

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 1

In “Technical Note: Adaptably diagnosing O₃-NO_x-VOC sensitivity evolution with routine pollution and meteorological data” Huang and Liao investigated the sensitivity of O₃ formation at selected sites in China, the US and Europe by applying different fit equations to the datasets. The authors identify most of the studied regions to be dominated by VOC-limited O₃ formation sensitivity.

While this is generally an important topic to investigate, unfortunately this study seems incoherent and is often difficult to follow. It remains largely unclear why and how the suggested fit equations are applied to the data and even more important what the added value of this analysis is. The in-situ observations investigated in this study can be used to directly infer the dominating sensitivity instead of using fit functions. Are any generalized conclusions drawn from the fitting? Could it be applied to other regions where observations are not available and how would that be possible considering that crossover points occur at NO_x to VOC ratios that are characteristic to each location?

1. **Why** the suggested fit equations are applied to the data?

As introduced in Section 1: Introduction, the ozone level has increased in most urban areas worldwide, and the O₃-NO_x-VOC sensitivity has likely evolved in response to the divergent trends in precursor emissions. **Elucidating its long-term evolution is critical for effective ozone mitigation.**

However, those commonly used methods for ozone formation regime (OFR) diagnosis, such as the Empirical Kinetic Modelling Approach (EKMA) isopleth plot and chemical indicators (e.g., H₂O₂/HNO₃, H₂O₂/NO_z, etc.), heavily rely on observation-based or numerical models, constrained by limited field data and computational demands. They are typically applied in case studies (Sillman and He, 2002; Sillman and West, 2009; Xue et al., 2014; Ou et al., 2016; Li et al., 2018). Although the satellite-derived HCHO/NO₂ ratio (FNR)-based method enables the regional scale long-term O₃-NO_x-VOC sensitivity diagnosis (Jin and Holloway, 2015; Ren et al., 2022; Wang et al., 2021; Zhang et al., 2024), its fixed daily sampling time restricts insights into other hours, and the sensitivity always varies over time. **These constraints highlight the necessity for more flexible and adaptable approaches.**

For a specific VOC reactivity (VOCR), the daytime ozone production (DPO₃) exhibits a characteristic skewed curve when plotted against NO_x or NO₂ (Graphical Abstract), which is transformed from the EKMA plot (Pusede and Cohen, 2012; Romer et al., 2018; Nussbaumer and Cohen, 2020; Guo et al., 2023; Yang et al., 2021). On a DPO₃-NO_x (or NO₂) curve (Graphical Abstract), the partition point and transition point are two key NO_x (or

NO₂) levels for differentiating the O₃-NO_x-VOC sensitivity. The **partition point** is defined as the peak DPO₃ corresponding NO_x (or NO₂) level, where the ozone formation equals to consumption, distinguishing the NO_x-limited/transition regime (to the left) and the VOC-limited regime (to the right); the **transition point** is defined as the NO_x (or NO₂) level at the position indicating the onset of diminishing ozone production with respect to NO_x, which further differentiates the 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.

Although the in-situ observations investigated in this study can be used to directly infer the dominating sensitivity via non-parametric approach (Huang et al., 2025), **two limitations still persist about that method: (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.). **The basic contour of the regular DPO₃-NO_x (or NO₂) curve would not vary with the relative humidity, temperature, season, altitude, mixing layer height and VOC species** (Guo et al., 2023). **This environmental stability makes it possible to be parametrically characterized. Therefore, seeking an effective empirical parametric model is necessary for more adaptably characterizing the DPO₃-NO_x (or NO₂) relation and figuring out both the partition and transition points.** This is the most important objective of the present study.

2. How the suggested fit equations are applied to the data?

As introduced in Section 2: Methodology, the DPO₃-NO_x (or NO₂) relation was regressed with the five-percentile-binned NO_x (or NO₂) concentrations (or logarithms) and their corresponding average DPO₃ levels. The DPO₃ was defined as the difference between the MDA8-daytime (7:00-19:00 Local Time (LT)) hourly ozone concentration and the ozone concentration at 6:00 LT. A total of seven parametric models (Equations 1-7) were individually applied to characterize the DPO₃-NO_x (or NO₂) relation. As in the bellowing **Response/Action 1-9** to Specific comment 1-9, we gave a more detail introduction of the rationale for selecting these studied models, which was added in the updated Supplement of the revised manuscript (Text S1).

