

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

5 **Reviewer 1**

6 In “Technical Note: Adaptably diagnosing O<sub>3</sub>-NO<sub>x</sub>-VOC sensitivity evolution with routine  
7 pollution and meteorological data” Huang and Liao investigated the sensitivity of O<sub>3</sub> formation  
8 at selected sites in China, the US and Europe by applying different fit equations to the datasets.  
9 The authors identify most of the studied regions to be dominated by VOC-limited O<sub>3</sub> formation  
10 sensitivity.

11 While this is generally an important topic to investigate, unfortunately this study seems  
12 incoherent and is often difficult to follow. It remains largely unclear why and how the suggested  
13 fit equations are applied to the data and even more important what the added value of this  
14 analysis is. The in-situ observations investigated in this study can be used to directly infer the  
15 dominating sensitivity instead of using fit functions. Are any generalized conclusions drawn  
16 from the fitting? Could it be applied to other regions where observations are not available and  
17 how would that be possible considering that crossover points occur at NO<sub>x</sub> to VOC ratios that  
18 are characteristic to each location?

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

20 **As introduced in Section 1: Introduction**, the ozone level has increased in most urban areas  
21 worldwide, and the O<sub>3</sub>-NO<sub>x</sub>-VOC sensitivity has likely evolved in response to the divergent  
22 trends in precursor emissions. **Elucidating its long-term evolution is critical for effective  
23 ozone mitigation.**

24 However, those commonly used methods for ozone formation regime (OFR) diagnosis, such  
25 as the Empirical Kinetic Modelling Approach (EKMA) isopleth plot and chemical indicators  
26 (e.g., H<sub>2</sub>O<sub>2</sub>/HNO<sub>3</sub>, H<sub>2</sub>O<sub>2</sub>/NO<sub>2</sub>, etc.), heavily rely on observation-based or numerical models,  
27 constrained by limited field data and computational demands. They are typically applied in  
28 case studies (Sillman and He, 2002; Sillman and West, 2009; Xue et al., 2014; Ou et al., 2016;  
29 Li et al., 2018). Although the satellite-derived HCHO/NO<sub>2</sub> ratio (FNR)-based method enables  
30 the regional scale long-term O<sub>3</sub>-NO<sub>x</sub>-VOC sensitivity diagnosis (Jin and Holloway, 2015; Ren  
31 et al., 2022; Wang et al., 2021; Zhang et al., 2024), its fixed daily sampling time restricts  
32 insights into other hours, and the sensitivity always varies over time. **These constraints  
33 highlight the necessity for more flexible and adaptable approaches.**

34 **For a specific VOC reactivity (VOCR), the daytime ozone production (DPO<sub>3</sub>) exhibits a  
35 characteristic skewed curve when plotted against NO<sub>x</sub> or NO<sub>2</sub> (Graphical Abstract),  
36 which is transformed from the EKMA plot (Pusede and Cohen, 2012; Romer et al., 2018;  
37 Nussbaumer and Cohen, 2020; Guo et al., 2023; Yang et al., 2021). On a DPO<sub>3</sub>-NO<sub>x</sub> (or NO<sub>2</sub>)  
38 curve (Graphical Abstract), the partition point and transition point are two key NO<sub>x</sub> (or**

39 **NO<sub>2</sub>** levels for differentiating the O<sub>3</sub>-NO<sub>x</sub>-VOC sensitivity. The **partition point** is defined  
40 as the peak DPO<sub>3</sub> corresponding NO<sub>x</sub> (or NO<sub>2</sub>) level, where the ozone formation equals to  
41 consumption, distinguishing the NO<sub>x</sub>-limited/transition regime (to the left) and the VOC-  
42 limited regime (to the right); the **transition point** is defined as the NO<sub>x</sub> (or NO<sub>2</sub>) level at the  
43 position indicating the onset of diminishing ozone production with respect to NO<sub>x</sub>, which  
44 further differentiates the NO<sub>x</sub>-limited and transition regimes. As referred to the study by Yang  
45 et al. (2021), we determined the transition point as the position with a half of the maximum  
46 curve slope.

