Is atmospheric oxidation capacity better in indicating tropospheric $O_3$ formation?

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HIGHLIGHTS

• This study summarizes and evaluates different approaches that indicate $O_3$ formation.
• Isopleth and sensitivity methods are useful but have many prerequisites.
• AOC is a better indicator of photochemical reactions leading to $O_3$ formation.

ABSTRACT

Tropospheric ozone ($O_3$) concentration is increasing in China along with dramatic changes in precursor emissions and meteorological conditions, adversely affecting human health and ecosystems. $O_3$ is formed from the complex nonlinear photochemical reactions from nitrogen oxides ($NO_x = NO + NO_2$) and volatile organic compounds (VOCs). Although the mechanism of $O_3$ formation is rather clear, describing and analyzing its changes and formation potential at fine spatial and temporal resolution is still a challenge today. In this study, we briefly summarized and evaluated different approaches that indicate $O_3$ formation regimes. We identify that atmospheric oxidation capacity (AOC) is a better indicator of photochemical reactions leading to the formation of $O_3$ and other secondary pollutants. Results show that AOC has a prominent positive relationship to $O_3$ in the major city clusters in China, with a goodness of fit ($R^2$) up to 0.6. This outcome provides a novel perspective in characterizing $O_3$ formation and has significant implications for formulating control strategies of secondary pollutants.

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Special Issue—Frontier Progresses from Chinese-American Professors of Environmental Engineering and Science (Responsible Editors: Xing Xie, Jinkai Xue & Hongliang Zhang)
1 Introduction

Tropospheric ozone ($O_3$) is considered a key air pollutant that causes, among others, respiratory problems on humans and damages and reduced growth on plants (Lippmann, 1989; Chen et al., 2007; Feng and Kobayashi, 2009; Van Dingenen et al., 2009). It is mainly produced by photochemical reactions of volatile organic compounds (VOCs) and nitrogen oxides (NO$_x$) from both natural and anthropogenic sources, although transport from the stratosphere is possible (Husain et al., 1977; Steinfeld, 1998). $O_3$ and its precursors can be transported at great distances (Clarke and Ching, 1983; Lelieveld et al., 2009). Thus, its ambient level at a specific location is impacted by transported $O_3$ from upwind sources and locally produced $O_3$ through photochemical reactions.

$O_3$ formation occurs due to NO$_2$ photolysis but formed $O_3$ reacts with NO to regenerate NO$_2$ without net $O_3$ accumulation (Steinfeld, 1998). However, $O_3$ accumulates when hydroperoxy radical (HO$_2$) or peroxy radicals (RO$_2$) formed in VOCs oxidation by the hydroxyl radical (OH) replace $O_3$ in converting NO back to NO$_2$ (Kentarchos and Roelofs, 2003; Pollack et al., 2013; Ren et al., 2013; Tan et al., 2018). Thus, $O_3$ formation depends on the levels of NO$_x$, VOCs, and oxidants (OH, HO$_2$, and RO$_2$, Fig. 1(a)). At different atmospheric conditions, $O_3$ formation varies nonlinearly to different mixtures of NO$_x$ and VOCs (Sillman et al., 1990; Sillman, 1999). In rural areas, where NO$_x$ levels are usually low, $O_3$ peak concentration decreases when NO$_x$ is further reduced (NO$_x$-limited regime). Contrary, in urban areas, where NO$_x$ amounts are elevated, a decrease in NO$_x$ at constant VOC levels will increase $O_3$ peak concentrations (VOC-limited regime). Following this methodology, $O_3$ formation is usually split into VOC-limited regime, NO$_x$-limited regime, and transition regime that governing by both NO$_x$ and VOCs (Sillman, 1995; Sillman and He, 2002). Numerical studies have focused on identifying $O_3$ formation regimes for designing controlling strategies (Feng et al., 2016; Ye et al., 2016; Lu et al., 2019; Wang et al., 2019b). Xing et al. (2011) investigated the nonlinear response of $O_3$ to precursor emission changes in China and found that NO$_x$ control appears to be beneficial for $O_3$ reduction in the downwind areas which usually experience high $O_3$ levels. Jin and Holloway (2015) investigated the $O_3$ sensitivity regime from 2005 to 2013 in China. They found that reducing NO$_x$ emissions is an effective way to control $O_3$ pollution in the Pearl River Delta (PRD). Wang et al. (2019a) reported that the $O_3$ sensitivity turned into a transition regime in eastern China in recent years, which required more comprehensive emissions control strategies to alleviate $O_3$ better. Ding et al. (2022) highlighted the necessity of simultaneous VOCs and NO$_x$ emission control in winter while enhancing the NO$_x$ control in the summer. However, most of these studies neglected the importance of atmospheric oxidation capacity (AOC) as the formation regimes are changing. The AOC is a key factor that describes $O_3$ formation (Kentarchos and Roelofs, 2003; Prinn, 2003; Li et al., 2019b). Generally, AOC is defined as the total oxidation reaction rates of primary pollutants (such as VOCs) by the oxidants (e.g. OH, HO$_2$, and NO$_3$). And the concentrations of these oxidants are used as an indicator to assess the AOC level in the modeling studies. Regardless of the changes in the regime and emissions of NO$_x$ and VOCs, faster reaction rates, indicated by higher levels of oxidants, would lead to enhanced $O_3$ formation. The following question is then posed: Which is a better indicator when assessing $O_3$ formation. Here, we discuss different ways to understand $O_3$ formation and propose that AOC should be additionally considered when designing control policies.

