Response to the reviewers — Article ACP-2021-708

We thank reviewers for their thoughtful comments. Our detailed responses for each comment are listed below, along with the changes made to the manuscript to make these findings clear to readers. Our responses to the comments are presented in blue. The comments are shown in black. All page and reference numbers in our response are based on the revised manuscript. The line and reference numbers mentioned in the reviewers' comments are kept intact and are based on the original manuscript.

Reviewer 1

Minor Issues:

RC1.1. Line 26 of the text has the phrase "… with baseline chamber O3 concentrations above 90 ppb …." I suggest that this sentence and the following sentence be reworded to reflect the discussion of Figure 6 based on ambient MDA8 ozone, rather than on the chamber ozone. I think this would make the information in the abstract more policy relevant, and better reflect the discussion in the paper.

We have revised the sentence mentioned by reviewer in Line 27:

"The O_3-nonattainment days (MDA8 O_3 > 70 ppb) have O_3 sensitivity in the NO_x-limited regime, suggesting that a NO_x emissions control strategy would be most effective at reducing these peak O_3 concentrations. In contrast, a large portion of the days with MDA8 O_3 concentrations below 55 ppb were in the VOC-limited regime, suggesting that an emissions control strategy focusing on NO_x reduction would increase O_3 concentrations."

RC1.2. Line 88: Should "source" be plural?

'source' has been changed to 'sources'.

RC1.3. I suggest adding a sentence to the end of Section 2.4 that mentions the extensive sensitivity tests that were performed to ensure the relevance of the results, and will be discussed in Section 4.1

A sentence has been added in the end of Section 2.4:

'Section 4.1 presents a sensitivity study on the chamber measurement result using the chamber model described here.'

RC1.4. The discussion of Figure 2 in Section 3.1.1 requires improvement. It is noted that good agreement is observed between the time trends of the chamber and TROPOMI satellite remote sensing measurements. It is also suggested that the upward trend in NO2 concentrations in October–December, 2020 is likely associated with decreased boundary layer heights and increased fuel consumption for heating during the colder fall – winter season. However, the satellite measurement is a column measurement; the in situ chamber NO2 concentrations would depend upon boundary layer height, but a true column measurement would be independent of the boundary layer height. A more accurate discussion is required here, and that discussion should fully consider the averaging kernel of the satellite column measurement.

Increased boundary layer heights are often associated with increased wind speeds in the boundary layer, leading to downwind advection and dispersion of pollutants. This increased dispersion reduces the column concentrations of NO_x or HCHO measured by the TROPOMI satellite. This point has been clarified on line 247 of the revised manuscript.

RC1.5. The discussion in the preceding comment also applies to the comparison between the in situ and satellite column measurements of HCHO and CO*Biogenic; this discussion also should be improved.

Same response to RC1.4.

RC1.6. The caption of Figure 3 mentions "variance", a term that has a specific statistical meaning. I suggest replacing it with "variation".

'variance' has been changed to 'variation'.

RC1.7. The figures are mis-numbered: two numbered 6 and figs. 8 and 9 incorrectly labeled Figs 7 and 8. The text does refer to the correct numbering.

Figure numbers have been corrected.

RC1.8. In Figure 10, the cities in the right graph are not all in SoCAB; some are in the Salton Sea AB; I also think that it would be useful to include the Salton Sea AB in the left graph,

Results for the Salton Sea Air Basin have been added to Figure 10(a) and the caption for Figure 10(b) now refers to the region as "Southern California", instead of "SoCAB".