3. What is the added value of this analysis?

In order to address the limitations of those commonly used diagnostic methods (**Line 39-46 in revised manuscript**) and the prior non-parametric fitting of DPO₃-NO_x (or NO₂) curve (**Line 62-77 in the revised manuscript**), the present study aims to seek an effective empirical parametric model for more adaptably characterizing the DPO₃-NO_x (or NO₂) relation and determining the dominating sensitivity of ozone formation. After a series of analyses as in Section 3.2: Which is the most capable parametric model, we identified that the *log-Bragg3*

model (Equation 3) performed the best.

Therefore, **one of the added values** of these analyses is that they make it **easier for OFR diagnosis that can be adaptable to different locations and different time**, even though the crossover points do not always occur at the same NO₂ mixing ratio. **This is particularly important for elucidating the evolution of O₃-NO_x-VOC sensitivity on the large spatiotemporal scale.** Furthermore, as discussed in Section 3.5: Implications of the *log-Bragg3* model's parameters (*b*, *d*), **the identified model (*log-Bragg3* model, Equation 3) is also able to provide implications of ozone formation intensity and the associated chemical processes, indicating by its parameters.**

4. Are any **generalized conclusions** drawn from the fitting?

Based on the above responses, the generalized conclusion from the fitting is that the *log-Bragg3* model (Equation 3) performed the best, compared with other models, in adaptably characterizing the DPO₃-NO_x (or NO₂) relation and diagnosing the dominating sensitivity of ozone formation, which is also able to provide the implications of ozone formation intensity and the associated chemical processes.

5. Could it be applied to other regions where observations are not available?

It could still be applied in other regions **where other reliable reanalysis data are available**, even though there is no observation.

6. How would that be possible considering that crossover points occur at NO_x to VOC ratios that are characteristic to each location?

Yes, the crossover points are characteristic to different locations, and they theoretically depend on local condition (e.g., VOCs or other relevant pollutants/radicals, meteorological factors, etc.). However, our identified model can solve this problem by adaptably fitting the data at different locations. **This is based on a hypothesis that the daytime ozone production (DPO₃) exhibits a characteristic skewed curve when plotted against NO_x or NO₂ for a specific VOC reactivity (VOCR) (Line 47-48 in the revised manuscript).** And indeed, **as in Section 3.1, the above hypothesized DPO₃-NO_x (or NO₂) relation was empirically validated worldwide**, even in regions with severe PM_{2.5} contamination, where the ozone formation is additionally influenced by the aerosol-inhibited photochemical regime, such as BTH, FWP and YRD in China (Ivatt et al., 2022; Geng et al., 2021; Kong et al., 2021; Xiao et al., 2022).

From this perspective, **the parameters in the identified model (*log-Bragg3*, Equation 3) reflect the local condition to some extent.** For example, the fitting parameter *e* varied with regions, as listed in Table S1 and Table S2; higher value indicates higher partition point (or crossover point as referred by the reviewer). Furthermore, as discussed in Section 3.5, the parameter *d* represents the maximum DPO₃ level, exhibiting higher ozone production with higher value; and higher value of parameter *b* characterizes a steeper curve, indicating a condition that favors faster change in ozone production efficiency for a given increment of NO_x.

It is further concerning that the authors have published a paper earlier this month (Huang et al., 2025), which they are now referring to have “critical limitations” which “fail” in respect to two different aspects (Line 54f.). This makes me wonder why the authors have not previously fixed these issues, considering that this previous paper was published one month after the submission of this manuscript.

We have clarified the distinctions and connections between the current study and our previous work (Huang et al., 2025), as outlined in the bellowing **Response/Action 1-1** to Specific comment 1-1.

Some statements are further not backed with the current literature. The authors often use terms that are not commonly known in literature and do not provide sufficient definitions or explanations. The same applies to abbreviations that are not defined when first used. The figures have too many panels, are too small and have a low resolution, which makes them difficult to read and understand the results.

1. The issue regarding statements not backed with literatures was addressed as outlined in the bellowing **Response/Action 1-16** to Specific comment 1-16.