2 Isopleth diagram

As mentioned before, an $O_3$ isopleth diagram is defined as a contour plot of interpolated maximum $O_3$ concentrations achieved as a function of VOC and NO$_x$ concentrations (Dodge, 1977). It is used to show the

![Fig. 1](a) The $O_3$ formation mechanism (highlighted parts are the major oxidants) modified from Steinfeld (1998) and (b) a typical $O_3$ isopleth.
degree of O$_3$ production by considering the relative abundances of NO$_x$ and VOCs, including constant VOCs composition and meteorological conditions (Milford et al., 1989; Jin and Demerjian, 1993; Menut et al., 2000). These isopleths provide characteristic O$_3$ ridgelines, which depict the maximum achievable O$_3$ concentration at the NO$_x$-limited, VOC-limited and transition regimes (Fig. 1(b)).

O$_3$ isopleth plots are widely used as a key methodology applied in O$_3$ control strategies. However, some complex prerequisites are required when interrelating the isopleths. To generate these plots, O$_3$ maximum concentration has to be predicted from a large number of numerical simulations performed with an atmospheric VOC/NO$_x$ chemical mechanism using varying initial concentrations of VOCs and NO$_x$. At the same time, all other variables are constant (Qian et al., 2019). In short, the following points need to be considered when interrelating O$_3$ isopleths:

1) A large amount of simulations is required to plot the isopleth;
2) The correlation of O$_3$ with NO$_x$ and VOCs varies greatly on the different assumptions and conditions (such as meteorological parameters), which is partly due to complex atmospheric processes such as regional transport;
3) The computation of isopleths is based on constant VOCs composition; however, ambient VOCs composition may vary in time as its total concentration changes;
4) Biogenic VOCs (BVOCs) are not considered in most cases as they are not controllable, although they represent a significant fraction of the total VOC budget, especially in suburban and rural areas;
5) There are no prescribed common conditions for determining O$_3$ formation regimes using the isopleths since they depend on multiple factors such as emission inventories, chemical mechanisms, and meteorological conditions;
6) Since the isopleths may encoushciner temporal variability (day-by-day changes), the methodology has limitations to represent a persistent O$_3$ pollution episode that lasts for several days.