47 **Although the in-situ observations investigated in this study can be used to directly infer**  
48 **the dominating sensitivity via non-parametric approach** (Huang et al., 2025), **two**  
49 **limitations still persist about that method:** (1) a fixed smoothing span, the key configuration  
50 for non-parametric smoothing, failed to exhibit robustness in fitting performance across studied  
51 sites, which leads to uncertainty in determining the partition point and inhibits the adaptability  
52 of this method across a broader spatiotemporal range; (2) the non-parametric approach  
53 provided no information on the curve's height and width, which determine the transition point  
54 and vary with locations, study periods and environmental factors (e.g., temperature, VOCs,  
55 etc.). **The basic contour of the regular DPO<sub>3</sub>-NO<sub>x</sub> (or NO<sub>2</sub>) curve would not vary with the**  
56 **relative humidity, temperature, season, altitude, mixing layer height and VOC species**  
57 **(Guo et al., 2023). This environmental stability makes it possible to be parametrically**  
58 **characterized. Therefore, seeking an effective empirical parametric model is necessary**  
59 **for more adaptably characterizing the DPO<sub>3</sub>-NO<sub>x</sub> (or NO<sub>2</sub>) relation and figuring out both**  
60 **the partition and transition points.** This is the most important objective of the present study.

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

62 **As introduced in Section 2: Methodology**, the DPO<sub>3</sub>-NO<sub>x</sub> (or NO<sub>2</sub>) relation was regressed  
63 with the five-percentile-binned NO<sub>x</sub> (or NO<sub>2</sub>) concentrations (or logarithms) and their  
64 corresponding average DPO<sub>3</sub> levels. The DPO<sub>3</sub> was defined as the difference between the  
65 MDA8-daytime (7:00-19:00 Local Time (LT)) hourly ozone concentration and the ozone  
66 concentration at 6:00 LT. A total of seven parametric models (Equations 1-7) were individually  
67 applied to characterize the DPO<sub>3</sub>-NO<sub>x</sub> (or NO<sub>2</sub>) relation. As in the bellowing **Response/Action**  
68 **1-9** to Specific comment 1-9, we gave a more detail introduction of the rationale for selecting  
69 these studied models, which was added in the updated Supplement of the revised manuscript  
70 (Text S1).

71 3. What is the **added value** of this analysis?

72 In order to address the limitations of those commonly used diagnostic methods (**Line 39-46 in**  
73 **revised manuscript**) and the prior non-parametric fitting of DPO<sub>3</sub>-NO<sub>x</sub> (or NO<sub>2</sub>) curve (**Line**  
74 **62-77 in the revised manuscript**), the present study aims to seek an effective empirical  
75 parametric model for more adaptably characterizing the DPO<sub>3</sub>-NO<sub>x</sub> (or NO<sub>2</sub>) relation and  
76 determining the dominating sensitivity of ozone formation. After a series of analyses as in  
77 Section 3.2: Which is the most capable parametric model, we identified that the *log-Bragg3*

78 model (Equation 3) performed the best.

79 Therefore, one of the added values of these analyses is that they make it easier for OFR  
80 diagnosis that can be adaptable to different locations and different time, even though the  
81 crossover points do not always occur at the same NO<sub>2</sub> mixing ratio. This is particularly  
82 important for elucidating the evolution of O<sub>3</sub>-NO<sub>x</sub>-VOC sensitivity on the large  
83 spatiotemporal scale. Furthermore, as discussed in Section 3.5: Implications of the *log-Bragg*  
84 3 model's parameters (*b*, *d*), the identified model (*log-Bragg3* model, Equation 3) is also  
85 able to provide implications of ozone formation intensity and the associated chemical  
86 processes, indicating by its parameters.