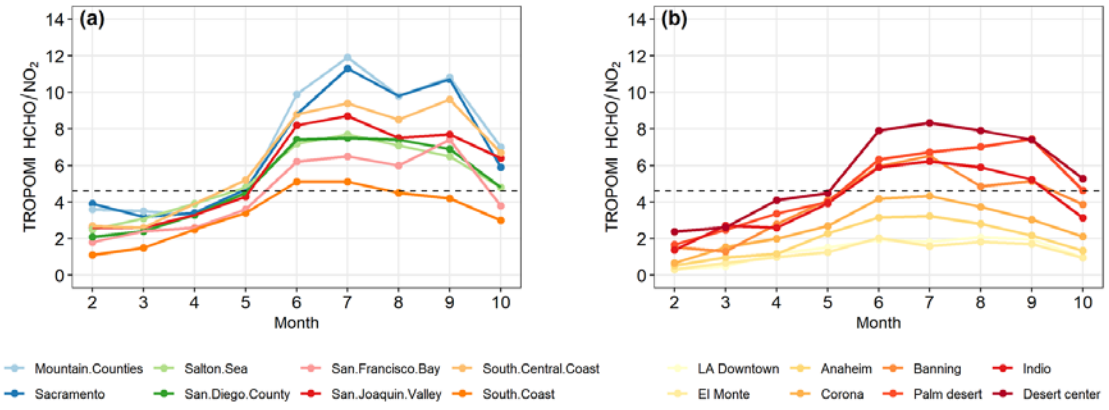

Figure 10. Monthly variation of TROPOMI HCHO/NO_2 in different air basins (left panel) and in different cities in Southern California (right panel). The darker colors in the right panel indicate increasing distance from the urban center of Los Angeles.

RC1.9. Line 460: please correct figure number and tense "Figure 10 shows that the chamber model can accurately predicted the measured …."

The figure number has been changed to 'Figure 11', and the word 'predicted' has been changed to 'predict'.

RC1.10. Line 464: should refer to Fig. S9.

This change has been made in revised manuscript.

RC1.11. The sentence beginning on Line 492 is easy to misinterpret; I suggest rewording: "Increasing the magnitude of the NOx perturbation increased the absolute magnitude of the ΔO_3 value but did not shift the chemistry into a different regime."

This change has been made in revised manuscript in Line 529.

RC1.12. The last sentence in Section 4.1.4 should be amplified slightly; I suggest rewording: "It should be noted that operation of the mobile smog chamber system in cities with higher ambient NO_x concentrations is expected to give O_3 sensitivity results that are even less dependent on the NO_2 perturbation size.

This change has been made in revised manuscript in Line 547.

RC1.13. Line 513: I suggest that the sentence begin with "Current California …", since California has a history of addressing a great many precursor emission sectors.

This change has been made in revised manuscript in Line 550.

RC1.14. I would strongly argue that the sentence beginning on line 518 is inaccurate. Ambient measurements indicate that over decades VOCs and NOx have decreased at average annual rates of about 7.5% (Warneke et al., 2012) and 2.6%, respectively. Over 30-years (1980 to 2010) those rates correspond to decreases of factors of 10 and 2.2, respectively. These are based on measurements in the SoCAB, but are relevant for the entire state. The Cox et al. and Rasmussen et al. references rely on emission inventories for their estimates, which are far inferior to actual ambient measurements. In my view it is important that the tremendous success of emission control efforts is highlighted at every opportunity. At the very least, this paper should discuss both the inventory and ambient measurement estimates of emission decreases.

We have added a sentence on line 556 of the revised manuscript to describe the trends in ambient measurement that support the decreasing emissions.

Revised text:

The estimated VOC emissions decreased by a factor of 3 while NO_x emission decreased by a factor of 1.5 between 1980 to 2010 according to the California inventory (Cox et al., 2013; Rasmussen et al., 2013). Long-term ambient measurements in the SoCAB confirm that ambient VOC concentrations decreased at an average rate of 7.5% yr^{-1} , while ambient NO_x concentrations decreased at an average rate of 2.6% yr^{-1} between the years 1980 to 2010 (Pollack et al., 2013; Warneke et al., 2012).

Reference:

Cox, P., Delao, A. and Komorniczak, A.: The California Almanac of Emissions and Air Quality - 2013 Edition.