### 3 Sensitivity methods

State-of-the-art chemical transport models (CTM) are also used to provide sensitivity analysis methods to identify O$_3$ formation regimes (Sillman et al., 1990). Thereby, NO$_x$ or VOCs are reduced or increased by a certain percentage. Then, the simulated O$_3$ concentrations are compared with the base case (without NO$_x$ and VOCs changes). A certain difference (e.g. of 5 ppb) is used as a criterion to verify if O$_3$ formation is sensitive to NO$_x$ or VOCs reduction. In sensitivity methods, NO$_x$-VOCs indicators from model prediction and ambient measurements are often used to split O$_3$ formation to different regimes, which involve secondary species produced concurrently with photochemical O$_3$ production (Sillman, 1995,1999; Sillman and He, 2002). These species are relatively long-lived, so they can be transported along with O$_3$, including peroxydes (e.g. H$_2$O$_2$), nitric acid (HNO$_3$), total reactive nitrogen (NO$_y$ = NO$_x$ + HNO$_3$ + peroxyacetyl nitrates + alky nitrates), and NO$_x$ reaction products (NO$_y$ = NO$_x$ - NO$_3$). The ratios among these species reveal information associated with O$_3$ formation chemistry. The purpose of deriving these indicators is to build a link of O$_3$-NO$_x$-VOC to reduce the peak O$_3$ by the controllable species. The widely used indicators include, but are not limited to, VOCs/NO$_x$, H$_2$O$_2$/NO$_x$, H$_2$O$_2$/NO$_y$, H$_2$O$_2$/HNO$_3$, O$_3$/HNO$_3$, O$_3$/NO$_y$, and O$_3$/NO$_2$. Also, productions rates are often used to calculate O$_3$ formation regimes, such as (P$_{HNO_3}$ + P$_{ROOH}$/P$_{HNO_3}$) and P$_{H_2O_2}$/P$_{HNO_3}$ based on the production rates of H$_2$O$_2$, HNO$_3$, and organic hydroperoxides (ROOH).

Some of these indicators are derived from routinely monitored pollutants (e.g. O$_3$) at observation sites or satellite retrievals (e.g. HCHO and NO$_2$). Although the above mentioned indicators can be calculated directly from model simulations or satellite data, they are not probing into the complex reactions leading to O$_3$ formation and having uncertainties in indentifying O$_3$ formation regimes.

In practical implementation, using these indicators to evaluate the O$_3$ formation regimes requires extensive measurement networks (Jin and Holloway, 2015). Also, results from these indicators should be evaluated separately on a daily even hourly basis, especially for specific hours such as noon to two hours before sunset to maintain effectiveness. Even all these prerequisites are met, the NO$_x$-VOCs indicators method may have limitations in understanding O$_3$ changes or identifying O$_3$ formation regimes when NO$_x$ or VOCs emissions are dramatically changed. For instance, during the COVID-19, O$_3$ concentrations in norther China increased when large emission reductions occurred due to reduced human activities during the lockdown (Gaubert et al., 2021; Zhu et al., 2021). One potential reason for this change was a shift in the O$_3$ formation regime due to the reduction in NO$_x$ emissions during the imposed lockdown (Wang et al., 2021b). However, this was concluded mainly based on indicators or isopleth analysis, which may not reflect reality accurately. In particular, during the COVID-19, the O$_3$ formation regime changed from VOC-limited to NO$_x$-limited or transition regimes, but the rising O$_3$ concentrations were found with the NO$_3$ reduction. Table 1 shows the ratios of grid cells attributed to the three O$_3$ formation regimes (NO$_x$-limited, VOC-limited and transition) during the COVID-19 lockdown (January and February 2020) under different precursors reductions in China. These regimes are calculated using the Community Multi-scale Air Quality (CMAQ) model.
Details on the simulation and analysis can be found in Zhu et al. (2021). It is evident that the depending on the selected indicators different results are produced when identifying \( \text{O}_3 \) formation regimes. The HCHO/\( \text{NO}_2 \) indicator predicts a higher percentage of grids for the \( \text{NO}_2 \)-limited regime and the \( \text{O}_3/\text{NO}_x \) predicts more for the transition one. Contrary, the \( \text{H}_2\text{O}_2/\text{NO}_y \) favors the VOCs-limited regime. In addition, these three indicators show different sensitivities when changes (reductions) in \( \text{O}_3 \) precursors species emissions are considered. When the VOC emissions decrease by 50%, there are no changes in the formation regimes indicated by the \( \text{H}_2\text{O}_2/\text{NO}_y \) ratio. In contrast, an enhancement of the grid percentage corresponding to the VOC-limited regime (∼46% increasing rate) is predicted using the HCHO/\( \text{NO}_2 \) ratio. Consequently, different conclusions can be reached using different indicators.