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

88 Based on the above responses, the generalized conclusion from the fitting is that the *log-Bragg3*  
89 model (Equation 3) performed the best, compared with other models, in adaptably  
90 characterizing the DPO<sub>3</sub>-NO<sub>x</sub> (or NO<sub>2</sub>) relation and diagnosing the dominating sensitivity of  
91 ozone formation, which is also able to provide the implications of ozone formation intensity  
92 and the associated chemical processes.

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

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

96 6. How would that be possible considering that crossover points occur at NO<sub>x</sub> to VOC ratios  
97 that are characteristic to each location?

98 Yes, the crossover points are characteristic to different locations, and they theoretically depend  
99 on local condition (e.g., VOCs or other relevant pollutants/radicals, meteorological factors,  
100 etc.). However, our identified model can solve this problem by adaptably fitting the data at  
101 different locations. This is based on a hypothesis that the daytime ozone production (DPO<sub>3</sub>)  
102 exhibits a characteristic skewed curve when plotted against NO<sub>x</sub> or NO<sub>2</sub> for a specific  
103 VOC reactivity (VOCR) (Line 47-48 in the revised manuscript). And indeed, as in Section  
104 3.1, the above hypothesized DPO<sub>3</sub>-NO<sub>x</sub> (or NO<sub>2</sub>) relation was empirically validated  
105 worldwide, even in regions with severe PM<sub>2.5</sub> contamination, where the ozone formation is  
106 additionally influenced by the aerosol-inhibited photochemical regime, such as BTH, FWP and  
107 YRD in China (Ivatt et al., 2022; Geng et al., 2021; Kong et al., 2021; Xiao et al., 2022).

108 From this perspective, the parameters in the identified model (*log-Bragg3*, Equation 3)  
109 reflect the local condition to some extent. For example, the fitting parameter *e* varied with  
110 regions, as listed in Table S1 and Table S2; higher value indicates higher partition point (or  
111 crossover point as referred by the reviewer). Furthermore, as discussed in Section 3.5, the  
112 parameter *d* represents the maximum DPO<sub>3</sub> level, exhibiting higher ozone production with  
113 higher value; and higher value of parameter *b* characterizes a steeper curve, indicating a  
114 condition that favors faster change in ozone production efficiency for a given increment of NO<sub>x</sub>.

115 It is further concerning that the authors have published a paper earlier this month (Huang et al.,  
116 2025), which they are now referring to have “critical limitations” which “fail” in respect to two  
117 different aspects (Line 54f.). This makes me wonder why the authors have not previously fixed  
118 these issues, considering that this previous paper was published one month after the submission  
119 of this manuscript.

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

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

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

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

133 “On a theoretically regular  $DPO_3$ - $NO_x$  (or  $NO_2$ ) curve (Graphical Abstract), the partition point  
134 and transition point are two key  $NO_x$  (or  $NO_2$ ) levels for differentiating the  $O_3$ - $NO_x$ -VOC  
135 sensitivity. The partition point is defined as the peak  $DPO_3$  corresponding  $NO_x$  (or  $NO_2$ ) level,  
136 where the ozone formation equals to consumption, distinguishing the  $NO_x$ -limited/transition  
137 regime (to the left) and the VOC-limited regime (to the right); the transition point is defined as  
138 the  $NO_x$  (or  $NO_2$ ) level at the position indicating the onset of diminishing ozone production  
139 with respect to  $NO_x$ , which further differentiates the  $NO_x$ -limited and transition regimes. As  
140 referred to the study by Yang et al. (2021), we determined the transition point as the position  
141 with a half of the maximum curve slope in the present study.”

