Response to Reviewer #1

We thank the reviewer for their comments on the manuscript. We have addressed these comments as described below. All reviewer comments are presented in italic font while the author responses are displayed in standard font. Specific text that was added to the updated manuscript is provided in blue text.

This is an interesting paper, great job. This paper is an advancement over previous papers on the topic such as Jin et al. 2020, Jin et al., 2015, Wang et al., 2021, Koplitz et al., 2022, and Nussbaumer et al. 2022 (please cite the last two, references at the end) due to the extension of the analysis to 2021 and capturing multiple continents in a single paper.

We are pleased that the reviewer enjoyed the paper. We have added the last two references to the revised manuscript. Koplitz et al. (2022) is referenced when discussing that emission control strategies of NO_x has resulted in a transition from VOC-limited to NO_x-limited regimes in many urban regions. Nussbaumer et al. (2022) is cited when discussing the impact of COVID-19 lockdown restrictions on NO_x emissions and O_3 photochemical regimes.

My two major concerns with this paper: 1. Some figures could be improved (primarily Figure 1 and 3 as discussed in minor suggestions). 2. Inclusion of TROPOMI data in some capacity would substantially improve this paper and very strongly believe this is not out of scope for this manuscript (as discussed in next paragraph).

One additional inclusion that would make this paper more novel, would be to include TROPOMI data in some capacity. It would be insightful to do a OMI vs. TROPOMI intercomparison for at least one of the cities investigated for multiple years. What extra detail does TROPOMI gather that OMI does not? How do the FNRs compare during the overlapping timeframes? This would also help corroborate many of the claims in the paper such as some of the OMI trends attributed to instrument drift. Previous studies (pre-dating 2019) did not have this opportunity. I understand conducting this analysis globally would be a major lift, but for 1-5 cities in the US, this should be a minor lift. I realize that some of this was addressed in Johnson et al. 2023 (Figure 5), but you now have the opportunity to do it for a longer timeframe (2018 - 2021) (not just LISTOS 2018) and a few other cities. It should be included as a case study in a new section (section 3.6)

We agree with the review that including some comparison of the ability of OMI and TROPOMI to retrieve FNRs would add to the novelty of this study. We are currently conducting a study which focuses on this topic for the time period between 2019-2021. For the current manuscript we have now added information about the ability of OMI and TROPOMI to reproduce inter-city (7 major US cities discussed in the current manuscript) and inter-annual (2018-2019) FNR variability measured by AQS data. We have added an additional section (Sect. 3.6) and Fig. 9 into the revised manuscript, including additional discussion in the conclusion section, comparing normalized FNRs from AQS, OMI, and TROPOMI for New York, Los Angeles, Chicago, Washington DC, Pittsburgh, Atlanta, and Houston for 2018 and 2019. The following text has been added to Sect. 3.6: "To expand upon previous studies which investigated OMI FNR trends published prior to the availability of TROPOMI retrievals (e.g., Mahajan et al., 2015; Jin and Holloway, 2015; Souri et al., 2017; Jin et al., 2017, 2020), here we compare the ability of OMI and TROPOMI to reproduced inter-city and interannual FNR variability in the US measured by EPA AQS sites in the 7 major US cities illustrated in Fig. 2. For this purpose, we applied TROPOMI L2 HCHO version 2.1 and NO_2 version 3.6.2 interpolated to a standardized 0.1° × 0.1° grid format. Figure 9 shows the normalized summer mean FNRs (city-specific annual FNR values normalized by the 7-city FNR mean) for the 7 selected US cities for 2018 and 2019. For both years, TROPOMI was able to reproduce the inter-city variability in normalized FNRs more accurately compared to OMI for 5 of the 7 US cities. This is further emphasized by the fact that TROPOMI reproduced 48% and 93% of the inter-city FNR variance (R^2) measured by AQS data for 2018 and 2019, respectively, while OMI only reproduced ~30% of the FNR variability measured in both years. Furthermore, TROPOMI accurately reproduced the direction of change in AQS measured FNRs between 2018 and 2019 for 6 of the 7 cities (85%) while OMI was only able to reproduce the FNR differences for 3 of the 7 cities (43%). The improved capability of TROPOMI to capture spatiotemporal FNR variability compared to OMI is to be expected as recent studies have demonstrated improved HCHO and NO_2 retrievals from the newer and higher spatial resolution sensor (e.g., Souri et al., 2023a; Johnson et al., 2023) and OMI is far past the expected lifetime of the sensor. Future studies should intercompare the two sensor's retrievals of FNRs for the entire lifetime of TROPOMI which overlaps with OMI (2017-present) to fully understand the improvements when applying TROPOMI.".

