

We thank the reviewer for the comments that improve the quality of the paper. The detailed responses are given as follows. The reviewers' comments are shown in italic font, the responses are in regular blue font, and the revised text is in blue bold font.

Response to Referee #2

This paper presented the examination of different indicators for O₃-NO_x-VOC sensitivity based on the chemical transport model CMAQ results. Four indicators were tested, i.e. the ratio of the production rates of H₂O₂ and HNO₃ (PH₂O₂/PHNO₃), HCHO/NO₂, HCHO/NO_y, and reactive nitrogen concentrations (NO_y) for the YRD region. This work determined and evaluated the threshold values of these indicators. Besides, the uncertainty caused by the method was also analyzed. Generally, the manuscript is well-written with a clear structure, and the analysis and discussion are scientifically sound. I recommend publication once the comments below have been addressed.

General comments:

Comment 1: *The determination of NO_x-limited and VOC-limited is changes of O₃ by more than 5 ppbv if NO_x and VOC emission reduction by 35% relative to the base run. This criterion is adopted from Sillman et al. (1998). However, the original analysis mainly focused on an ozone episode at the Nashville and vicinity area with relatively high O₃ concentration (>80 ppbv). As indicate in Figure 2(a), the O₃ concentration could match this criterion for the south part of YRD but not the North part. The relatively low O₃ in the north part leads to relative low absolute change of O₃ concentration when NO_x or VOC emission reduce by 35%. In this case, the north part can still be attributed to NO_x/VOC limited regime. It's not clear to me how the determination of threshold for different indicators depends on this classification.*

Response 1: We agree that the northern YRD can be attributed to NO_x or VOC limited regimes, but not with O₃ changes of >5ppb when perturbing NO_x or VOC emissions by 35%. As a result, many grids in the northern YRD were excluded when determining the thresholds of indicators (see Figure 2(d)), as the criteria ($\Delta O_3 \geq 5\text{ppb}$) are relatively high for these low-O₃ areas. It could induce uncertainties when applying the derived thresholds (mostly based on O₃ changes in the

polluted areas) to determine O₃ formation regimes in clean areas. Therefore, we adopted relative changes of O₃ instead of the absolute changes as the criteria in Section 3.4 (see Figure 6 (b)). The results show that the thresholds with O₃ changes $\geq 5\%$, corresponding to 2-3 ppb or larger in the northern YRD (with O₃ concentrations mostly in the range of 40-60 ppb), are comparable to the original values, while the relative change of 2% is too small to distinguish the NO_x-VOC transition. The lower limits of the thresholds were more affected due to fewer VOC-limited grids in the YRD. In general, the thresholds derived from the grids with O₃ changes of >5ppb are also appropriate for the northern YRD to determine O₃ formation regimes.

Comment 2: *In section 3.4, the $P_{H_2O_2}/P_{HNO_3}$ is used for an example but also suggest to add similar discussion on HCHO/NO₂ or HCHO/NO_y to address the statement that “By comparing with the O₃ isopleths, it was found that HCHO/NO₂ and HCHO/NO_y showed the most consistency”.*

Response 2: Thank you for your suggestion. We added Figures S6-S8 as the results of HCHO/NO₂, HCHO/NO_y, and NO_y in supplemental materials, which were referred to in Lines 360-362:

The results of HCHO/NO₂, HCHO/NO_y, and NO_y (Figures S6-S8) were similar. They all indicated that the thresholds of the photochemical indicators are dependent on the methods or parameters in the methodology.