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

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

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

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

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

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

155 “In our previous study (Huang et al., 2025), the  $DPO_3-NO_x$  (or  $NO_2$ ) relation, as depicted in  
156 the Scenario A, B or C within a completed skewed curve in Graphical Abstract, was proved  
157 widespread based on the routine monitoring data in the Greater Bay Area, South China, which  
158 was smoothed by a non-parametric regression technique. The smoothing curve was able to  
159 effectively characterize the regional spatial pattern of  $O_3-NO_x$ -VOC sensitivity, differentiating  
160 the ozone formation regimes (OFRs) into the  $NO_x$ -limited/transition regime and the VOC-  
161 limited regime, and was further utilized to examine temperature-dependent sensitivities. The  
162 non-parametric approach is a commonly used method for smoothing a fluctuating numerical  
163 series within local neighbourhoods, enabling the identification of intrinsic  $DPO_3-NO_x$  (or  $NO_2$ )  
164 relation. However, two limitations still persist: (1) a fixed smoothing span, the key  
165 configuration for non-parametric smoothing, failed to exhibit robustness in fitting performance  
166 across studied sites, which leads to uncertainty in determining the partition point and inhibits  
167 the adaptability of this method across a broader spatiotemporal range; (2) the non-parametric  
168 approach provided no information on the curve’s height and width, which determine the  
169 transition point and vary with locations, study periods and environmental factors (e.g.,  
170 temperature, VOCs, etc.). As studied by Guo et al. (2023), the basic contour of the regular  
171  $DPO_3-NO_x$  (or  $NO_2$ ) curve would not vary with the relative humidity, temperature, season,  
172 altitude, mixing layer height and VOC species. This environmental stability makes it possible  
173 to be parametrically characterized. Therefore, seeking an effective empirical parametric model  
174 is necessary for more adaptably characterizing the  $DPO_3-NO_x$  (or  $NO_2$ ) relation and figuring  
175 out both the partition and transition points. This is the most important objective of the present  
176 study.”

177 However, it remains uncertain whether or not the regular  $DPO_3-NO_x$  (or  $NO_2$ ) relation is  
178 globally prevalent. Therefore, it is essential to firstly verify the universality of this relation  
179 using data from routine monitoring networks worldwide. Furthermore, based on the non-  
180 parametric approach, our previous study (Huang et al., 2025) revealed that the applicability  
181 and reliability for OFR diagnosis differed between the  $DPO_3-NO_x$  and  $DPO_3-NO_2$  curves at  
182 several observation stations in Hong Kong. Accordingly, the present study also attempts to  
183 compare the reliability between the two curves in diagnosing  $O_3-NO_x$ -VOC sensitivity across  
184 a broader spatial range.”

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

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

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

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

192 **Response/Action 1-3:** The primary objective of our previous study (Huang et al., 2025) was  
193 to investigate whether or not and how the OFRs shift with temperature in the Greater Bay Area,  
194 South China. In this region, the theoretical  $DPO_3-NO_x$  (or  $NO_2$ ) curve was firstly convinced  
195 regionally prevalent and able to effectively diagnose OFRs' spatial pattern. This encourages us  
196 to further verify whether this kind of curve is also globally widespread. In order to do so, we  
197 have to spend more time to collect more pollution and meteorological monitoring data  
198 worldwide and conduct the data pre-processing. Furthermore, the smoothing span, a key  
199 configuration for non-parametric smoothing, failed to exhibit robustness in fitting performance  
200 across studied sites, which leads to uncertainty in determining the partition point; therefore, we  
201 had spent plenty of time to define the reasonable smoothing spans specific to the individual  
202 studied observation stations and the relevant reanalysis data grids in that previous study. In a  
203 similar way, we also have to spend sufficient time to firstly define the smoothing spans as  
204 reasonable as possible for the stations in the present study, which is for further comparisons of  
205 fitting performances between the empirical parametric models and the non-parametric  
206 approach (as illustrated in Figures 1-3, Figures S2-S10, and Figures S13).

207 We do acknowledge that it would have been ideal by incorporating all potential improvements  
208 into that previous study. However, the limitations of the previous work (outlined in Line 67-71  
209 of the revised manuscript) did not hinder our ability to understand the temperature-related shift  
210 of OFRs within a limited scope, such as the Greater Bay Area. However, in response to those  
211 limits, the present study aims to find out a more adaptable method, the empirical parametric  
212 modelling, for characterizing the theoretical  $DPO_3-NO_x$  (or  $NO_2$ ) curve and diagnosing OFRs.