Minor suggestions:

Line 19. Add some nuance that NOx emission reductions have been more prevalent than VOC emissions reductions. Or that NOx emission controls have been more effective at reducing NO2 concentrations, while VOC controls are important, especially in major cities, but represent a smaller fraction of overall VOC reductions. Both NOx and VOC reductions have occurred in many urban areas and it is important to acknowledge this in some capacity in both the Abstract and Discussion.

Due to limitations in the length of the abstract we did not add this discussion into the abstract. However, we have added the following information to the discussion section of the revised manuscript: "Emission control strategies for VOCs have also been shown to have caused regional reductions in the concentrations of these compounds; however, it is challenging to derive and assess the impact of VOC emission control strategies as there are thousands of different VOC compounds all with different chemical reactivity (Pei et al., 2022). Furthermore, a large fraction of VOCs is emitted from biogenic sources which cannot be controlled through changes in human activities (Guenther et al., 1995).".

Line 53. Used "inherent" twice. Remove one of them, preferably the second one.

This has been corrected.

Line 190. Modify "NO2" —> "urban NO2"

This has been modified.

Figure 1 and 3 are helpful to give a wider view of NO2 and HCHO, but it would also be helpful to have zoom-ins of some of the cities, such as Figure 6. I'd recommend as follows: add a completely new figure (or amend Figure 1) that would be similar to Figure 6, but only showing NO2 and HCHO for this 16-year average. For Figure 3, I recommend only showing this at the urban scale. Too much is lost in a hemispheric image such as Figure 3. Maybe put current Figure 3 in the supplemental if you would still like to include.

We thank the reviewer for suggesting these extra figures to help present our results. To address the comment about adding visualizations of HCHO and NO_2 column concentrations over time, we added two new supplemental figures to display the underlying indicator species information used to derive the values in Fig. 6. We decided to keep Fig. 1 as is since we organized the manuscript to discuss OMI-derived FNR trends from hemispheric- to city-scales. By keeping Fig. 1 we show the 17-year averages of HCHO, NO_2, and FNRs on a hemispheric-scale and by adding new supplemental figures (Fig. S7 and S8) we show the HCHO and NO_2 values for 2005-2010 and 2016-2021 time periods zoomed in for the 18 select cities shown in Fig. 6. We felt this was the most efficient way to address the reviewer's comment but keep the information in the paper the authors wished to present. We have added some text to Sect. 3.4 in the revised manuscript to introduce these new figures: "The spatial maps of OMI-derived HCHO and NO_2 VCD values for these same time periods over the 18 cities are displayed in Fig. S8 and S9, respectively." and discuss some of the results displayed in these new supplemental figures: "These spatial distributions of increasing FNR values retrieved by OMI are clearly correlated with decreasing tropospheric NO_2 over the vast majority of cities displayed in Fig. S9.".

To address the reviewer's comment about Fig. 3 we have added a new supplemental figure (Fig. S2 in the revised manuscript) displaying the same information but zoomed in for North America, Europe, and Asia separately. This new figure is introduced in Sect. 3.3: "The same information shown in Fig. 3 is displayed individually for North America, Europe, and Asia in the supplemental information (see Fig. S2).".

Line 225. "Combustion" —> "fossil fuel combustion"

Updated.

Line 233. Add somewhere in this sentence "due to biogenic emissions". Relatedly, I don't see HCHO/NO2 enhancement over South Asia, are you referring to Malaysia? It's hard to tell from this image. Please clarify to a subsection of South Asia.

These suggestions have been added to the updated manuscript. This sentence now reads: "The highest FNR values are observed in regions of the southeast US and south Asia (e.g., Malaysia) where there are no large cities and enhanced tropospheric column HCHO abundances, primarily from biogenic emissions, are observed.".