### 4 Atmospheric oxidation capacity (AOC)

Reassessing its formation mechanism, \( \text{O}_3 \) is formed through photochemical reactions. The overall processes are related to the atmospheric oxidation capacity (AOC), which can be characterized by the levels of atmospheric oxidants, mainly including hydrogen oxide radicals (H\( \text{O}_3 = \text{OH} + \text{HO}_2 \)) during daytime, and nitrogen oxide radical (NO\( \text{y}_x \)) (Jacob, 2000; Monks, 2005) during nighttime. There are also other approaches to define the AOC. Elshorbany et al. (2009) identified the AOC as the sum of the explicit oxidation rates of primary pollutants by the oxidants. Liu et al. (2021) calculated the AOC based on electron transfer during the secondary pollutants formation process. The AOC is a fundamental factor in understanding the formation of \( \text{O}_3 \) and other secondary species. Despite its potential, only a handful of studies reported the importance of AOC in explaining changes in \( \text{O}_3 \) formation in China. Li et al. (2019a) found that since 2013, high AOC along with PM\( \text{2.5} \) reductions is the reason for the reported \( \text{O}_3 \) increases in China. Since the PM\( \text{2.5} \) is considered as the sink of the HO\( \text{x} \) (a major component of AOC), its reduction leads to the increase of AOC. Wang et al. (2021c) and Zhu et al. (2021) also reported that the unexpectedly rising amounts of \( \text{O}_3 \) during the COVID-19 were mainly attributed to the enhanced AOC since they had similar variation trends and spatial distributions.

Herewith, we aim at better understanding the interplay between AOC and \( \text{O}_3 \) formation. Among all oxidants, HO\( \text{x} \) concentrations are related to \( \text{O}_3 \) formation significantly, and even their day-to-day co-variations have been noticed in different cities and episodes (Porter et al., 2017; Chen et al., 2020; Zhu et al., 2020). Zhao et al. (2021) reported that the average day-to-day co-variations between HO\( \text{x} \) and \( \text{O}_3 \) is larger than 0.55 in the PRD. However, there is no further evidence in the relationships between AOC with \( \text{O}_3 \) concentrations and changes in \( \text{O}_3 \) concentrations. As a very preliminary test, the results from COVID-19 periods are summarized. Table 2 shows the correlations between the HO\( \text{x} \) and \( \text{O}_3 \) in the major city clusters, including the North China Plain (NCP), the Yangtze River Delta (YRD), and the PRD in China during the COVID-19 (January and February, 2020). \( \text{O}_3 \) from biogenic source is also considered, since recent studies reported that the BVOCs played an important role in the city clusters due to the urban greening impacts (Ma et al., 2019; Ma et al., 2022). In general, there is a significant positive correlation between non-background \( \text{O}_3 \) and HO\( \text{x} \) in all these three regions. The maximum goodness of fit (\( R^2 \)) is predicted for the NCP (\( R^2 = 0.47 \)) where, the most significant increase of \( \text{O}_3 \) (average increase rate ∼54%) was reported (Zhu et al.,

### Table 1 Ratios of grid cells falling in different \( \text{O}_3 \) formation regimes (VOC-limited, \( \text{NO}_2 \)-limited, and transition regimes) suggested by three different indicators during COVID-19 lockdown (January and February 2020) under different precursors reductions cases. All results are calculated using Community Multi-scale Air Quality (CMAQ) model.

