Dear editor:

We thank you for your suggestions for further improvements to our manuscript. We have taken your concerns into account and give responses below on behalf of all authors. For clarity, editor comments are given in bold, followed by our responses. Modified text in our revised manuscript are given in quotes, italics and blue.

Responses:

1. **This time the co-editor has evaluated the revised manuscript, on behalf of the two reviewers. I find the revised manuscript has been improved, based on the reviewers' comments. However, the following minor points still need to be clarified and justified. I would appreciate it if the authors could take them into account.**

(line numbers are those for acp-2020-1251-ATC1.pdf, with track changes)

We thank the editor for the recognition of our work here, and address specific concerns below.

2. **Have the authors compared the simulated VOC concentration levels (Fig. 5c and d) with the observations to confirm the adequacy? For biogenic VOC levels over 16 ppb (line 275), any support observations are present? The concentration range would be too high for isoprene/monoterpenes - is it mostly contributed from less reactive species such as methanol? Some discussion should be added, as this point is also critical in the determination of the regimes.**

Since VOC concentrations are not measured by the standard air pollution monitoring networks in China, we cannot provide a comprehensive evaluation. Methanol is included in biogenic VOCs in Fig. 5, and contributes to the overall biogenic VOC concentrations. The editor is correct to note that methanol makes a substantial contribution to biogenic VOC over southern central China, as shown in Figure 1 below. Despite the relatively high concentrations of methanol, its reactivity is lower than most anthropogenic VOC species, and it makes relatively little contribution to O_3 sensitivity. In this study we have focused on the effect of anthropogenic VOC emissions, and we note that biogenic VOC are relatively low in all but one of the regions we investigate here. We now state this clearly in section 4 and 6:

Page 13, Line 272:
"The distribution of biogenic VOC concentrations (including isoprene and methanol) differs from that of anthropogenic VOCs (Fig. 5c, 5d)."

Page 17, Line 349:
"The determination of O_3 sensitivity regimes here is based on the O_3 responses to decreasing anthropogenic NO_x and/or VOC emissions, and any potential impacts of

changing BVOC emissions has not been assessed. Decreasing BVOC emissions may offset the increase in O_3 levels due to decreased NO_x emissions for the NCP, the YRD and the PRD, and would make all regions more VOC limited. We note that our conclusion of NO_x limitation in Chongqing may be sensitive to our underestimation of NO_2 levels (section 3), and to the higher BVOC emissions in this region, both of which reduce the ratio of NO_x to VOC in the region (Table 3).”

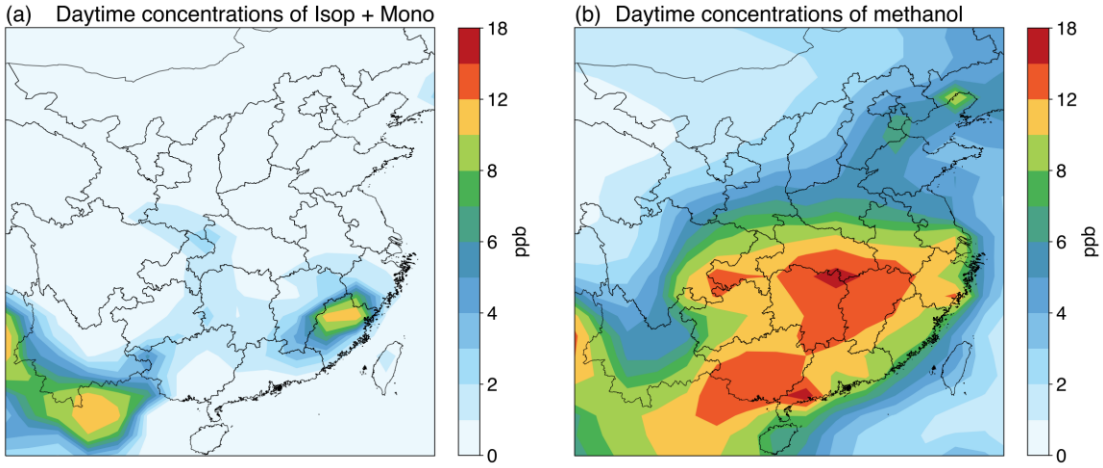

Figure 1. Spatial distributions of simulated surface daytime concentrations of isoprene and monoterpene (a) and methanol (b) in JJA, 2016, China.

3. **Line 257, Figure 5 and Table 3. How did the authors define the "daytime"? Any specific time period with the local time?**

In this study, we refer to MDA8 O_3 concentrations as daytime O_3 concentrations to enhance readability. Following the standard definition of MDA8, we use the Maximum Daily Average 8 hour concentrations calculated from consecutive 8 hour running mean values over 24 hours. We have then used the same time period (typically 11:00-18:00 or 12:00-19:00) for all species. We state this on line 255, but we have rephrased this to make it clearer.