2. We speculate that the terms concerned by the reviewer, which are less frequently used in the literatures, might be the two terms "partition point" and "transition point". **The definitions as below were added in the revised manuscript (Line 55-62).**

“On a theoretically regular $\text{DPO}_3\text{-NO}_x$ (or NO_2) curve (Graphical Abstract), the partition point and transition point are two key NO_x (or NO_2) levels for differentiating the $\text{O}_3\text{-NO}_x\text{-VOC}$ sensitivity. The partition point is defined as the peak DPO_3 corresponding NO_x (or NO_2) level, where the ozone formation equals to consumption, distinguishing the NO_x -limited/transition regime (to the left) and the VOC-limited regime (to the right); the transition point is defined as the NO_x (or NO_2) level at the position indicating the onset of diminishing ozone production with respect to NO_x , which further differentiates the 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.”

3. All abbreviations were defined upon their first use in the revised manuscript, such as the term “OFR” as exemplified in bellowing **Response/Action 1-2** to Specific comment 1-2.

4. The Clearer figures were provided in the revised manuscript.

Considering these various drawbacks, unfortunately, I cannot recommend this manuscript for publication in its current state as it does neither meet the scientific nor the methodological standards of an ACP publication. If the authors wish to improve their manuscript in the future, please find more detailed comments and questions in the following, which might be helpful for revising the study.

Specific comment 1-1: Line 53 ff.: Could the authors describe the study of Huang et al., 2025?

What were the methods applied and the findings of this study?

Response/Action 1-1: Thank you for this valuable comment. The distinctions and connections between the current study and our previous work (Huang et al., 2025), detailed as below, were added in the revised manuscript (Line 62-84).

“In our previous study (Huang et al., 2025), the $\text{DPO}_3\text{--NO}_x$ (or NO_2) relation, as depicted in the Scenario A, B or C within a completed skewed curve in Graphical Abstract, was proved widespread based on the routine monitoring data in the Greater Bay Area, South China, which was smoothed by a non-parametric regression technique. The smoothing curve was able to effectively characterize the regional spatial pattern of $\text{O}_3\text{--NO}_x\text{--VOC}$ sensitivity, differentiating the ozone formation regimes (OFRs) in to the NO_x -limited/transition regime and the VOC-limited regime, and was further utilized to examine temperature-dependent sensitivities. The non-parametric approach 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 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 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 NO_2) relation and figuring out both the partition and transition points. This is the most important objective of the present study.

However, it remains uncertain whether or not the regular $\text{DPO}_3\text{--NO}_x$ (or NO_2) 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 (Huang et al., 2025) revealed that the applicability and reliability for OFR diagnosis differed between the $\text{DPO}_3\text{--NO}_x$ and $\text{DPO}_3\text{--NO}_2$ curves at several observation stations in Hong Kong. Accordingly, the present study also attempts to compare the reliability between the two curves in diagnosing $\text{O}_3\text{--NO}_x\text{--VOC}$ sensitivity across a broader spatial range.”

Specific comment 1-2: Line 53: What is “OFR”? Please define abbreviations when first used.

Response/Action 1-2: Thank you for pointing it out. OFR is short for the term of ozone formation regime. It has been defined in the revised manuscript (Line 65-66).

Specific comment 1-3: Line 54: The authors have published the study they are referring to here (Huang et al., 2025) earlier this month and are now referring to critical limitations of their

work. I find this a bit irritating. Why do the authors have not implemented the improvements in the previous study?

Response/Action 1-3: The primary objective of our previous study (Huang et al., 2025) was to investigate whether or not and how the OFRs shift with temperature in the Greater Bay Area, South China. In this region, the theoretical $\text{DPO}_3\text{--NO}_x$ (or NO_2) curve was firstly convinced regionally prevalent and able to effectively diagnose OFRs' spatial pattern. This encourages us to further verify whether this kind of curve is also globally widespread. In order to do so, we have to spend more time to collect more pollution and meteorological monitoring data worldwide and conduct the data pre-processing. Furthermore, the smoothing span, a 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; therefore, we had spent plenty of time to define the reasonable smoothing spans specific to the individual studied observation stations and the relevant reanalysis data grids in that previous study. In a similar way, we also have to spend sufficient time to firstly define the smoothing spans as reasonable as possible for the stations in the present study, which is for further comparisons of fitting performances between the empirical parametric models and the non-parametric approach (as illustrated in Figures 1-3, Figures S2-S10, and Figures S13).