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

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

226 **The relation between the 95<sup>th</sup> percentile of  $O_3$  production and the transition point,  
227 detailed as below, was added in the revised manuscript (Line 237-244).**

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

229 curve was defined as the  $\text{NO}_x$  (or  $\text{NO}_2$ ) concentration corresponding to the position with a half  
230 of the maximum fitting curve slope (the blue dotted lines in Figures 3 and S13), after which  
231 ozone formation became less dependent on  $\text{NO}_x$  but significantly more dependent on VOCR.  
232 This parametric transition point exactly corresponded to the  $\text{DPO}_3$  level in the top 4.9% of the  
233 *log-Bragg3* model predictions, so that the transition point for the non-parametric smoothing  
234 curve was determined as the  $\text{NO}_x$  (or  $\text{NO}_2$ ) level corresponding to the top 4.9% smoothing  
235  $\text{DPO}_3$  level (the red dotted lines in Figures 3 and S13)."

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

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

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

252 **Response/Action 1-6:** The sentence "The  $\text{DPO}_3$ - $\text{NO}_x$  (or  $\text{NO}_2$ ) curve shows environmental  
253 stability (Guo et al., 2023), enabling the parametric characterization" was rewritten as ".....the  
254 basic contour of  $\text{DPO}_3$ - $\text{NO}_x$  (or  $\text{NO}_2$ ) curve would not vary with the relative humidity,  
255 temperature, season, altitude, mixing layer height and VOC species. This environmental  
256 stability makes it possible to be parametrically characterized." **This is shown in Line 74-75**  
257 **of the revised manuscript.**

258 **Specific comment 1-7:** Line 58: Please elaborate on the "bend" – what is it and where is it  
259 coming from? The cited literature Romer et al., 2018 and Guo et al. 2023 do not seem to  
260 mention / explain this bend. How can  $\text{PO}_3$  have two different values for the same  $\text{NO}_2$ ?

261 **Response/Action 1-7:** As referred to the Supplementary Information for Guo et al. (2023), the  
262  $\text{DPO}_3$ - $\text{NO}_2$  curve exhibited a bend at the end of the curve for some cases, especially for some  
263 VOC species (like alkanes in Fig. S5) and when excluding the reaction of  $\text{NO}+\text{NO}+\text{O}_2=2\text{NO}_2$   
264 in box model (Fig. S6:  $\text{C}\rightarrow(\text{C})$ ), while the  $\text{DPO}_3$ - $\text{NO}_x$  curve did not show such bending  
265 behavior (Figure 2(b) in Romer et al. 2018).

266 The ends of both curves reflect the relatively low  $\text{DPO}_3/\text{NO}_2$  ratio and high  $\text{NO}_x$  level, and this

267 typically indicates a condition that the reaction of OH with NO<sub>2</sub> dominates the fate of HO<sub>x</sub>,  
268 slowing the oxidation of organic precursor, and gradually terminating the ozone production  
269 (Pusede et al., 2015; Romer et al., 2018). When applying the DPO<sub>3</sub>-NO<sub>2</sub> curve, the ozone  
270 production might decrease with NO<sub>2</sub> under this condition, potentially leading to a pseudo  
271 diagnostic result indicative of a NO<sub>x</sub>-limited regime under a realistic NO<sub>x</sub>-saturated condition.  
272 In contrast, when applying the DPO<sub>3</sub>-NO<sub>x</sub> curve, the ozone production continues to decline  
273 with the increasing NO<sub>x</sub> level under this low-DPO<sub>3</sub>/NO<sub>2</sub>-ratio condition, thereby diagnosed as  
274 the VOC-limited regime.

275 The above explanation was detailed in Section 3.3: Comparison of reliabilities between the  
276 DPO<sub>3</sub>-NO<sub>2</sub> and DPO<sub>3</sub>-NO<sub>x</sub> curves (Line 216-227 in the revised manuscript).