Page 12, Line 255:
“We use the standard definition of the Maximum Daily Average 8-hour (MDA8) Ozone metric, and consider this same time period for other species, which we refer to hereafter as daytime concentrations.”

4. **Section 6. How much were the total VOC CONCENTRATIONS (or reactivity) reduced with the 20% emission change? I am wondering if biogenic VOCs might have been dominant and thus there is no virtual change in the total concentrations of VOCs and their role, even if the emissions of "anthropogenic" VOCs are varied over a wide range (in Chongqing for example).**

This is an interesting and valuable point, and the editor is correct to point out that the higher BVOC emissions influence the result and partly explain the higher NO_x needed to drive the region into VOC limitation. Our results reflect the effects of anthropogenic emission changes, which remain fully valid, but the differing underlying response may indeed reflect higher BVOC. In Table 3 we have shown the concentration of anthropogenic and biogenic VOC along with the ratio of NO_x to total VOC. This highlights the different chemical environment in Chongqing, and provides an additional reason for the different O_3 sensitivity. We have now amended the text to acknowledge the contribution of the higher background BVOC in Chongqing:

Page 17, Line 352:
"We note that our conclusion of NO_x limitation in Chongqing may be sensitive to our underestimation of NO_2 levels (section 3), and to the higher BVOC emissions in this region, both of which reduce the ratio of NO_x to VOC in the region (Table 3)."

5. **Line 351. Satellite observations of NO2 and HCHO have large uncertainties (of several tens of percents, e.g., Pinardi et al., 2020 for OMI NO2) and therefore the determination of regimes from the ratio would not be very certain.**

Pinardi, G., Van Roozendael, M., Hendrick, F., Theys, N., Abuhassan, N., Bais, A., Boersma, F., Cede, A., Chong, J., Donner, S., Drosoglou, T., Dzhola, A., Eskes, H., Fries, U., Granville, J., Herman, J. R., Holla, R., Hovila, J., Irie, H., Kanaya, Y., Karagkiozidis, D., Kouremeti, N., Lambert, J.-C., Ma, J., Peters, E., Piters, A., Postylyakov, O., Richter, A., Remmers, J., Takashima, H., Tiefengraber, M., Valks, P., Vlemmix, T., Wagner, T., and Wittrock, F.: Validation of tropospheric NO2 column measurements of GOME-2A and OMI using MAX-DOAS and direct sun network observations, Atmos. Meas. Tech., 13, 6141?6174, https://doi.org/10.5194/amt-13-6141-2020, 2020.

We agree that estimating O_3 sensitivity from satellite data carries substantial uncertainty as the editor mentions but we note that it provides a very useful indication of O_3 sensitivity regimes. We acknowledge this and we now use the word "*suggest*" to replace "*identify*" in the manuscript. We have modified text in section 6:

Page 17, Line 354:
"However, satellite observation based studies have also suggested this region as one that is largely NO_x limited in 2016, in contrast to the heavily populated coastal regions (Wang et al., 2021)."

6. **Figure 8f, lines 397-400. To my eye, the point with 40% NOx increase and 0% VOC change would be within the VOC limited side. In Figure 9a also, at the point of NOx emission change of 40%, surface O3 change is almost saturated. Please double check.**

The transition point for Chongqing lies at roughly 40% NO_x emissions increase, as the editor notes. We hence remove sentences on lines 401 – 403 to avoid the confusion.

7. **Line 433. Although the authors simply state that all selected regions across the globe outside of China are NOx limited, I would suggest that the chosen resolution would affect and the real situation is not that simple. For example in the central Tokyo, when studied at a resolution of ca. 10 km, summertime ozone formation is clearly VOC limited (Inoue et al., 2019). The effectiveness of legislated VOC emissions reduction is seriously studied. I hope the authors could acknowledge this point. Maybe in line 433, after "NOx limited," it would be better to add "with the studied horizontal resolution".**

Inoue, K., Tonokura, K. & Yamada, H. Modeling study on the spatial variation of the sensitivity of photochemical ozone concentrations and population exposure to VOC emission reductions in Japan. Air Qual Atmos Health 12, 1035?1047 (2019). https://doi.org/10.1007/s11869-019-00720-w

We agree that these conclusions are scale-dependent, and note that our results apply to the model scales resolved here that are representative of wider urban regions. Clearly smaller regions with more intense emissions may still be VOC limited, but these lie within wider regions that are NO_x limited at the scales considered here. We have added the following text in the manuscript:

Page 20, Line 437:
"We find that all selected high emission regions across the globe outside of China are NO_x limited at the model resolution considered here, such that NO_x emissions decreases yield regional O_3 decreases. Current levels of NO_x emissions in these regions are considerably lower than for the industrial regions of China, reflecting the different O_3 sensitivity regimes (Table 5). We note that these results apply to the wide urban regions considered here, and that local O_3 sensitivity in some parts of these regions may be different."