Response to referee #1

Referee Comment

Title: Ozone–NOx–VOC Sensitivity of the Lake Michigan Region Inferred from TROPOMI Observations and Ground-Based Measurements

General comments
Acdan et al. use TROPOMI column density retrievals and PAMS surface concentration measurements of HCHO and NO2 to study HCHO, NO2, and ozone production sensitivities (indicated by FNRs) in the lake Michigan region. The authors carefully composite for typical ozone season days, ozone exceedance days, weekdays, and weekends. They identify a spatial heterogeneity in ozone chemistry sensitivity for Chicago metropolitan area and its surrounding region, where the metropolitan area remains VOC-sensitive. They find that changes in FNRs on ozone exceedance days indicate an increase in NOx-sensitivity in NOx-sensitive areas and an increase in VOC-sensitivity in VOC-sensitive areas. Connecting wind fields with lake breeze provides a nice illustration of a stronger lake breeze effect on higher-ozone days.

Overall, the paper provides important implications for ozone mitigation in the Lake Michigan Region. However, a few major issues should be addressed before I recommend publication.

Introductory comment

We thank referee #1 for providing thorough feedback on our manuscript. Based on both referees' comments, we have made major revisions to the paper, including:

1. Changing the title of the manuscript to "Examining TROPOMI formaldehyde to nitrogen dioxide ratios in the Lake Michigan region: implications for ozone exceedances"
2. Removing all text/figures/references/etc. relating to PAMS data
3. Re-processing the data composites, specifically:
   a. Using the reprocessed TROPOMI NO_2 PAL dataset so that all data come from the same processor version
   b. Removing the use of detection limit thresholds
   c. Addressing the HCHO artifact over water through a bias-correcting approach
   d. Using the same number of days for the TROPOMI weekday-weekend composites
4. Adding new 2-meter temperature composites from the NAM analysis dataset
5. Expanding on the discussions of FNR errors (e.g., citing Souri et al., 2023), the usage of the J20 thresholds, and comparisons to similar studies (e.g., Tao et al., 2022)
6. Moving some of the appendices to a supplemental information document along with new supplemental figures/tables

We believe that these changes have greatly added to the scientific content of the paper and look forward to another round of discussion, if needed.

Our responses to referee #1's specific comments are as follows:

(1) The uncertainties in using OMI threshold values to interpret TROPOMI FNRs are briefly mentioned. However, despite similar signal-to-noise ratios, TROPOMI still shows disagreement with OMI, and it would be good to see some discussion on how this difference, along with TROPOMI bias, may affect the study results.

Response

We agree that these topics should be addressed in the manuscript.

Changes to manuscript

We have added a subsection to the data & methodology section entitled "2.4 Analysis of data composites". In this subsection we note that the primary method of analysis is through taking the difference between composite categories and providing a qualitative interpretation of what that means in terms of ozone chemistry sensitivity; we can then determine if the changes are due to changes in HCHO or NO_2 levels, or both.

Because we use the J20 threshold values as an additional analysis tool to interpret our TROPOMI derived FNRs, we also highlight some of the disagreements between TROPOMI and OMI data, as well as assumptions and uncertainties from the J20 study, and how these factors may affect the study results.

This new subsection can be found in the revised manuscript on lines 207–223.

(2) The unresolved biases and noises in FNR would be amplified as opposed to using HCHO and NO2 columns individually. The equation for uncertainty propagation was not properly implemented for division in Appendix B. For division ($z=xy$): $\delta z = \sqrt{(\delta[x][x])2 + (\delta[y][y])2}|z|$. More discussion on retrieval errors can be found in Souri et al. 2022 (https://acp.copernicus.org/preprints/acp-2022-410/).

Response

We thank the referee for pointing out the error in the equation.

Changes to manuscript

We have removed the equation from appendix section and instead added a subsection to the data & methodology section entitled "2.1.1 Errors associated with FNRs derived from S5P TROPOMI data". In this section, we reference the Souri et al. (2023) paper to provide a more detailed discussion of FNR errors and discuss what these errors imply for the FNRs we calculated for the Lake Michigan region. We reference Souri et al. (2023) eq. 15, which is the correct equation as the referee pointed out.

This new subsection can be found in the revised manuscript on lines 141–152.

(3) For weekday/weekend analysis, there seem to be many more days selected for weekdays (327 days) than weekends (132 days). Would the result be impacted by averaging over more days? It may be necessary to test if the same number of days were selected.

Response

We agree that it is best to test if the number of days selected impacts the results.