[online] Available from: https://www.arb.ca.gov/aqd/almanac/almanac13/almanac13.htm, 2013.
Pollack, I. B., Ryerson, T. B., Trainer, M., Neuman, J. A., Roberts, J. M., Parrish, D. D., Pollack, C. :, Ryerson, T. B., Trainer, M., Roberts, J. M. and Parrish, D. D.: Trends in ozone, its precursors, and related secondary oxidation products in Los Angeles, California: A synthesis of measurements from 1960 to 2010, J. Geophys. Res. Atmos., 118(11), 5893–5911, doi:10.1002/JGRD.50472, 2013.
Rasmussen, D. J., Hu, J., Mahmud, A. and Kleeman, M. J.: The ozone-climate penalty: Past, present, and future, Environ. Sci. Technol., 47(24), 14258–14266, doi:10.1021/es403446m, 2013.
Warneke, C., De Gouw, J. A., Holloway, J. S., Peischl, J., Ryerson, T. B., Atlas, E., Blake, D., Trainer, M., Parrish, D. D., Warneke, C. :, De Gouw, J. A., Holloway, J. S., Peischl, J., Ryerson, T. B., Atlas, E., Blake, D., Trainer, M. and Parrish, D. D.: Multiyear trends in volatile organic compounds in Los Angeles, California: Five decades of decreasing emissions, J. Geophys. Res. Atmos., 117(D21), 0–17, doi:10.1029/2012JD017899, 2012.

Reviewer 4

This paper provides results about the response of ozone concentration to changes in precursor NO_x and VOC concentrations. The bulk of the experimental and analysis setup relies on smog chambers analysis. They authors offer quantitative results illustrating the complexities of near surface ozone chemistry and atmospheric conditions and its seasonal variation.

The paper also makes use of TROPOMI observations. To better understand the validity of the results obtained from TROPOMI some questions rise:

RC4.1. Discuss and provide references evaluating the accuracy of TROPOMI HCHO and NO_2 products. Furthermore, are the satellite retrievals corrected to account for the validation results reported in the literature? A good starting point for NO_2 will be https://amt.copernicus.org/articles/14/481/2021/, and for HCHO https://amt.copernicus.org/articles/13/3751/2020/amt-13-3751-2020.html. If there is no correction, how do these uncertainties translate into results derived using TROPOMI NO_2 and HCHO?
We have added discussions and references evaluating the accuracy of TROPOMI HCHO and NO_2 products (Verhoelst et al. 2021, Vigouroux et al. 2020) on Line 189-200 in the revised manuscript as the reviewer recommended.

"Correction factors were not applied to TROPOMI data in the current study. Verhoelst et al. (2021)and Vigouroux et al. (2020a) analyzed the accuracy of the TROPOMI data using ground-based measurement sites across the globe. Measurements were not made in California, but several of the evaluation sites had attributes similar to locations in California. Bias in daily TROPOMI NO_2 retrievals varied between -15% to -56% in moderately polluted areas with NO_2 column measurements between 3×10^{15} - 14×10^{15} molec cm^{-2} (typical for moderate-sized cities in California). The bias in TROPOMI HCHO measurements ranged between +26%±5% at low HCHO levels to -30.8%±1.4% at

high HCHO levels. HCHO levels measured in Sacramento (~0.6 x 10^{15} molec cm^{-2}) had a bias of approximately zero. These results suggest that TROPOMI measurements over California almost certainly contain some amount of bias that could only be removed through a comparison to measurements from a ground-based network. Application of global-average bias correction factors would not change the trends in HCHO and NO_2 in time and space even if they would change the absolute magnitude of those values. The current analysis will therefore focus on trends in the TROPOMI measurements."

RC4.2. What is the reason behind using QA bigger than 0.5 for both products? QA for NO_2 and HCHO have slightly different definition. From the TROPOMI HCHO user document: "In order to avoid misinterpretation of the data quality, it is recommended to only use those TROPOMI pixels associated with a qa_value above 0.5 (no error flag, cloud radiance fraction at 340 nm<0.5, Solar Zenith Angle (SZA)<=70°, surface albedo<=0.2, no snow/ice warning, air mass factor>0.1).". The NO2 user guide has a different definition: "qa_value > 0.75. For most users this is the recommended pixel filter. This removes cloud-covered scenes (cloud radiance fraction > 0.5), part of the scenes covered by snow/ice, errors and problematic retrievals.