Figure 2. Recommend having legend on each plot individually OR have the legend be more prominent (larger), either option is OK. This plot is a bit busy and not intuitive, but don't have any easy suggestions to amend, other than potentially having three separate panels for each city (24 total), but maybe that'd be worse.

Based on this suggested we have updated Fig. 2 in the revised manuscript to have a more pronounced legend at the bottom of the figure.

Figure 2 and Table 1. Pittsburgh typo. Also be more descriptive with the title "USA" instead say "USA urban areas"

These have been corrected.

Line 244. Units of -0.05 and 0.15? I believe yr-1? Also slight preference to modify 0.15 to +0.15? Same comments for Table 1. I would prefer units of %/yr, but this is personal preference.

Yes, the units are yr^{-1}. We have also added "+" in this sentence. Based on personal preference, we did not however update the values to be % yr^{-1}.

Line 246 Spatial footprint of OMI must also be playing a role here too, since HCHO in urban areas can be heterogeneous. As you know, OMI has 13 x 24 km resolution at best, often much worse.

"coarse spatial resolution of the OMI footprint" has been added to the list of reasons why OMI is unable to replicate the variability of HCHO observed in the in situ data.

Line 260. What does "near unity" mean in this context? Not a FNR of 1, but something else? Hard to discern.

The statement in the original manuscript reads: "that ratios of mid-day tropospheric VCD FNRs to PBL and surface-level concentrations are near unity". To make this clearer we have altered this sentence in the revised manuscript to read: "Correlation between OMI VCD and AQS NO_2 was near 1.0 (R=0.98) and both data sources had normalized linear regression slopes of ~-0.20.".

Line 266. Why the HCHO increase hemispherically? Global temperature rising / more biogenic emissions? I think it's too cavalier to imply that anthropogenic VOC emissions are increasing from 2005 - 2021 as alluded to in the next sentence. Some cities have done a great deal reducing local anthropogenic VOC emissions.

In the original manuscript we discuss the potential positive drift in OMI HCHO version 3 collection 3 in the discussion section (Sect. 4). However, we realize this issue needs to be discussed earlier in the manuscript and is introduced in Sect. 3.3 of the revised manuscript with the following text: "It should be noted that the NASA-released operational OMI HCHO version 3 collection 3 data product used in this study has been shown to have a positive drift due to instrument aging (e.g., Marais et al., 2012; Zhu et al., 2014, 2017b). This positive trend in OMI HCHO data displayed in Fig. 3 is likely largely impacted by the artificial positive drift in the collection 3 OMI data. A new NASA OMI HCHO version 3 collection 4 product is in development using the SAO algorithm which has removed this positive drift in HCHO (Ayazpour et al., 2024; personal communication with the SAO HCHO algorithm team). This new HCHO retrieval product shows that HCHO has a near-neutral trend across most of the populated cities in the Northern Hemisphere. This new collection 4 retrieval data is not yet peer-reviewed or available to the public therefore is not used here and the remaining results in this study use OMI HCHO version 3 collection 3 data. However, to test the potential impact on the results of this study using an OMI VCD product with this average positive drift eliminated, we removed the mean annual Northern Hemispheric HCHO trend (~0.004 DU yr^{-1}) from the collection 3 data and evaluate the resulting FNR trends over 18 selected large cities in the Northern Hemisphere which is discussed in Sect. 3.4.".

We further discuss this issue in Sect. 3.4 of the revised manuscript with the following text: "To test whether the positive drift in the NASA OMI HCHO collection 3 data significantly impacted the results of the FNR trends over the 18 selected large cities in the Northern Hemisphere we present these same results in Fig. S5 with the OMI data which has the annual average Northern Hemispheric HCHO trend remove (more representative of OMI HCHO version 3 collection 4 data) and Fig. S6 shows the spatial trends of HCHO, NO_2, and FNRs over the Northern Hemisphere using this detrended HCHO data. Comparing Figs. S5 and 4, it is seen that while some of the FNR values are slightly lower in magnitude the positive trends are very similar using collection 3 HCHO retrievals and a data product with the positive drift removed. Throughout the Northern Hemisphere HCHO trends now display both positive and negative values (see Fig S6) instead of the constant positive trends from the OMI HCHO collection 3 product. Using the detrended OMI HCHO data does result in more negative FNR trends in remote regions outside of large urban regions; however, over urban areas, and rural regions surrounding large cities, the FNR trends are still positive as displayed in Fig. S5 and S6. Overall, using the OMI HCHO version 3 collection 3 data product does not significantly impact the FNR results in large cities in the Northern Hemisphere focused on in this study. Future studies investigating FNRs conducted when the NASA OMI HCHO version 3 collection 4 data is available to the public should however use this new product to present more accurate results compared to those shown here using the NASA OMI HCHO version 3 collection 3 product.".