We do acknowledge that it would have been ideal by incorporating all potential improvements into that previous study. However, the limitations of the previous work (outlined in Line 67-71 of the revised manuscript) did not hinder our ability to understand the temperature-related shift of OFRs within a limited scope, such as the Greater Bay Area. However, in response to those limits, the present study aims to find out a more adaptable method, the empirical parametric modelling, for characterizing the theoretical $\text{DPO}_3\text{--NO}_x$ (or NO_2) curve and diagnosing OFRs.

Specific comment 1-4: Line 55 f.: How do the authors define the NO_x -limited/transition boundary? The transition point is commonly referred to as the crossover from NO_x - to VOC-sensitive chemistry, but there is no exact definition of a transition region in textbook literature. If I read the graphical abstract correctly it is related to the 95th percentile of O_3 production. Where does this definition come from and what's the reasoning for it?

Response/Action 1-4: Thank you for pointing it out. The crossover from NO_x -limited/transition regime to VOC-sensitive regime is the partition point, rather than the transition point. The transition point is the crossover from NO_x -limited regime to transition regime, and it indicates the onset of diminishing ozone production with respect to NO_x . As referred to the study by Yang et al. (2021), we determined the transition point as the NO_x (or NO_2) level corresponding to the position with a half of the maximum curve slope in the present study. **The definition and determination of the two key points were added in the revised manuscript Line (Line 54-59).**

The relation between the 95th percentile of O_3 production and the transition point, detailed as below, was added in the revised manuscript (Line 237-244).

“According to the study by Yang et al. (2021), the transition point for the parametric fitting

curve was defined as the NO_x (or NO_2) concentration corresponding to the position with a half of the maximum fitting curve slope (the blue dotted lines in Figures 3 and S13), after which ozone formation became less dependent on NO_x but significantly more dependent on VOCR. This parametric transition point exactly corresponded to the DPO_3 level in the top 4.9% of the *log-Bragg3* model predictions, so that the transition point for the non-parametric smoothing curve was determined as the NO_x (or NO_2) level corresponding to the top 4.9% smoothing DPO_3 level (the red dotted lines in Figures 3 and S13)."

Specific comment 1-5: Line 56: What do the authors mean by parametric modeling? Is this a reference to parameterizations in atmospheric models or something different?

Response/Action 1-5: The term "parametric modeling" refers to regression with empirical parametric model (Equations 1-7), which is distinct from the parameterization schemes incorporated in atmospheric models. By fitting data with empirical models, we can adaptively obtain the parameters of these models that are specific to the studied locations. The theoretical meanings of the models' parameters were described in Section 2.1. For instance, by fitting data from different locations with the *log-Bragg3* model (Equation 3), we can obtain different sets of fitting values for its three parameters (b , d , e). The fitting values of parameters b and d determine the transition point (Section 3.4) and respectively imply ozone production intensity and the related chemical processes (Section 3.5), and the parameter e corresponds to the partition point (Section 3.4). Therefore, it is possible to conveniently compare the characteristics of ozone formation amongst different locations using the *log-Bragg3* model (Equation 3).

Specific comment 1-6: Line 57: What do the authors mean by environmental stability? Whenever using non-textbook terms, I recommend a full definition and explanation.

Response/Action 1-6: The sentence "The DPO_3 - NO_x (or NO_2) curve shows environmental stability (Guo et al., 2023), enabling the parametric characterization" was rewritten as ".....the basic contour of DPO_3 - NO_x (or 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." This is shown in Line 74-75 of the revised manuscript.

Specific comment 1-7: Line 58: Please elaborate on the "bend" – what is it and where is it coming from? The cited literature Romer et al., 2018 and Guo et al. 2023 do not seem to mention / explain this bend. How can PO_3 have two different values for the same NO_2 ?