As the reviewer mentioned, the ESA recommends QA values > 0.75 for most users retrieving values for individual days. In our use case, we averaged TROPOMI data to monthly concentrations, which mitigates the effects of uncertainty inherent in any individual data point. According to the ESA (ATBD document of TROPOMI NO_2), QA values > 0.50 add "the good quality retrievals over clouds and over scenes covered by snow/ice. Errors and problematic retrievals are still filtered out." Data with QA values > 0.50 therefore seems reasonable when constructing monthly averages. Incorporating TROPOMI NO_2 data with QA values > 0.5 increased the number of available data points and produced more robust statistics than calculations that only used individual data points that passed the highest level of QA. These points have been included in the discussion on Line 186 of the revised manuscript.

Revised text:

Quality assurance (QA) values were obtained alongside the HCHO and NO_2 data, and only measurements with QA values ≥ 0.50 were retained to ensure good data quality and sufficient data points when computing monthly averages (Van Geffen et al., 2021).

RC4.3. How was the satellite data averaged in the spatial domain for the state wide results?

For the statewide analysis, we re-gridded all the satellite data into 5 km grids and calculated the monthly averages of each data product in each grid. The monthly averages of HCHO and NO_2 were used to calculate the ratios. We have added the following in the caption of Figure 9: "TROPOMI NO_2 and HCHO data are re-gridded to 5 km resolution when calculating monthly-average ratios."

RC4.4. While references to TROPOMI NO_2 and HCHO retrieval algorithm papers are provided the paper could benefit of a short description of each one of them.

As the reviewer recommended, we have added a short description of each retrieval algorithm in 1st paragraph of Section 2.2 as follows: "The retrieval algorithms for TROPOMI NO_2 data use the measurements of the earth's radiance in the visible absorption wavelengths (405 – 465 nm) made by the hyperspectral imaging spectrometer. The algorithms first derive the total slant column density of NO_2 using a Differential Optical Absorption Spectroscopy (DOAS) method. The total slant column NO_2 is then separated into stratospheric and tropospheric slant column densities of NO_2 while utilizing information from a data assimilation system. Finally, the tropospheric vertical column density of NO_2 is obtained by applying conversion factors, called air mass factors (AMFs), to the tropospheric slant column density of NO_2. The retrievals of TROPOMI HCHO data apply a similar DOAS method to the ultraviolet (UV) wavelengths (328.5 – 359 nm) of the solar spectrum."

Other minor comments:

RC4.5. Lines 86-90 provide information about previous studies using satellite observations to derive HCHO/NO_2 ratios. It will be good to include some more recent papers using instruments recently launched such as TROPOMI. Motivated by the COVID-19 lockdowns there is significant amount of literature looking at it, for example https://www.science.org/doi/10.1126/sciadv.abe1178. The reference to OMI nadir pixel resolution (13 k x 24 km) in line 89 is ambiguous since it seems to refer to the resolution at NADIR of the instrument not the HCHO/NO_2 ratio studies.

We have removed 'with 13 km x 24 km resolution' to avoid confusing readers. We have added recent publications (Chossiere et al. 2021) in the revised manuscript in Line 75.

Revised text:

Satellite retrievals of HCHO/NO_2 from Global Ozone Monitoring Experiment (GOME), SCanning Imaging Absorption spectroMeter for Atmospheric CartograpHY (SCIAMACHY), Ozone Monitoring Instrument (OMI) and TROPOspheric Monitoring Instrument (TROPOMI) have extended these O_3 sensitivity calculations over broad geographical regions (Chossière et al., 2021; Duncan et al., 2010; Jin et al., 2017; Martin et al., 2004; Schroeder et al., 2017a).

RC4.6. While discussing figure 3 and the seasonal trends on O_3 sensitivity elevated fire plumes are mentioned repeatedly to explain discrepancies. It would be a more convincing argument if those satellite observations were clearly associated to fires by quantitatively explaining the range of NO_2 and HCHO TROPOMI columns in fire and non-fire plumes as well as if considered necessary, utilizing other sources of information such as back trajectories or satellite aerosol retrievals.

The wildfire detection method used in the current manuscript uses ground-based measurements to detect rapid changes in concentrations that are indicative of wildfire impacts. Many of the wildfire plumes detected using this approach were also detected by TROPOMI. Figure 1 compares TROPOMI HCHO and NO_2 measurements based on a 5 km radius buffer during days classified as "no-wildfire" and "wildfire" based on ground-based measurements between August to October 2020. The median TROPOMI NO_2 and HCHO measurements on "wildfire" days are approximately 14% and 44% higher than measurements on "no-wildfire" days, respectively.