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

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

284 For more clarity, the sentence “The parametric model validity was confirmed when its curve  
285 and partition point aligned well with the non-parametric results” was rewritten as “The  
286 parametric model validity was confirmed when its curve (the blue fitting curve in Figure 3,  
287 Figure S2-S10, and Figure S13) and partition point (blue dashed dotted vertical line in Figure  
288 3, Figure S2-S10, and Figure S13) aligned well with those obtained from non-parametric  
289 approach (the red smoothing curve in Figure 3, Figure S2-S10, and Figure S13; red dashed  
290 dotted vertical line in Figure 3, Figure S2-S10, and Figure S13)” This is shown in **Line 99-102**  
291 **of the revised manuscript.**

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

294 **Response/Action 1-9:** Yes, we use models (Equations 1-7) to fit our data, respectively. The  
295 rationale for selecting the studied models, detailed as below, were **added in the updated**  
296 **supplement (Text S1).**

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

305 
$$Y = d \times \exp[-b \times (X - e)^2] \quad (S1)$$

306 
$$Y = c + (d - c) \times \exp[-b \times (X - e)^2] \quad (S2)$$

307 
$$Y = \frac{d}{1+b \times (X - e)} \quad (S3)$$

308 
$$Y = c + \frac{d - c}{1+b \times (X - e)} \quad (S4)$$

309 The Poly2 model (Equation S5) provided as below can also be used to describe a symmetric  
 310 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>-  
 311 NO<sub>x</sub>-VOC sensitivity (Jin et al., 2020).

312 
$$Y = b_0 + b_1 \times X + b_2 \times X^2 \quad (A5)$$

313 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  
 314 asymmetric curve with a maximum, and thus can be appropriately described by Equation 1. To  
 315 explore more alternative fitting approaches, we attempted to reduce the skewness of the DPO<sub>3</sub>-  
 316 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  
 317 Equations 2-7 in the article text are the transformed forms of the Equation 1 and Equations S1-  
 318 S5 with the X-coordinate logarithmized.”

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

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

322 **Specific comment 1-11:** Line 117: What are “records  $\leq 4$ ”? Can the authors provide a  
 323 reasoning for this “no precipitation” definition? I am not aware of being able to accurately infer  
 324 rainfall from cloud cover.

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

327 “The cloud records provided in the NOAA-Integrated Surface Database (ISD), obtained via  
 328 the R package *worldmet*, range from 0 (representing no visible cloud cover) to 9 (representing  
 329 a completely overcast sky). The cloud records with values  $\leq 4$  indicate that < 50% of the sky  
 330 is obscured by clouds. However, there are limited rainfall recordings in the ISD compared to  
 331 cloud cover, especially in the US the studied regions in China (except Hong Kong). Based on  
 332 the meteorological data for the Europe and Hong Kong, where both rainfall and cloud cover  
 333 recordings are comprehensively available, the precipitation was significantly lower when < 50 %  
 334 of the sky is covered by clouds, compared to the instances where > 50% of the sky is obscured.  
 335 Therefore, the “no precipitation” scenario for Europe and US was defined as the hours of 50%  
 336 cloud cover with the records  $\leq 4$  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<sub>x</sub> (or NO<sub>2</sub>) 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 + NO<sub>2</sub> is a termination reaction of the HO<sub>x</sub> cycle and its dominance  
362 characterizes a VOC-limited O<sub>3</sub> formation regime. Unlike the authors state, the cited studies  
363 do not show the existence of a pseudo NO<sub>x</sub> limited under a NO<sub>x</sub> 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 O<sub>3</sub> 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<sub>x</sub>-limited regime under a realistic  
369 NO<sub>x</sub>-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

372 evidence regarding the diagnostic uncertainty associated with the application of DPO<sub>3</sub>-NO<sub>2</sub>  
373 curve; while the studies by Pusede et al. (2015) and Romer et al. (2018) provide a possible  
374 explanation for this kind of uncertainty.