Response/Action 1-7: As referred to the Supplementary Information for Guo et al. (2023), the DPO_3 - NO_2 curve exhibited a bend at the end of the curve for some cases, especially for some VOC species (like alkanes in Fig. S5) and when excluding the reaction of $\text{NO} + \text{NO} + \text{O}_2 = 2\text{NO}_2$ in box model (Fig. S6: $\text{C} \rightarrow (\text{C})$), while the DPO_3 - NO_x curve did not show such bending behavior (Figure 2(b) in Romer et al. 2018).

The ends of both curves reflect the relatively low DPO_3/NO_2 ratio and high NO_x level, and this

typically indicates a condition that the reaction of OH with NO₂ dominates the fate of HO_x, slowing the oxidation of organic precursor, and gradually terminating the ozone production (Pusede et al., 2015; Romer et al., 2018). When applying the DPO₃-NO₂ curve, the ozone production might decrease with NO₂ under this condition, potentially leading to a pseudo diagnostic result indicative of a NO_x-limited regime under a realistic NO_x-saturated condition. In contrast, when applying the DPO₃-NO_x curve, the ozone production continues to decline with the increasing NO_x level under this low-DPO₃/NO₂-ratio condition, thereby diagnosed as the VOC-limited regime.

The above explanation was detailed in Section 3.3: Comparison of reliabilities between the DPO₃-NO₂ and DPO₃-NO_x curves (**Line 216-227 in the revised manuscript**).

Specific comment 1-8 Line 77 f.: What exactly are parametric vs non-parametric results?

Response/Action 1-8: The parametric results are the parametric fitting curves (the blue fitting curves in Figure 3, Figure S2-S10, and Figure S13) and their corresponding partition points (the blue dashed dotted vertical lines in Figure 3, Figure S2-S10, and Figure S13), while non-parametric results referred to the non-parametric smoothing curves (the red smoothing curves in Figure 3, Figure S2-S10, and Figure S13) and their corresponding partition points (the red dashed dotted vertical lines in Figure 3, Figure S2-S10, and Figure S13).

For more clarity, the sentence “The parametric model validity was confirmed when its curve and partition point aligned well with the non-parametric results” was rewritten as “The parametric model validity was confirmed when its curve (the blue fitting curve in Figure 3, Figure S2-S10, and Figure S13) and partition point (blue dashed dotted vertical line in Figure 3, Figure S2-S10, and Figure S13) aligned well with those obtained from non-parametric approach (the red smoothing curve in Figure 3, Figure S2-S10, and Figure S13; red dashed dotted vertical line in Figure 3, Figure S2-S10, and Figure S13)” This is shown in **Line 99-102 of the revised manuscript**.

Specific comment 1-9: Line 80 ff.: Did the authors use these equations to fit their data? How were these fits chosen?

Response/Action 1-9: Yes, we use models (Equations 1-7) to fit our data, respectively. The rationale for selecting the studied models, detailed as below, were **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 O₃-HCHO/NO₂ ratio for diagnosing the O₃-NO_x-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 DPO₃-NO_x (or NO₂) 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 DPO₃-NO_x (or NO₂) curve by logarithmizing the NO_x (or NO₂) 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."

Specific comment 1-10: Line 108: What is the study period?

Response/Action 1-10: Thank you for pointing it out. The study period was 2014-2019, which was added in the revised manuscript (Line 136).

Specific comment 1-11: Line 117: What are "records ≤ 4"? Can the authors provide a reasoning for this "no precipitation" definition? I am not aware of being able to accurately infer rainfall from cloud cover.

Response/Action 1-11: Thank you for this question. The explanation, detailed as below, was added in the revised manuscript (Line 144-151).

"The cloud records provided in the NOAA-Integrated Surface Database (ISD), obtained via the R package *worldmet*, range from 0 (representing no visible cloud cover) to 9 (representing a completely overcast sky). The cloud records with values ≤ 4 indicate that < 50% of the sky is obscured by clouds. However, there are limited rainfall recordings in the ISD compared to cloud cover, especially in the US the studied regions in China (except Hong Kong). Based on the meteorological data for the Europe and Hong Kong, where both rainfall and cloud cover recordings are comprehensively available, the precipitation was significantly lower when < 50 % of the sky is covered by clouds, compared to the instances where > 50% of the sky is obscured. Therefore, the "no precipitation" scenario for Europe and US was defined as the hours of 50% cloud cover with the records ≤ 4 in ISD, rather than the zero-precipitation hours."