The transport of fire plumes is strongly affected by smoke injection height, which is a function of fire intensity. Plumes with large amounts of thermal energy can be injected above the daytime mixing depth and can be transported aloft without reaching the ground. These plumes would not trigger the ground-based wildfire detection method, but they would still be visible to TROPOMI. In September 2020, many wildfires occurred in high-elevation areas such as the Sierra Nevada Mountains to the east of the ground-based measurement site. We have visually checked satellite images provided by NASA WorldView and confirmed the presences of fire plumes transported from those mountainous areas (example below).

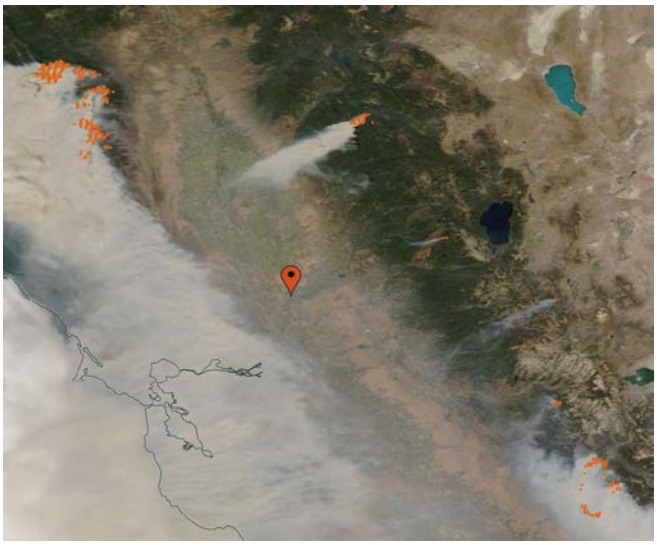

Image: September 8, 2020 (center: Sacramento; orange dots: fire locations detected by MODIS).

CALIPSO satellite products report the vertical profile of aerosols, but coverage over the study period is limited. More widely available aerosol optical depth (AOD) from MODIS MAIAC (1 km resolution; (Lyapustin et al., 2018)) confirms the presence of wildfire plumes during fall 2020, but does not differentiate between elevated plumes and plumes that reach the ground. We describe the potential influence of elevated plumes as a plausible explanation for the discrepancies between ground-based measurements and TROPOMI measurements, but further research would be needed to test this hypothesis.

Figure 1 has been added in the revised SI as Figure S7. A sentence has been added in revised manuscript to describe Figure 1 in Line 312:

'Figure S7 compares TROPOMI HCHO and NO_2 on wildfire days and non-wildfire days. Median TROPOMI HCHO measurements increased by 44% and TROPOMI NO_2 measurements increased by 14% on wildfire days.'