Figure 6. I have a slight preference if 2020 data was excluded from this spatial plot analysis. Low NO2 during 2020 was driven by stay-at-home measures and not polices, so from a policymaking perspective, I don't think inclusion of 2020 is warranted. Section 3.5 is great, and is how I recommend the 2020 data to be discussed. Personally, I also feel that black grid boxes on this figure are not helpful. Maybe include a copy of this figure with the black boxes in the supplemental for those interested?

We understand the reviewer's preference about excluding OMI 2020 data from this portion of the analysis, especially from a policy-making perspective. However, the goal of this study was to demonstrate the long-term trends of FNRs in OMI data, not to evaluate the effectiveness of satellite-derived data to be used in a decision-making capacity. In fact, we discuss in the manuscript about how this data likely has errors much too large for applying in policy making. Therefore, we decided to keep the 2020 data in the analysis.

Figure 6 has been updated as suggested by the reviewer and text has been added to the revised manuscript to point the reader to the new supplemental figure displaying the CGLC-MODIS-LCZ urban grids for each city: "Figure 6 shows that OMI is able to retrieve the differences in FNRs in urban and rural regions surrounding large cities in the Northern Hemisphere (Fig. S10 shows the same information in Fig. 6 except with the CGLC-MODIS-LCZ urban grids used to separate urban and rural values).".

Discussion in Lines 428 - 440 falls flat for me because you are projecting future policy recommendations based on an old instrument (OMI). TROPOMI is better. TEMPO will be even better. This is not discussed here and should be. This is one of many reasons, why I believe that including TROPOMI data in any capacity in this paper is necessary, and not out of scope. I see that some of this discussion is in Lines 469 - 482. Maybe Lines 428 - 440 & Lines 469 - 482 need to be merged together.

We agree with the reviewer. As explained above we have added a comparison of OMI and TROPOMI FNRs for 7 US cities in 2018 and 2019. This clearly shows that TROPOMI is more accurately able to reproduce observed FNR spatiotemporal variability. This is further discussed in the conclusion section of the revised manuscript.

References:

Ayazpour, Z., Abad, G. G., Nowlan, C. R., Sun, K., Kwon, H. A., Miller, C. C., et al.: Aura Ozone Monitoring Instrument (OMI) Collection 4 Formaldehyde Product, ESS Open Archive, http://dx.doi.org/10.22541/essoar.171804891.19520982/v1, 2024.

Guenther, A., Hewitt, N., Erickson, D., Fall, R., Geron, C., Graedel, T., Harley, P., Klinger, L., Lerdau, M., McKay, W., Pierce, T., Scholes, B., Steinbrecher, R., Tallamraju, R., Taylor, J., and Zimmerman, P.: A global model of natural volatile organic compound emissions, J. Geophys. Res., 100, 8873–8892, 1995.

Johnson, M. S., Souri, A. H., Philip, S., Kumar, R., Naeger, A., Geddes, J., Judd, L., Janz, S., Chong, H., and Sullivan, J.: Satellite remote-sensing capability to assess tropospheric-column ratios of formaldehyde and nitrogen dioxide: case study during the Long Island Sound Tropospheric Ozone Study 2018 (LISTOS 2018) field campaign, Atmos. Meas. Tech., 16, 2431–2454, https://doi.org/10.5194/amt-16-2431-2023, 2023.

Jin, X., and Holloway, T.: Spatial and temporal variability of ozone sensitivity over China observed from the Ozone Monitoring Instrument, J. Geophys. Res. Atmos., 120, 7229–7246, doi:10.1002/2015JD023250, 2015.