337 **Specific comment 1-12:** Line 130: The European Union does not describe a geographical
338 region and some parts of the map in Figures S1 are not part of the EU. I recommend referring
339 to the region as, e.g. Europe.

340 **Response/Action 1-12:** Thank you for this kind recommendation. **It was revised throughout**
341 **the manuscript and the updated supplement.**

342 **Specific comment 1-13:** Figure 1: The panels are too small and the resolution too low. It is
343 difficult to read the legend. It is further difficult to distinguish between any of the stations
344 because the data points are overlapping.

345 **Response/Action 1-13:** Thank you for this comment. We have improved the resolution of this
346 figure in the revised manuscript.

347 **Specific comment 1-14:** Line 170 ff.: It is unclear what exactly the authors are trying to show
348 in Figure 1. What is a parametric y-axis and a non-parametric x-axis approach?

349 **Response/Action 1-14:** The y-axis represents the NO_x (or NO₂) concentrations corresponding
350 to the partition points obtained from the parametric models, while the x-axis represents those
351 obtained from the non-parametric approach.

352 **Specific comment 1-15:** Line 174 f.: What are the definitions of the scenarios the authors are
353 referring to?

354 **Response/Action 1-15:** As illustrated in the graphical abstract, taking the red curve specific to
355 the lower VOC reactivity (VOCR1) as the example, the Scenario A is referred to as the curve
356 portion within the yellow dashed box, while the Scenarios B and C corresponding to the curve
357 portion within the green and blue dashed boxes, respectively. The non-parametric fitting can
358 only feature one of the three scenarios based on the realistic data within a theoretically
359 completed curve

360 **Specific comment 1-16:** Line 180 ff.: Is this the bend that the authors were referring to earlier?
361 The reaction of OH + NO₂ is a termination reaction of the HO_x cycle and its dominance
362 characterizes a VOC-limited O₃ formation regime. Unlike the authors state, the cited studies
363 do not show the existence of a pseudo NO_x limited under a NO_x saturated regime. Further
364 evidence would be required to prove this statement of the authors, which does not agree with
365 our current knowledge of O₃ formation sensitivity.

366 **Response/Action 1-16:** Thank you for this comment. We do acknowledge that the reference
367 citations presented here may lead to potential ambiguity in interpretation.

368 The statement “a pseudo diagnostic result indicative of a NO_x-limited regime under a realistic
369 NO_x-saturated condition” is the finding derived from our present study, rather than the
370 conclusions drawn from the cited references of Guo et al., 2023; Romer et al., 2018; Pusede et
371 al., 2015. However, the studies by Guo et al. (2023) and Romer et al. (2018) provided the

evidence regarding the diagnostic uncertainty associated with the application of DPO₃-NO₂ curve; while the studies by Pusede et al. (2015) and Romer et al. (2018) provide a possible explanation for this kind of uncertainty.

More specifically, as referred to the **Supplementary Information for Guo et al. (2023)**, the DPO₃-NO₂ curve exhibited a bend at the end of the curve for some cases, especially for some VOC species (like alkanes in Fig. S5) and when excluding the reaction of NO+NO+O₂=2NO₂ in box model (Fig. S6: C→(C)), while the DPO₃-NO_x curve did not display such bending behavior (Figure 2(b) in Romer et al., 2018). The ends of both curves reflect the relatively low DPO₃/NO₂ ratio and high NO_x level, and this typically indicates a condition that the reaction of OH with NO₂ dominates the fate of HO_x, slowing the oxidation of organic precursor, and gradually terminating the ozone production (Pusede et al., 2015; Romer et al., 2018). When applying the DPO₃-NO₂ curve, the ozone production might decrease with NO₂ under this condition, potentially leading to a pseudo diagnostic result indicative of a NO_x-limited regime under a realistic NO_x-saturated condition. In contrast, when applying the DPO₃-NO_x curve, the ozone production continues to decline with the increasing NO_x level under the low-DPO₃/NO₂-ratio condition, thereby diagnosed as the VOC-limited regime.