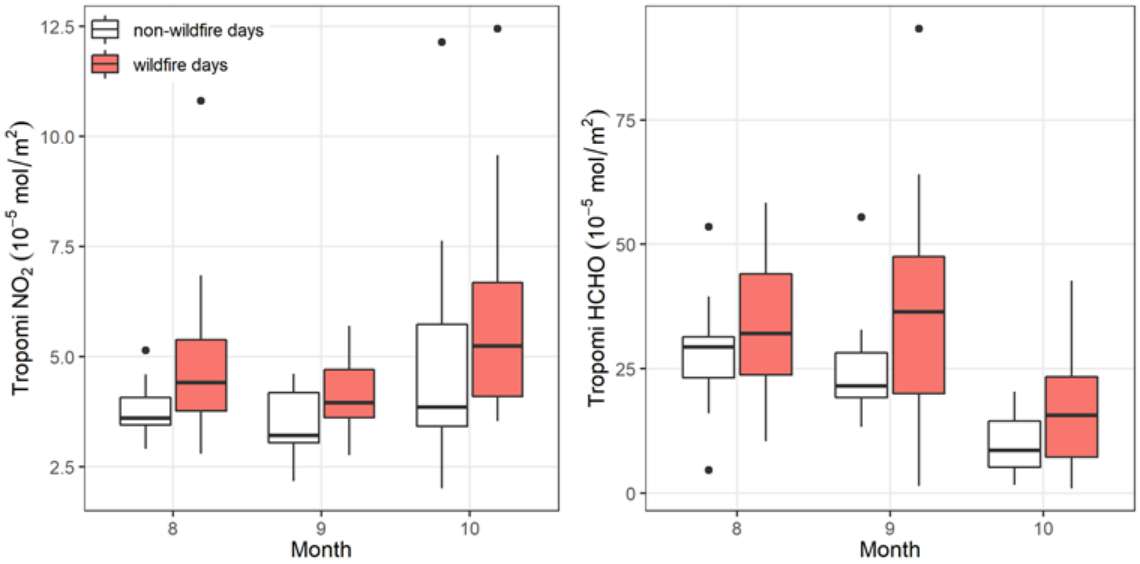

Figure 1. Monthly box and whisker plot of TROPOMI HCHO and NO_2 in wildfire days (solid box) and non-wildfire days (open box) from August to October, 2020. TROPOMI HCHO and NO_2 is in the 5km radii buffer of the chamber measurement site in Sacramento.

RC4.7. Figure 3 may be easier to interpret if the TRPOMI HCHO/NO_2 scale for panels a) and b) is similar. Why is it inverted in panel b)?

Figure 3a shows the monthly variation of O_3 response to NO_x perturbation, while Figure 3b shows the monthly variation of O_3 response to VOC perturbation. Those two parameters have the exact opposite seasonal trend due to the NO_x-O_3-VOC chemistry. To show the consistency in the seasonal trend of chamber-measured and satellite-based O_3 sensitivity, we inverted the right Y axis (TROPOMI HCHO/NO_2) in Figure 3b. The result shows that the seasonal trend of chamber ΔO_3^{+VOC} is quite similar to the inverted TROPOMI HCHO/NO_2 trend. This helps to build confidence in the ground-based chamber measurements for ΔO_3^{+VOC}.

RC4.8. Regarding the TROPOMI HCHO/NO_2 ratio, regime transition value of 4.6 it would be very interesting if the authors could provide some context of how it compares to previously published studies. As far as I can tell 4.6 is in the higher end of the values reported in the literature for urban areas.

As the reviewer recommended, we have added some context about previous studies of HCHO/NO_2 regime transition value in the past paragraph in Section 3.2 in revised manuscript:

Revised text:

'The $HCHO/NO_2$ transition point directly measured in the current study is consistent with previous estimates constructed from the combination of satellite measurements and routine ground-based O_3 monitoring data (Jin et al., 2020). Other previous efforts to estimate $HCHO/NO_2$ value at the transition point between NO_x-limited and VOC-limited regimes typically couple satellite $HCHO/NO_2$ measurements with O_3 sensitivity or O_3 sensitivity indicators (i.e., LNO_x/LRO_x) predicted using reactive chemical transport models. These hybrid studies predict $HCHO/NO_2$ transition points lower than the value of 4.6 derived in the current study. Martin (2004) used $HCHO/NO_2$ from GOME to calculate the regime transition value $HCHO/NO_2$=1.0 for polluted areas across the globe. Duncan (Duncan et al., 2010) used OMI to estimate the regime transition value $HCHO/NO_2$=1~2 across the continental U.S.. Schroeder (2017) found the transition range could between $HCHO/NO_2$=1.3~5.0 during DISCOVER-AQ in Houston. These estimated $HCHO/NO_2$ transition values vary due to the different satellite resolution, retrieval algorithms, and inherent air pollution patterns over the different study areas. The finer resolution satellite data used in the current study combined with direct ground-based measurements of O_3 sensitivity should provide accurate information for the $HCHO/NO_2$ transition point between chemical regimes over California.'

RC4.9. Do the correlation plots and equations shown in figure 7 and S6 consider the variance and uncertainty of both parameters?