Changes to manuscript

We tested using only Tuesdays/Wednesdays in the weekday composites and Saturdays/Sundays in the weekend composites. Both composites were created with the same number of days over the 3-year period (114 days). The analysis of these new composites is largely the same as before. We replaced the old plots (created with unequal number of days) with the new weekday/weekend composites (created with equal number of days) in the revised manuscript (Figures 7–10 in the revised manuscript).

(4) It would be interesting to see how different regions are similar/different in changes on higher-ozone days. Is there any broader implication of this study on similar urban environments? For example, Tao et al. 2022 (https://pubs.acs.org/doi/full/10.1021/acs.est.2c02972) compare TROPOMI HCHO, NO2, and FNRs on ozone exceedance days versus non-exceedance days and weekdays versus weekends, for summer 2018 over New York City.

Response

We agree that comparisons to similar studies increases the scientific content of this paper.

Changes to manuscript

We added a paragraph talking about how our study is similar to Tao et al. (2022), which can be found on lines 379–383 in the revised manuscript. We end the paragraph with the following to highlight potential broader implications:

"These similarities suggest that the results presented here are broadly applicable to other coastal urban environments with O_3 exceedance problems. Future work could investigate FNRs over Detroit, Michigan, and Los Angeles, California, to see if this implication is true." (lines 381–383 in the revised manuscript)

Specific comments

P3. Line 89. The definition of "typical O3 season days" and "exceedance days" could be moved from P3. Line 104 to here (or briefly mentioned), as they appear for the first time.

Response

We agree with this suggestion.

Changes to manuscript

We moved the definitions of the "ozone season" and "exceedance days" to the introduction when the terms first appear. (line numbers 103–107 in the revised manuscript)

P4. Line 116-122. It is controversial to conclude that the errors affecting HCHO and NO2 retrievals can be canceled out rather than amplified by using their ratio. Please see more in the general comments.

Response

We agree with this comment.

Changes to manuscript

We added a subsection to the data & methodology section entitled "2.1.1 Errors associated with FNRs derived from S5P TROPOMI data" to provide a more detailed discussion of FNR errors. Please see our response to the related general comment above for more information.

P7. Section 2.2. What are the uncertainties in the PAMS surface measurements?

Response & changes to manuscript

To address the specific comments from Referee #2, we have decided to entirely remove the sections involving PAMS surface measurements and focus on analyzing TROPOMI and meteorological data composites.

P9. Line 233-235. This sentence on diurnal cycles seems confusing. Not sure how having diurnal information would make a difference in the current observations. What time of the day were the 6-day interval HCHO measurements collected? Or is it daily mean?

Response & changes to manuscript

To address the specific comments from Referee #2, we have decided to entirely remove the sections involving PAMS surface measurements and focus on analyzing TROPOMI and meteorological data composites.

P12. Line 309-311. May examine the mean temperatures for each composite and verify whether the higher-ozone days co-occur with hotter temperatures.

Response

We agree with this suggestion and believe it would add scientific content to the paper.

Changes to manuscript

We added 2-meter temperature data composite images for comparison. The data come from the NCEP North American Mesoscale (NAM) 12 km Analysis (ds609.0 | DOI: 10.5065/G4RC-1N9 | https://rda.ucar.edu/datasets/ds609.0/). These composites confirm our original suggestion that higher ozone days co-occur with hotter temperatures. These composites are Figures 2d–f and 7d–f and in the revised manuscript.

Technical comments

P2. Line 36. Could say "exceed the NAAQS" as O3 is mentioned three times in this sentence.

Response

We agree with this suggestion.

Changes to manuscript

We removed "O_3" from before NAAQS so that the sentence does not sound too repetitive.

P6. Line 164. 15 individual monthly composites?

Response

Originally, we created 15 individual monthly composites (May-September = 5 months) for 2019–2021 (3 years) → 5 × 3 = 15 composites. We then combined them using a weighted average to get an overall 2019-2020-2021 mean O_3 season composite. This was originally done to reduce processing time while creating the composites.

However, to address comments by referee #2, we decided to re-process the TROPOMI data. During this re-processing, we made our compositing script more efficient, which allowed us to create a single 2019–2020–2021 mean O_3 season composite. We have confirmed that taking the weighted average of individual composites and creating a single composite of all the data produce the same average values.

Changes to manuscript

We have removed any mention of "individual monthly composites" from the revised manuscript and updated the data & methodology section to reflect the changes in the satellite data processing.

P6. Line 166. "Next, we ~~next~~ created"

Response

We thank the referee for catching this grammatical error.

Changes to manuscript

We have removed the second "next" as suggested.