Based on the above interpretation, we have re-organized the discussion and citations in the revised manuscript (Line 214-227) as below:

“The DPO₃/NO₂ ratios at these stations in Hong Kong ranged from 0.1 to 0.6, much lower than other stations/grid (BTH: 1.1-4.0, FWP: 1.3-3.4, YRD: 1.4-4.5, PRD: 1.3-4.5, Macao: 2.1, Europe region/US: 0.3-16.5, other stations in Hong Kong: 0.8-6.5). Such low DPO₃/NO₂ ratios, accompanied by high NO_x level, typically occur at the ends of both DPO₃-NO₂ and DPO₃-NO_x curves. As referred to the Figures S5-S6 in Guo et al. (2023), the DPO₃-NO₂ curve was found to exhibit a bend at the its end in certain cases, especially for specific VOC species (like alkanes) and when the reaction of NO+NO+O₂=2NO₂ is excluded in box model, while the DPO₃-NO_x curve did not display such bending behaviour (Romer et al., 2018). A low DPO₃/NO₂ ratio at high NO_x level typically indicates a condition that the reaction of OH with NO₂ dominates the fate of HO_x, slowing the oxidation of organic precursor, and gradually terminating the ozone production (Pusede et al., 2015; Romer et al., 2018). When applying the DPO₃-NO₂ curve, the ozone production might decrease with NO₂ under this condition, potentially leading to a pseudo diagnostic result indicative of a NO_x-limited regime under a realistic NO_x-saturated condition. In contrast, when applying the DPO₃-NO_x curve, the ozone production continues to decline with the increasing NO_x level under this low-DPO₃/NO₂-ratio condition, thereby diagnosed as the VOC-limited regime. Hence, the DPO₃-NO_x curve is considered more reliable for diagnosing O₃-NO_x-VOC sensitivity at any NO_x level, and it is recommended to check the DPO₃/NO₂ ratio before employing the DPO₃-NO₂ curve.”

Specific comment 1-17: Line 193 ff. / Figure 3: What is the added value of these fits? It is possible to determine the dominating sensitivity of O₃ formation based on the observational data of O₃ and NO₂. Why are the fits needed? The individual fit parameters are likely different for each location, as the crossover does not always occur at the same NO₂ mixing ratio

(depending on the availability of VOCs).

Response/Action 1-17: In Figure 3, the blue solid curves represent the parametric fittings based on the *log-Bragg3* model (Equation 3). This model was proved the best to adaptably characterize both the $\text{DPO}_3\text{-NO}_2$ and $\text{DPO}_3\text{-NO}_x$ curves and determine the dominating sensitivity of ozone formation based on the routine recordings of O_3 and NO_2 or O_3 and NO_x .

One of the added values of these fits shown in Figure 3 is that **they make it easier for OFR diagnosis that can be adaptable to different locations and different time**, even though the crossover points does not always occur at the same NO_2 mixing ratio. **This is particularly important for elucidating the evolution of $\text{O}_3\text{-NO}_x\text{-VOC}$ sensitivity on the large spatiotemporal scale.**

Furthermore, as discussed in Section 3.5: Implications of the *log-Bragg 3* model's parameters (*b*, *d*), **the other value** is that the parametric fits in Figure 3 **also provide some implications of ozone formation intensity and the associated chemical processes**, indicated by the parameters *b* and *d* (Table S1).

However, according to the comparison of diagnostic reliability between the $\text{DPO}_3\text{-NO}_2$ and $\text{DPO}_3\text{-NO}_x$ curves (in Section 3.3), the use of $\text{DPO}_3\text{-NO}_2$ curve may introduce significant uncertainty when the DPO_3/NO_2 ratio is excessively low, and it is recommended to evaluate the DPO_3/NO_2 ratio prior to applying this curve. Therefore, before applying the $\text{DPO}_3\text{-NO}_2$ curve for OFR diagnosis on the regional scale (in Section 3.4), we firstly checked the DPO_3/NO_2 ratios, ranging from 0.88 to 4.98 for our studied regions, which were at the median levels compared with those stations of pseudo-diagnosis in Hong Kong (0.1-0.6). Even for the European region with the lowest ratios amongst out studied regions, the $\text{DPO}_3\text{-NO}_2$ curve is still applicable, where the diagnostic results agreed well between the $\text{DPO}_3\text{-NO}_2$ and $\text{DPO}_3\text{-NO}_x$ curves. **In a word, it is conditional to determine the dominating sensitivity of ozone formation based on the observational data of O_3 and NO_2 .**

Specific comment 1-18: Line 217 ff.: How was the *log-Bragg 3* fit chosen? Does it provide the best result? How was this evaluated?

