic VOCs is most efficient for O₃ abatement. Additionally, it should be noted that the averaged RIR of CO was relatively higher than that of isoprene, demonstrating that the positive effect of CO on O₃ formation cannot be neglected during summer in Nanjing. This is consistent with the findings in Wuhan reported by Zeng *et al.* (2018), but in contrary to the finding by Wang *et al.* (2017) that the RIR of CO is lower than biogenic VOCs in summer in Hong Kong.



Figure 5: The RIR analysis for AHC, CO, isoprene (ISOP) and NO_x in Nanjing during O_3 pollution episode.

According to the analysis above, it is clear that mitigating ozone pollution should consider AVOCs reduction as a priority, although the efficient way is



Figure 6: The RIR analysis for the top 10 of anthropogenic VOC species in Nanjing during O_3 pollution episode.

difficult to establish under real situations. Thus, this study further conducts simulations of individual anthropogenic VOCs using the same model setups to determine the dominant AVOCs of reducing O₃. As shown in Figure 6, the top 10 anthropogenic VOCs include 6 aromatics, 3 alkenes, and isopentane (lumped into alkanes), with the RIRs of 0.01-0.06. The speciation of the top 10 VOCs for OFP and RIR are basically similar, including m/p-Xylene, propylene, ethylene, toluene, o-Xylene, 1,2,4-Trimethylbenzene and 1,2,3-Trimethylbenzene listed together. Particularly, m/p-Xylene has the highest RIR value, which is also the most important VOCs for OFP as presented in Figure 4. Therefore, m/p-Xylene has a dominant influence on O₃ formation compared to other AVOCs. It should be noted that the RIRs of AVOCs are not always positive, with negative values for alkanes such as n-octane, n-nonane and n-decane.

3.3. Implications for Control Measures

As mentioned above, O_3 formation in Nanjing is located in the VOC-limited regime during summer. However, it is not realistic to reduce VOC emissions alone for O_3 pollution control. Firstly, NO_x and VOCs are emitted into atmosphere by various emission sources simultaneously, which makes it difficult to reduce VOCs but to maintain NO_x emissions. Secondly, the inappropriate reduction ratio between VOCs and NO_x can hinder the decreases of O_3 due to the non-linear relationships for O_3 -VOCs- NO_x . Thus, this study conducted another 35 sensitive cases with different reduction ratios for VOCs and NO_x concentrations to find the most reasonable reduction ratio of the precursors. Figure **7** presents the averaged net O₃ increment (predicted values of sensitive cases minus that of original case) with different reduction percentages of AVOCs and NOx. The x-axis corresponds the NO_x reduction percentages from 0-50% with an interval of 10%. Each colored line means VOCs concentrations are constrained with the same reduction scale of NO_x. Not surprisingly, the averaged net O₃ increment gradually decreases with the increased reduction percentage of VOCs and increases with the increased reduction percentage of NO_x. This finding agrees with the analysis of O₃ formation process which is limited by VOCs during summer in Nanjing in Section 3.2. Especially, O₃ variations are always positive when VOCs are kept without reduction. As shown, with respect to zero O_3 positive increment, the maximum percentages of NO_x emission reduction are 21%-43% when the percentages of VOC emission reduction are 10%-20%. Hence, a reasonable reduction ratio (0.46) of VOCs to NOx is determined for O₃ abatement in the urban areas of Nanjing.

Currently, the estimation of VOC emissions and the source apportionment of VOCs in Yangtze River Delta have been widely documented in previous studies (Fu *et al.*, 2013; Liu *et al.*, 2018; Kudo *et al.*, 2014; Wang *et al.*, 2014). Zhao *et al.* (2020) used the positive matrix factorization (PMF) model to identify important VOC sources in the urban area of Nanjing and found that the highest contribution was from transportation, including diesel vehicular exhaust $(34\pm5\%)$ and gasoline vehicular exhaust $(27\pm3\%)$, followed by industrial emissions $(19\pm2\%)$, fuel evaporation $(15\pm2\%)$, and biogenic emissions $(4\pm1\%)$. An *et al.* (2017) pointed out vehicular emissions, which is related to transportation source, contributed significantly to VOCs (38%),



Figure 7: The averaged net O_3 increment with different percentages of AVOCs and NO_x reduction in Nanjing during O_3 pollution episode.

followed by industry source, including solvent usage (19%) and industrial production (10%), liquefied petroleum gas/natural gas (LPG/NG) usage (20%), and biomass/biofuel burning (13%) during summertime at Nanjing urban site. Overall, transportation and industry are the top two sources to contribute VOC emission. In addition, according to 2015 Nanjing anthropogenic emission inventory established by Nanjing Municipal Academy of Ecology and Environment Protection Science emission inventory, the significant contribution of VOC emissions is from industry (89%), followed by transportation (8.1%) and residential (repast and biomass burning, totally accounting for 2%).

It should be noted that without considering transportation, the VOC emissions from industry should be reduced, especially for the solvent usage and industrial process. Wu *et al.* (2016) also proposed that reducing VOC emissions from important industrial sources is effective for controlling VOCs in some regions where VOC emissions from industry source are high. Based on the results of section 3.3, the emission reduction ratio of VOCs to NO_x should be greater than 0.46.

4. CONCLUSIONS

In this study, we used the observations of 56 VOCs to evaluate their contributions to OFP during summer of 2015 in Nanjing, compared to concentration proportions. An Observation-Based Model with a near-explicit photochemical mechanism was applied to simulate the formation of O_3 and analyze the top precursors by calculating RIR.

Although observed alkanes and acetylene are the top two abundant VOCs (46.8% and 26.5%, respectively), their contributions to OFP are relatively small due to lower MIR. Aromatics has major contribution to OFP (61.6%), followed by alkenes accounting for about 18%. The OBM model performed а good agreement between simulation and which well-reproduced observation. the O_3 concentration trend and diurnal variations with higher IOA (0.86) during the entire period. The averaged RIR results of different VOC groups illustrated that O₃ pollution was in VOC-limited regime and AVOCs reduction was the most efficient way to mitigate local O₃ pollution in Nanjing, while NO_x reduction emission could enhance O₃ pollution due to the negative RIR value. OFP and RIR of all individual VOC are calculated and the top ten VOCs of OFP and RIR are mainly aromatics and alkenes, suggesting that these two VOC groups are of importance in producing O₃. In addition, m/p-Xylene has a significant contribution to OFP with the highest value over 30 μ g/m³, which also shows the highest RIR value among all VOCs, indicating that m/p-Xylene should be focused firstly in reducing VOC emission. The OBM has implications to propose an effective control strategy by simulating sensitive cases based on various reduction percent of AVOCs and NO_x . The cutting ratio of AVOCs to NO_x is supposed to be higher than 0.46 under the real situation.

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