| Scenarios                          | Indicators | VOC-limited | \( \text{NO}_2 \)-limited | Transition |
|-----------------------------------|------------|-------------|---------------------------|------------|
| During COVID-19                   | HCHO/\( \text{NO}_2 \) | 0.13 | 0.70 | 0.17 |
|                                   | \( \text{O}_3/\text{NO}_y \) | 0.11 | 0.40 | 0.49 |
|                                   | \( \text{H}_2\text{O}_2/\text{NO}_y \) | 0.42 | 0.15 | 0.43 |
| VOCs reduced by 50%               | HCHO/\( \text{NO}_2 \) | 0.19 | 0.65 | 0.16 |
|                                   | \( \text{O}_3/\text{NO}_y \) | 0.14 | 0.39 | 0.47 |
|                                   | \( \text{H}_2\text{O}_2/\text{NO}_y \) | 0.42 | 0.15 | 0.43 |
| \( \text{NO}_x \) reduced by 50% | HCHO/\( \text{NO}_2 \) | 0.02 | 0.85 | 0.13 |
|                                   | \( \text{O}_3/\text{NO}_y \) | 0.03 | 0.51 | 0.46 |
|                                   | \( \text{H}_2\text{O}_2/\text{NO}_y \) | 0.24 | 0.19 | 0.57 |
| VOCs and \( \text{NO}_x \) reduced by 50% | HCHO/\( \text{NO}_2 \) | 0.04 | 0.79 | 0.17 |
|                                   | \( \text{O}_3/\text{NO}_y \) | 0.03 | 0.52 | 0.45 |
|                                   | \( \text{H}_2\text{O}_2/\text{NO}_y \) | 0.25 | 0.19 | 0.56 |
In addition, O₃ concentrations also show high sensitivity to the changes of AOC (average increase rate ~98%) in the NCP, represented by the higher regression slope (slope = 3.7). Compared to the NCP, the correlation is less strong (R² = 0.27) for the PRD. The latter is consistent with the findings of previous studies denoting that O₃ concentration remained almost constant during the COVID-19 (Wang et al., 2021a; Zhu et al., 2021). Considering O₃ concentrations (sum of background and non-background O₃), the positive correlations between it and AOC are even more significant with the R² up to 0.60 in the YRD during COVID-19 (Table 2). Besides, previous studies also reported that the elevated O₃ was corresponding to the enhanced AOC during the summertime (Li et al., 2019a; Qin et al., 2022), indicating that the AOC may be a better indicator to elucidate the O₃ formation. However, these correlations vary greatly under different emission control strategies in different regions. In the YRD, the R² drops from 0.6 to 0.03 when NOₓ emissions are reduced by 50%. While in the PRD, the R² value is more sensitive to VOC decreases (from 0.43 to 0.05). The AOC levels highly depend on the meteorological parameters and emission conditions. Thus, the correlation between O₃ and AOC changes accordingly to the variations in meteorology and emissions. More comprehensive simulations are required to determine their relationships better under different emissions control scenarios.

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### Table 2

The correlation between HOₓ and non-background (NB) O₃ and Oₓ in the major city clusters (NCP, YRD and PRD regions) in China during COVID-19 under different emission control conditions. The NB O₃ is the sum of O₃ from all emission sources (including both anthropogenic and biogenic sources). Units for NB O₃/Oₓ are ppb and ppt. Total grid cells are 284, 277, and 158 in the NCP, YRD, and PRD, respectively. Total data points are 10508, 10247, and 5846 in the NCP, YRD, and PRD, respectively.

| NB      | NCP       | YRD       | PRD       |
|---------|-----------|-----------|-----------|
| O₃      |           |           |           |
| During COVID-19 | O₃ = 3.7*HOₓ+0.2, R² = 0.47 | O₃ = 1.8*HOₓ+0.6, R² = 0.40 | O₃ = 1.2*HOₓ+1.6, R² = 0.27 |
| NOₓ reduced by 50% | O₃ = 1.5*HOₓ+0.7, R² = 0.49 | O₃ = 2.2*HOₓ–4.4, R² = 0.03 | O₃ = 2.0*HOₓ–9.2, R² = 0.55 |
| VOCs reduced by 50% | O₃ = 1.3*HOₓ+0.7, R² = 0.14 | O₃ = 2.8*HOₓ–3.4, R² = 0.39 | O₃ = 1.9*HOₓ–7.0, R² = 0.05 |
| VOCs and NOₓ reduced by 50% | O₃ = 1.2*HOₓ+0.6, R² = 0.29 | O₃ = 2.8*HOₓ–6.5, R² = 0.69 | O₃ = 2.3*HOₓ–8.9, R² = 0.46 |
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