We used ordinary lease square (OLS) linear regression in the original versions of Figure 7 and S6. This assumes that O_3 sensitivity measured in chambers has little to no error. This assumption is supported by the consistency tests described in the 3rd paragraph of Section 2.1 that show good agreement among 3 chambers, with only 1% uncertainty between measurements. In the revised manuscript, we repeated the test for the transition $HCHO/NO_2$ threshold between chemical regimes using reduced major axis (RMA) regression. RMA regression assumes both x and y variables include errors. Figure 2 (has been added in revised SI) compares the results of the two regression models. The RMA regression estimates a transition $HCHO/NO_2$=4.4 between chemical regimes, which is in good agreement with the original OLS result of 4.6.

We have described the method of linear regression (OLS) in the captions for Figures 7 and S6. We have also added the text below to Section 3.2 of the revised manuscript:

'Ordinary lease square (OLS) regression was used to estimate the transition point $HCHO/NO_2$=4.6 between chemical regimes. This approach does not account for uncertainty in chamber $\Delta O_3^{+NO_x}$. Repeating the analysis using reduced major axis (RMA) regression that accounts for errors in both x and y yields an estimated transition point $HCHO/NO_2$=4.4 between chemical regimes.'

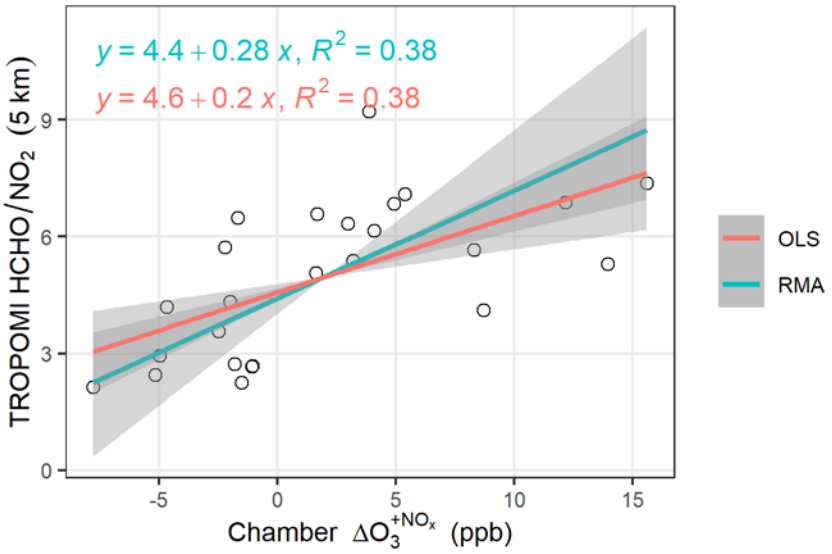

Figure 2. Correlation between weekly averaged TROPOMI HCHO/NO_2 at 5 km circular buffers and the weekly averaged $\Delta O_3^{+NO_x}$ from ground-based measurement during non-wildfire days. The shaded area shows the 95% confidence interval of the mean response of the predicted value. Red regression line generated using ordinary least squares regression. Green regression line generated using reduced major axis regression.

Reference:

Chossière, G. P., Xu, H., Dixit, Y., Isaacs, S., Eastham, S. D., Allroggen, F., Speth, R. L. and Barrett, S. R. H.: Air pollution impacts of COVID-19–related containment measures, Sci. Adv., 7(21), doi:10.1126/SCIADV.ABE1178/ASSET/C9139A3B-89F3-4EC2-B4A0-1E6157B0E236/ASSETS/GRAPHIC/ABE1178-FX2.JPEG, 2021.

Duncan, B. N., Yoshida, Y., Olson, J. R., Sillman, S., Martin, R. V., Lamsal, L., Hu, Y., Pickering, K. E., Retscher, C., Allen, D. J. and Crawford, J. H.: Application of OMI observations to a space-based indicator of NOx and VOC controls on surface ozone formation, Atmos. Environ., 44(18), 2213–2223, doi:10.1016/j.atmosenv.2010.03.010, 2010.

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Ialongo, I., Virta, H., Eskes, H., Hovila, J. and Douros, J.: Comparison of TROPOMI/Sentinel-5 Precursor NO2 observations with ground-based measurements in Helsinki, Atmos. Meas. Tech., 13(1), 205–218, doi:10.5194/amt-13-205-2020, 2020.

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anthropogenic emissions, Geophys. Res. Lett., 31(6), 2–5, doi:10.1029/2004gl019416, 2004.
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