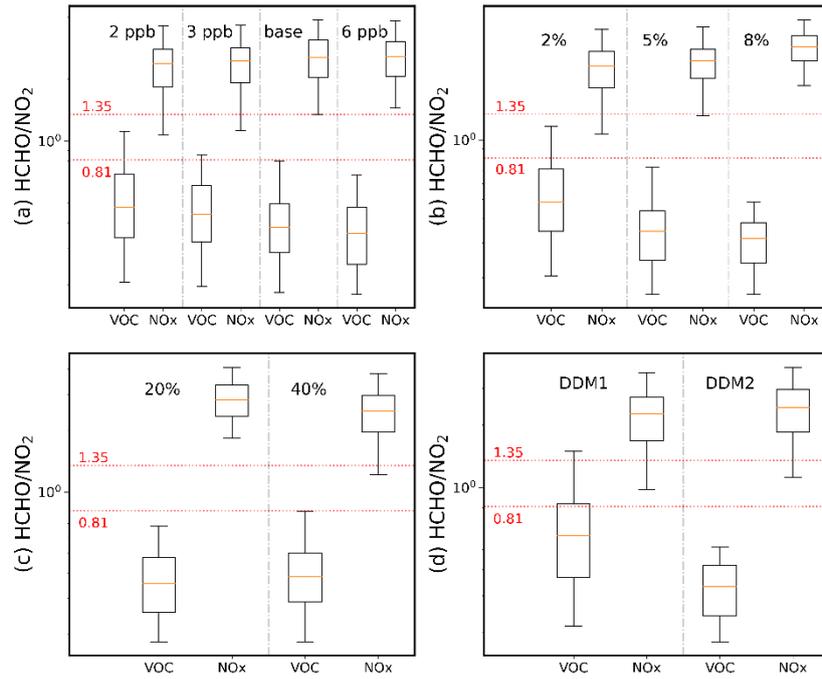


Figure S6: The percentile distributions of the HCHO/NO₂ values at the VOC- or NO_x-limited grid cells with different setups in methodology. The threshold intervals of HCHO/NO₂ in Table 3 are indicated with red dotted lines.

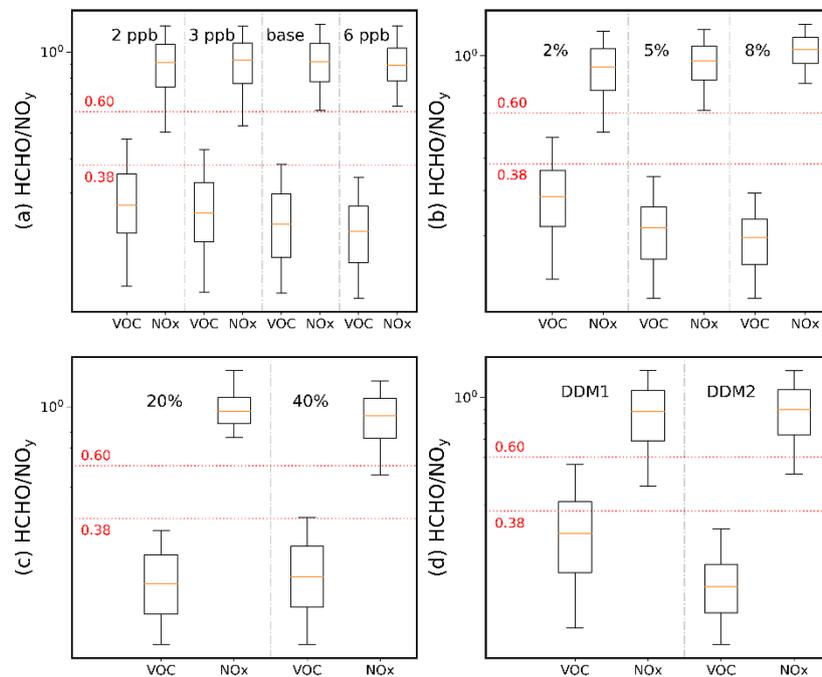


Figure S7: The percentile distributions of the HCHO/NO_y values at the VOC- or NO_x-limited grid cells with different setups in methodology. The threshold intervals of HCHO/NO_y in Table 3 are indicated with red dotted lines.

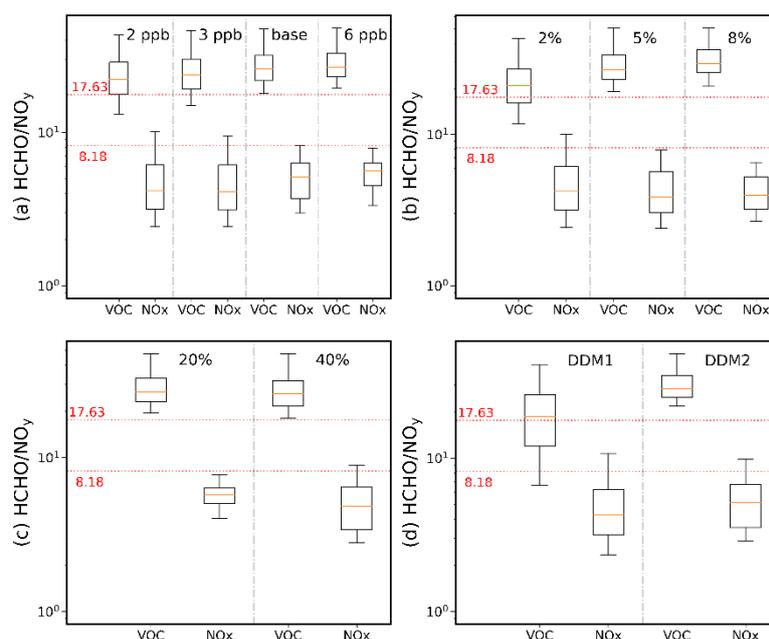


Figure S8: The percentile distributions of the NO_y values at the VOC- or NO_x -limited grid cells with different setups in methodology. The threshold intervals of NO_y in Table 3 are indicated with red dotted lines.

