

We appreciate the reviewer for the valuable comments and suggestions which have helped improve our manuscript. We addressed all of the specific comments individually. For clarity, the changes are **highlighted (in blue font) in the revised manuscript**. The itemized response/actions made to the manuscript are listed as below.

## **Reviewer 2**

Huang et al. established statistical fitting equations for data sets from China, the United States, and Europe, and studied the sensitivity of O<sub>3</sub> formation to precursors. This is an important topic at present. The author claimed that this parametric methodology overcomes the complexities and resource constraints inherent in conventional methods, and is expected as a unified tool to facilitate global ozone mitigation under evolving precursor emission patterns and climate change. Unfortunately, I do not agree with this view. The main reasons are as follows:

The authors established a unified statistical fitting equation to fit the nonlinear relationship between ozone and precursors. After comparing multiple nonparametric regression models, the authors found that the log-Bragg3 model is universal. However, the authors did not explain why the log-Bragg3 model equation is universal. The model lacks the basic connotation of atmospheric chemistry and atmospheric physics, but is only a statistical equation obtained by fitting. In terms of mathematics, one can completely fit a very complex nonlinear relationship with high precision by constructing high-order statistical equations. This is just a process of approximating fitting by the infinite series method in mathematics. However, this operation has not made any essential improvement to the study of ozone sensitivity. It is just a mathematical technique. Even if, as the author says, common results have been obtained in the existing data of different cities, there is no guarantee that universal results will be obtained in the future.

**Response/Action 2-1: All of the seven studied empirical parametric models (Equations 1-7) are selected on the basis of the basic connotation of ozone formation chemistry**, rather than just a process of approximating fitting by the infinite series method in mathematics.

**Based on the Empirical Kinetic Modelling Approach (EKMA) isopleth plot, one of the most validated diagnostic methods, the daytime ozone production (DPO<sub>3</sub>) exhibits a characteristic skewed curve when plotted against NO<sub>x</sub> or NO<sub>2</sub> for a specific VOC reactivity (VOCR) (Graphical Abstract). This type of skewed and asymmetric diagram describing the DPO<sub>3</sub>–NO<sub>x</sub> (or NO<sub>2</sub>) relation had been reported in previous atmospheric chemistry relevant studies** (Pusede and Cohen, 2012; Romer et al., 2018; Nussbaumer and Cohen, 2020; Guo et al., 2023; Yang et al., 2021). As shown in the Graphical Abstract, ozone production rises with NO<sub>x</sub> is insensitive to VOCR. Reducing NO<sub>x</sub> is more effective than controlling VOCs to mitigate ozone. As NO<sub>x</sub> rises, it reaches its maximum and became limited by both NO<sub>x</sub> and VOCR. This indicates that controlling either or both precursors can effectively reduce ozone acceleration. With further NO<sub>x</sub> increase, it grows with VOCR but declines with NO<sub>x</sub>. Here, VOC control becomes the key for ozone mitigation, while NO<sub>x</sub> reduction potentially leads to increase in ozone pollution. On a DPO<sub>3</sub>–NO<sub>x</sub> (or NO<sub>2</sub>) curve, the partition

point and transition point are two key  $\text{NO}_x$  (or  $\text{NO}_2$ ) levels for differentiating the  $\text{O}_3$ - $\text{NO}_x$ -VOC sensitivity. The partition point is defined as the peak  $\text{DPO}_3$  corresponding  $\text{NO}_x$  (or  $\text{NO}_2$ ) level, where the ozone formation equals to consumption, distinguishing the  $\text{NO}_x$ -limited/transition regime (to the left) and the VOC-limited regime (to the right); the transition point is defined as the  $\text{NO}_x$  (or  $\text{NO}_2$ ) level at the position indicating the onset of diminishing ozone production with respect to  $\text{NO}_x$ , which further differentiates the  $\text{NO}_x$ -limited and transition regimes. As referred to the study by Yang et al. (2021), we determined the transition point as the position with a half of the maximum curve slope in the present study.

**The above basic connotation of ozone formation chemistry for the hypothesized skewed and asymmetric  $\text{DPO}_3$ - $\text{NO}_x$  (or  $\text{NO}_2$ ) curve was detailed in Section 1 of the revised manuscript (Line 47-61).**

Among all studied models, the *log-Bragg3* model was identified as the best model for globally characterizing the  $\text{DPO}_3$ - $\text{NO}_x$  (or  $\text{NO}_2$ ) relation and diagnosing  $\text{O}_3$ - $\text{NO}_x$ -VOC sensitivity (or referred to as ozone formation regimes, OFRs), based on the routine recordings of  $\text{O}_3$  and  $\text{NO}_2$  or  $\text{O}_3$  and  $\text{NO}_x$ . The **improvements** of this model to the study of ozone sensitivity include:

- (1) This model effectively **addresses the limitations of those commonly used methods (Line 39-46) as well as the prior non-parametric fitting of  $\text{DPO}_3$ - $\text{NO}_x$  (or  $\text{NO}_2$ ) curve (Line 62-77)**, both of which are inadequate for elucidating the evolution of  $\text{O}_3$ - $\text{NO}_x$ -VOC sensitivity on the large spatiotemporal scale; therefore, this model **makes it easier for OFR diagnosis that can be adaptable to different locations and different time**, even though the partition and transition points, two key diagnostic  $\text{NO}_x$  (or  $\text{NO}_2$ ) levels, are not always the same at different locations or time.
- (2) As discussed in Section 3.5, this model is also able to **provide implications of ozone formation intensity and the associated chemical processes, indicating by its parameters.**

The **universal results** of the present study include: **(1) the basic contour of  $\text{DPO}_3$ - $\text{NO}_x$  (or  $\text{NO}_2$ ) diagram is a skewed and asymmetric curve**, and was verified universally prevalent, including regions with severe  $\text{PM}_{2.5}$  contamination where ozone formation is additionally influenced by aerosol-inhibited photochemical regime **(Line 171-174 in the revised manuscript)**; **(2) the *log-Bragg3* model (Equation 3)** performed the best in characterizing this relation, adaptably resolves the  $\text{O}_3$ - $\text{NO}_x$ -VOC sensitivity continuum, and provide parametric insights into ozone formation intensity (*d*), associated chemical processes (*b*), and the  $\text{O}_3$ - $\text{NO}_x$ -VOC sensitivity partition threshold (*e*). **In fact, we have confirmed that this model can also fit the data from other regions and periods very well.**

**In the future, it is expected to be able to adaptably obtained the parameters of the  $\text{DPO}_3$ - $\text{NO}_x$  (or  $\text{NO}_2$ ) curves and diagnose the  $\text{O}_3$ - $\text{NO}_x$ -VOC sensitivities at different locations and different time.**

In fact, since 2020, there have been many breakthroughs in the research on the sensitivity of  $\text{O}_3$  formation to precursors. The city lockdown measures during the COVID-19 period

provided natural experiments for the emission reduction of precursors. Some new insights have even overturned the past understanding of atmospheric chemistry's sensitivity to ozone, and thereby improved the simulation effect of atmospheric ozone generation. This paper only focuses on research data before 2019, and it seems to deliberately avoid the adverse effects of emission reduction on simulation during the epidemic. Therefore, it is difficult to believe that the results of this study have achieved universal success. Are the study findings applicable during the COVID-19 pandemic in 2020?

**Response/Action 2-2:** Yes, in addition to the stations and periods incorporated in the present study, we have confirmed that such a skewed and asymmetric diagram of  $\text{DPO}_3\text{-NO}_x$  (or  $\text{NO}_2$ ) relation is also prevalent in other regions (e.g., Taiwan) and periods (2020-2022), where the data can also be fitted well by the *log-Bragg3* model.

The reason using the data before 2019 is that we aim to verify whether such a skewed and asymmetric diagram of  $\text{DPO}_3\text{-NO}_x$  (or  $\text{NO}_2$ ) relation can also be observed in those regions where the ozone formation is additionally influenced by the aerosol-inhibited photochemical regime, and the  $\text{PM}_{2.5}$  pollution during our studied period was significantly more severe in the regions of BTH, FWP and YRD in China compared to other regions (e.g., the European region and US).

The paper mentioned their past research results, but did not explain in depth the improvements of this method.

**Response/Action 2-3:** Thank you for this valuable comment. The distinctions and connections between the current study and our previous work (Huang et al., 2025) were **added in the revised manuscript (Line 62-83)**.

The non-parametric approach in our previous study is a commonly used method for smoothing a fluctuating numerical series within local neighbourhoods, enabling the identification of intrinsic  $\text{DPO}_3\text{-NO}_x$  (or  $\text{NO}_2$ ) relation. However, **two limitations still persist: (1)** a fixed smoothing span, the key configuration for non-parametric smoothing, failed to exhibit robustness in fitting performance across studied sites, which leads to uncertainty in determining the partition point and inhibits the adaptability of this method across a broader spatiotemporal range; **(2)** the non-parametric approach provided no information on the curve's height and width, which determine the transition point and vary with locations, study periods and environmental factors (e.g., temperature, VOCs, etc.). As studied by Guo et al. (2023), the basic contour of the regular  $\text{DPO}_3\text{-NO}_x$  (or  $\text{NO}_2$ ) curve would not vary with the relative humidity, temperature, season, altitude, mixing layer height and VOC species. This environmental stability makes it possible to be parametrically characterized. **Therefore, seeking an effective empirical parametric model is necessary for more adaptably characterizing the  $\text{DPO}_3\text{-NO}_x$  (or  $\text{NO}_2$ ) relation and figuring out both the partition and transition points. This is the most important objective of the present study.**