375 More specifically, as referred to the **Supplementary Information for Guo et al. (2023)**, the  
376 DPO<sub>3</sub>-NO<sub>2</sub> curve exhibited a bend at the end of the curve for some cases, especially for some  
377 VOC species (like alkanes in Fig. S5) and when excluding the reaction of NO+NO+O<sub>2</sub>=2NO<sub>2</sub>  
378 in box model (Fig. S6: C→(C)), while the DPO<sub>3</sub>-NO<sub>x</sub> curve did not display such bending  
379 behavior (Figure 2(b) in Romer et al., 2018). The ends of both curves reflect the relatively  
380 low DPO<sub>3</sub>/NO<sub>2</sub> ratio and high NO<sub>x</sub> level, and this typically indicates a condition that the  
381 reaction of OH with NO<sub>2</sub> dominates the fate of HO<sub>x</sub>, slowing the oxidation of organic precursor,  
382 and gradually terminating the ozone production (Pusede et al., 2015; Romer et al., 2018). When  
383 applying the DPO<sub>3</sub>-NO<sub>2</sub> curve, the ozone production might decrease with NO<sub>2</sub> under this  
384 condition, potentially leading to a pseudo diagnostic result indicative of a NO<sub>x</sub>-limited regime  
385 under a realistic NO<sub>x</sub>-saturated condition. In contrast, when applying the DPO<sub>3</sub>-NO<sub>x</sub> curve, the  
386 ozone production continues to decline with the increasing NO<sub>x</sub> level under the low-DPO<sub>3</sub>/NO<sub>2</sub>-  
387 ratio condition, thereby diagnosed as the VOC-limited regime.

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

390 “The DPO<sub>3</sub>/NO<sub>2</sub> ratios at these stations in Hong Kong ranged from 0.1 to 0.6, much lower than  
391 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,  
392 Europe region/US: 0.3-16.5, other stations in Hong Kong: 0.8-6.5). Such low DPO<sub>3</sub>/NO<sub>2</sub> ratios,  
393 accompanied by high NO<sub>x</sub> level, typically occur at the ends of both DPO<sub>3</sub>-NO<sub>2</sub> and DPO<sub>3</sub>-NO<sub>x</sub>  
394 curves. As referred to the Figures S5-S6 in Guo et al. (2023), the DPO<sub>3</sub>-NO<sub>2</sub> curve was found  
395 to exhibit a bend at the its end in certain cases, especially for specific VOC species (like alkanes)  
396 and when the reaction of NO+NO+O<sub>2</sub>=2NO<sub>2</sub> is excluded in box model, while the DPO<sub>3</sub>-NO<sub>x</sub>  
397 curve did not display such bending behaviour (Romer et al., 2018). A low DPO<sub>3</sub>/NO<sub>2</sub> ratio at  
398 high NO<sub>x</sub> level typically indicates a condition that the reaction of OH with NO<sub>2</sub> dominates the  
399 fate of HO<sub>x</sub>, slowing the oxidation of organic precursor, and gradually terminating the ozone  
400 production (Pusede et al., 2015; Romer et al., 2018). When applying the DPO<sub>3</sub>-NO<sub>2</sub> curve, the  
401 ozone production might decrease with NO<sub>2</sub> under this condition, potentially leading to a pseudo  
402 diagnostic result indicative of a NO<sub>x</sub>-limited regime under a realistic NO<sub>x</sub>-saturated condition.  
403 In contrast, when applying the DPO<sub>3</sub>-NO<sub>x</sub> curve, the ozone production continues to decline  
404 with the increasing NO<sub>x</sub> level under this low-DPO<sub>3</sub>/NO<sub>2</sub>-ratio condition, thereby diagnosed as  
405 the VOC-limited regime. Hence, the DPO<sub>3</sub>-NO<sub>x</sub> curve is considered more reliable for  
406 diagnosing O<sub>3</sub>-NO<sub>x</sub>-VOC sensitivity at any NO<sub>x</sub> level, and it is recommended to check the  
407 DPO<sub>3</sub>/NO<sub>2</sub> ratio before employing the DPO<sub>3</sub>-NO<sub>2</sub> curve. ”

408 **Specific comment 1-17:** Line 193 ff. / Figure 3: What is the added value of these fits? It is  
409 possible to determine the dominating sensitivity of O<sub>3</sub> formation based on the observational  
410 data of O<sub>3</sub> and NO<sub>2</sub>. Why are the fits needed? The individual fit parameters are likely different  
411 for each location, as the crossover does not always occur at the same NO<sub>2</sub> mixing ratio

412 (depending on the availability of VOCs).