Response/Action 1-18: Yes, the *log-Bragg3* model (Equation 3) provided the best fitting result, compared to other models (Equations 1-2 and 4-7). In the present study, the parametric model validity was confirmed when its curve and partition point aligned well with those obtained from non-parametric approach, which revealed the intrinsic $\text{DPO}_3\text{-NO}_x$ (or NO_2) relation by smoothing a numerical series within local neighborhoods. The studied parametric models were individually applied to regress the $\text{DPO}_3\text{-NO}_x$ (or NO_2) relation for all the studied stations and the Macao grid (494 fits).

Firstly, we identified that the models of *log-Bragg3*, *log-Bragg4*, *log-Lorentz3*, *log-Lorentz4* and *log-Poly2* (Equations 3-7) exhibited **the highest fitting convergence**, with all 494 parametric fits successfully converging. However, not all the convergent fits were able to characterize the regular diagram as in Graphical Abstract to effectively partition the $\text{O}_3\text{-NO}_x$ -

450 VOC sensitivity.

451 Secondly, we further observed that the models of *log-poly2* (Equation 7), *log-Bragg3*
452 (Equation 3) and *log-Lorentz3* (Equation 5) were able to regress the largest number of
453 convergent and effective fits (*log-poly2*: 142/142 DPO₃-NO_x fits, 494/494 DPO₃-NO₂ fits;
454 *log-Bragg3*: 141/142 DPO₃-NO_x fits, 490/494 DPO₃-NO₂ fits; *log-Lorentz3*: 141/142 DPO₃-
455 NO_x fits, 489/494 DPO₃-NO₂ fits).

456 Although all the *log-Poly2* fits (Equation 7) were convergent and effective, quite certain portion
457 of them did not achieve the statistical significance ($p > 0.1$) (Figures S11-S12 (g)). Amongst
458 all models, the *log-Bragg3* (Equation 3) and *log-Lorentz3* (Equation 5) models performed
459 the best fitting significance, with over 95% of fits achieving the statistical significance ($p <$
460 0.1) (Figures S11-S12 (c, e)).

461 Furthermore, we compared the partition points identified between the parametric and non-
462 parametric fits. It also showed that only the *log-Bragg3* (Equation 3) and *log-Lorentz3*
463 (Equation 5) models were able to identify the partition points for all fits under Scenario
464 B as illustrated in Graphical Abstract (Figure 1 (c, e, j, l)).

465 Despite comparable performance in terms of amounts of convergent and effective fits, fitting
466 statistical significance, and ability to identify partition point between the *log-Bragg3* and *log*-
467 *Lorentz3* models, the *log-Bragg3* model is finally preferred due to the generally inferior
468 statistical properties exhibited by *Lorentz* models (Ratkowsky, 1990)

469 Technical:

470 **Specific comment 1-19:** Line 29.: Please check the author of this reference (“Collaborators”).

471 **Response/Action 1-19:** Thank you for pointing it out. This citing was corrected as “[GBD 2019](#)
472 [Risk Factors Collaborators, 2020](#)” (Line 30), and the relevant reference was corrected as “[GBD](#)
473 [2019 Risk Factors Collaborators.: Global burden of 87 risk factors in 204 countries and](#)
474 [territories, 1990-2019: a systematic analysis for the Global Burden of Disease Study 2019,](#)
475 [Lancet, 396, 1223-1249, 10.1016/s0140-6736\(20\)30752-2, 2020.](#)” (Line 365) in the revised
476 manuscript.

477 **Specific comment 1-20:** Line 49 f.: There seems to be a part of the sentence missing “As NO_x
478 increases.”

479 **Response/Action 1-20:** Thank you for pointing it out. This is a repetitive statement, and it was
480 removed in the revised manuscript.

481

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