Even though the  $\text{DPO}_3\text{-NO}_x$  (or  $\text{NO}_2$ ) relation, as depicted in the Scenario A, B or C within a completed skewed curve in Graphical Abstract, was firstly convinced regionally prevalent and

able to effectively diagnose OFRs' spatial pattern in that previous study, **it remains uncertain whether or not this kind of regular DPO<sub>3</sub>-NO<sub>x</sub> (or NO<sub>2</sub>) relation is globally prevalent. Therefore, it is essential to firstly verify the universality of this relation using data from routine monitoring networks worldwide.** Furthermore, based on the non-parametric approach, our previous study revealed that the applicability and reliability for OFR diagnosis differed between the DPO<sub>3</sub>-NO<sub>x</sub> and DPO<sub>3</sub>-NO<sub>2</sub> curves at several observation stations in Hong Kong. **Accordingly, the present study also attempts to compare the reliability between the two curves in diagnosing O<sub>3</sub>-NO<sub>x</sub>-VOC sensitivity across a broader spatial range.**

Many details of the paper are obscure and difficult to understand. Especially in the explanation of professional terms and details of model parameters. At the same time, the visualization of the graph is also very vague, which seriously affects normal reading.

**Response/Action 2-4:** Thank you for this valuable comment. We speculate that the key professional terms concerned by the reviewer, might be the two words "partition point" and "transition point". **The definitions as below were added in the revised manuscript (Line 55-61).**

“On a theoretically regular DPO<sub>3</sub>-NO<sub>x</sub> (or NO<sub>2</sub>) curve (Graphical Abstract), the partition point and transition point are two key NO<sub>x</sub> (or NO<sub>2</sub>) levels for differentiating the O<sub>3</sub>-NO<sub>x</sub>-VOC sensitivity. The partition point is defined as the peak DPO<sub>3</sub> corresponding NO<sub>x</sub> (or NO<sub>2</sub>) level, where the ozone formation equals to consumption, distinguishing the NO<sub>x</sub>-limited/transition regime (to the left) and the VOC-limited regime (to the right); the transition point is defined as the NO<sub>x</sub> (or NO<sub>2</sub>) level at the position indicating the onset of diminishing ozone production with respect to NO<sub>x</sub>, which further differentiates the NO<sub>x</sub>-limited and transition regimes. As referred to the study by Yang et al. (2021), we determined the transition point as the position with a half of the maximum curve slope in the present study.”

**The model parameters were detailed in Section 2.1. Additionally, the rationale for selecting these studied models, detailed as below, was added in the updated Supplement (Text S1).**

“The Equation 1 in the article text and the Equations S1-S4 provided here are usually used to describe a phenomenon where the Y variable increases to reach a maximum at a certain level of the X variable, and decreases afterwards. For example, they can be applied to determine the maximum growth rate of plant at its corresponding optimal temperature level, as well as in the cases related to bioassays in toxicology/biology study: low doses of exogenous substances induce irritation effects. Only Equation 1 in the article text is capable to describe a skewed and asymmetric curve with a maximum, whereas Equations S1-S4 provided here are limited to describing the normal and symmetric curves.

$$Y = d \times \exp[-b \times (X - e)^2] \quad (S1)$$

$$Y = c + (d - c) \times \exp[-b \times (X - e)^2] \quad (S2)$$

$$Y = \frac{d}{1+b \times (X-e)} \quad (S3)$$

$$Y = c + \frac{d-c}{1+b \times (X-e)} \quad (S4)$$

The *Poly2* model (Equation S5) provided as below can also be used to describe a symmetric curve and is ever applied in fitting the relation of O<sub>3</sub>-HCHO/NO<sub>2</sub> ratio for diagnosing the O<sub>3</sub>-NO<sub>x</sub>-VOC sensitivity (Jin et al., 2020).

$$Y = b_0 + b_1 \times X + b_2 \times X^2 \quad (A5)$$

In the present study, the DPO<sub>3</sub>-NO<sub>x</sub> (or NO<sub>2</sub>) diagram is hypothesized to be a skewed and asymmetric curve with a maximum, and thus can be appropriately described by Equation 1. To explore more alternative fitting approaches, we attempted to reduce the skewness of the DPO<sub>3</sub>-NO<sub>x</sub> (or NO<sub>2</sub>) curve by logarithmizing the NO<sub>x</sub> (or NO<sub>2</sub>) concentrations. Therefore, the Equations 2-7 in the article text are the transformed forms of the Equation 1 and Equations S1-S5 with the X-coordinate logarithmized.”

Finally, the Clearer figures were provided in the revised manuscript.

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