Jin, X., Fiore, A. M., Murray, L. T., Valin, L. C., Lamsal, L. N., Duncan, B., Boersma, K. F., De Smedt, I., Abad, G. G., Chance, K., and Tonnesen, G. S.: Evaluating a Space-Based Indicator of Surface Ozone-NOx-VOC Sensitivity Over Midlatitude Source Regions and Application to

Decadal Trends, J. Geophys. Res.-Atmos., 122, 10439–10461, https://doi.org/10.1002/2017JD026720, 2017.

Jin, X., Fiore, A., Boersma, K. F., De Smedt, I., and Valin, L.: Inferring Changes in Summertime Surface Ozone–NOx–VOC Chemistry over U.S. Urban Areas from Two Decades of Satellite and Ground-Based Observations, Environ. Sci. Technol., 54, 6518–6529, https://doi.org/10.1021/acs.est.9b07785, 2020.

Mahajan, A. S., De Smedt, I., Biswas, M. S., Ghude, S., Fadnavis, S., Roy, C., and van Roozendael, M.: Inter-annual variations in satellite observations of nitrogen dioxide and formaldehyde over India, Atmos. Environ., 116, 194–201, https://doi.org/10.1016/j.atmosenv.2015.06.004, 2015.

Marais, E. A., Jacob, D. J., Kurosu, T. P., Chance, K., Murphy, J. G., Reeves, C., Mills, G., Casadio, S., Millet, D. B., Barkley, M. P., Paulot, F., and Mao, J.: Isoprene emissions in Africa inferred from OMI observations of formaldehyde columns, Atmos. Chem. Phys., 12, 6219–6235, https://doi.org/10.5194/acp-12-6219-2012, 2012.

Pei, C. L., Yang, W. Q., Zhang, Y. L., Song, W., Xiao, S. X., Wang, J., Zhang, J. P., Zhang, T., Chen, D. H., Wang, Y. J., Chen, Y. N., and Wang, X. M.: Decrease in ambient volatile organic compounds during the COVID-19 lockdown period in the Pearl River Delta region, South China, Sci. Total Environ., 823, 153720, https://doi.org/10.1016/j.scitotenv.2022.153720, 2022.

Souri, A. H., Choi, Y., Jeon, W., Woo, J. -H., Zhang, Q., and Kurokawa J.-i.: Remote sensing evidence of decadal changes in major tropospheric ozone precursors over East Asia, J. Geophys. Res. Atmos., 122, 2474-2492, doi:10.1002/2016JD025663, 2017.

Souri, A. H., Johnson, M. S., Wolfe, G. M., Crawford, J. H., Fried, A., Wisthaler, A., Brune, W. H., Blake, D. R., Weinheimer, A. J., Verhoelst, T., Compernolle, S., Pinardi, G., Vigouroux, C., Langerock, B., Choi, S., Lamsal, L., Zhu, L., Sun, S., Cohen, R. C., Min, K.-E., Cho, C., Philip, S., Liu, X., and Chance, K.: Characterization of errors in satellite-based HCHO ⁄ NO2 tropospheric column ratios with respect to chemistry, column-to-PBL translation, spatial representation, and retrieval uncertainties, Atmos. Chem. Phys., 23, 1963–1986, https://doi.org/10.5194/acp-23-1963-2023, 2023a.

Zhu, L., Jacob, D. J., Mickley, L. J., Marais, E. A., Cohan, D. S., Yoshida, Y., Duncan, B. N., González Abad, G., and Chance, K. V.: Anthropogenic emissions of highly reactive volatile organic compounds in eastern Texas inferred from oversampling of satellite (OMI) measurements of HCHO columns, Environ. Res. Lett., 9, 114004, https://doi.org/10.1088/1748-9326/9/11/114004, 2014.

Zhu, L., Mickley, L. J., Jacob, D. J., Marais, E. A., Sheng, J., Hu, L., González Abad, G., and Chance, K.: Long-term (2005–2014) trends in formaldehyde (HCHO) columns across North America as seen by the OMI satellite instrument: Evidence of changing emissions of volatile organic compounds, Geophys. Res. Lett., 44, 7079–7086, https://doi.org/10.1002/2017GL073859, 2017b.