Comment 3: *The overall accuracy values of NO_y in some cases are higher than other photochemical indicators as shown in Table.4, however in the section 3.3, the indicator was not recommended. Please explain the discrepancy between the result mentioned above.*

Response 3: Table 4 and Section 3.3 show the evaluation of the indicators from two aspects. Table 4 shows the accuracy of the photochemical indicators with the base emissions in the YRD. However, in Section 3.3, with significant emission changes, NO_y is substantially inconsistent with O_3 isopleths, particularly with VOC emission reductions. The former is based on statistics throughout the entire YRD in one emission scenario, while the latter emphasizes the comparison with O_3 isopleths with emission perturbations in one location.

Specific comments:

Comment 4: *The language needs improved. For example, the tense of one paragraph should be consistent. I only list a few and suggest the authors to carefully go through the paper. Line 14: examines to examined, Line 49: is VOC-limited to was VOC-limited...*

Response 4: Thank you for your comment. We have checked the tense and made the following revisions:

Line 14: This study examined ...

Line 49: ... was VOC-limited in the morning ...

Line 71: this work aimed to ...

Line 112: Figure S2 shows that ...

Line 151: ...were not classified...were lower than...

Line 161: This provided ...

Line 168: were indicated with ...

Comment 5: *Line 48: Explain “NO_z” when it appeared for the first time.*

Response 5: Thanks for pointing this out. NO_z has been defined at the first mention:

Line 48: “...presented hourly H₂O₂/NO_z (NO_z = NO_y - NO_x) ...”

Comment 6: *Line 50: If the threshold for an indicator is varying, it seems contradict to “robust”,*

Response 6: Thank you for your comment. In each of the listed studies, the indicator H₂O₂/HNO₃ can well distinguish O₃ formation regimes, and even shows better performance than other indicators (Xie et al., 2014; Ye et al., 2016; Peng et al., 2011). The thresholds in each study are different. As such, H₂O₂/HNO₃ can be regarded as a reliable indicator to identify O₃ sensitivity with localized thresholds, although the thresholds could vary among regions.

Comment 7: *Please Briefly introduce HDDM in the section of “Methods”.*

Response 7: Thank you for your comment. We have added the introduction of the HDDM as below:

Lines 182-183: “The HDDM can quantify the responsiveness of air pollutants to infinitesimal perturbations of a model parameter or input (e.g., an emission rate of a precursor) with sensitivity coefficients (Cohan et al., 2005).”

Comment 8: *The introduction of OA expression in Table 4 was incomplete*

Response 8: Thanks. We have corrected it.

Comment 9: *Please elaborate the determination of thresholds for different photochemical indicators in Fig.3.*

Response 9: Thank you for your comment. Detailed information on the determination of the thresholds is provided in Section 2.3. We clarified it in the caption of Figure 3:

Lines 225-226: **The thresholds derived in this study (see Table 3, with the method detailed in Section 2.3) are indicated as the grey vertical lines.**

Comment 10: *Delete the extra brackets in Fig.3(a).*

Response 10: Thanks. Figure 3 has been updated in the main text.

Comment 11: *Please elaborate the approach to distinguish the O₃ formation regime with shading colors as shown in Fig.5.*

Response 11: Thank you for your comment. The shading colors that distinguish O₃ formation regimes are based on the indicator values in each emission scenario, with the thresholds given in Table 3. We clarified it in the caption of Figure 5:

Lines 229-300: **Figure 5 O₃ isopleths (red lines) overlap with the O₃ formation regimes (shading color) identified with $P_{\text{H}_2\text{O}_2}/P_{\text{HNO}_3}$, HCHO/NO_2 , HCHO/NO_y , and NO_y (the thresholds given in Table 3) at the CCM site.**

References

- Peng, Y.-P., Chen, K.-S., Wang, H.-K., Lai, C.-H., Lin, M.-H., and Lee, C.-H.: Applying model simulation and photochemical indicators to evaluate ozone sensitivity in southern Taiwan, *Journal of Environmental Sciences*, 23, 790-797, 10.1016/s1001-0742(10)60479-2, 2011.
- Xie, M., Zhu, K., Wang, T., Yang, H., Zhuang, B., Li, S., Li, M., Zhu, X., and Ouyang, Y.: Application of photochemical indicators to evaluate ozone nonlinear chemistry and pollution control countermeasure in China, *Atmospheric Environment*, 99, 466-473, 10.1016/j.atmosenv.2014.10.013, 2014.
- Ye, L., Wang, X., Fan, S., Chen, W., Chang, M., Zhou, S., Wu, Z., and Fan, Q.: Photochemical indicators of ozone sensitivity: application in the Pearl River Delta, China, *Frontiers of Environmental Science & Engineering*, 10, 1-14, 2016.