413 **Response/Action 1-17:** In Figure 3, the blue solid curves represent the parametric fittings  
414 based on the *log-Bragg3* model (Equation 3). This model was proved the best to adaptably  
415 characterize both the DPO<sub>3</sub>-NO<sub>2</sub> and DPO<sub>3</sub>-NO<sub>x</sub> curves and determine the dominating  
416 sensitivity of ozone formation based on the routine recordings of O<sub>3</sub> and NO<sub>2</sub> or O<sub>3</sub> and NO<sub>x</sub>.

417 **One of the added values** of these fits shown in Figure 3 is that **they make it easier for OFR**  
418 **diagnosis that can be adaptable to different locations and different time**, even though the  
419 crossover points does not always occur at the same NO<sub>2</sub> mixing ratio. **This is particularly**  
420 **important for elucidating the evolution of O<sub>3</sub>-NO<sub>x</sub>-VOC sensitivity on the large**  
421 **spatiotemporal scale.**

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

426 However, according to the comparison of diagnostic reliability between the DPO<sub>3</sub>-NO<sub>2</sub> and  
427 DPO<sub>3</sub>-NO<sub>x</sub> curves (in Section 3.3), the use of DPO<sub>3</sub>-NO<sub>2</sub> curve may introduce significant  
428 uncertainty when the DPO<sub>3</sub>/NO<sub>2</sub> ratio is excessively low, and it is recommended to evaluate  
429 the DPO<sub>3</sub>/NO<sub>2</sub> ratio prior to applying this curve. Therefore, before applying the DPO<sub>3</sub>-NO<sub>2</sub>  
430 curve for OFR diagnosis on the regional scale (in Section 3.4), we firstly checked the  
431 DPO<sub>3</sub>/NO<sub>2</sub> ratios, ranging from 0.88 to 4.98 for our studied regions, which were at the median  
432 levels compared with those stations of pseudo-diagnosis in Hong Kong (0.1-0.6). Even for the  
433 European region with the lowest ratios amongst out studied regions, the DPO<sub>3</sub>-NO<sub>2</sub> curve is  
434 still applicable, where the diagnostic results agreed well between the DPO<sub>3</sub>-NO<sub>2</sub> and DPO<sub>3</sub>-  
435 NO<sub>x</sub> curves. **In a word, it is conditional to determine the dominating sensitivity of ozone**  
436 **formation based on the observational data of O<sub>3</sub> and NO<sub>2</sub>.**

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

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

446 Firstly, we identified that the models of *log-Bragg3*, *log-Bragg4*, *log-Lorentz3*, *log-Lorentz4*  
447 and *log-Poly2* (Equations 3-7) exhibited **the highest fitting convergence**, with all 494  
448 parametric fits successfully converging. However, not all the convergent fits were able to  
449 characterize the regular diagram as in Graphical Abstract to effectively partition the O<sub>3</sub>-NO<sub>x</sub>-

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 DPO<sub>3</sub>-NO<sub>x</sub> fits, 494/494 DPO<sub>3</sub>-NO<sub>2</sub> fits;  
454 *log-Bragg3*: 141/142 DPO<sub>3</sub>-NO<sub>x</sub> fits, 490/494 DPO<sub>3</sub>-NO<sub>2</sub> fits; *log-Lorentz3*: 141/142 DPO<sub>3</sub>-  
455 NO<sub>x</sub> fits, 489/494 DPO<sub>3</sub>-NO<sub>2</sub> 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<sub>x</sub>  
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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