Interactive comment on "Mobile MAX-DOAS observations of tropospheric NO₂ and HCHO during summer over the Three Rivers' Source region in China" *by* Siyang Cheng et al.: Reply to Anonymous

Referee #2

Referee comments are in black. Author responses are in blue.

This paper presents mobile MAX-DOAS measurement of tropospheric nitrogen dioxide (NO2) and formaldehyde (HCHO) during summer months over the Tibetan plateau. Mobile MAX-DOAS made four closed loop journeys each spanning 3 days. Measurements of slant column densities (SCDs) at 15 degrees elevation angles (EA) are converted to vertical column densities (VCDs) using geometric approximation. The paper presents diurnal variation, and spatial variation of NO2 and HCHO VCDs in the Tibetan plateau. Using the terrain altitude of the drive track, it also presents the vertical profile of NO2 and HCHO VCDs over this remote background region. Finally, the measured NO2 and HCHO VCDs are used to validate TROPOMI measurements over the region. This paper provides a rare measurement over a data scarce region and hence is worthy of publication to ACP. However, major revision is needed, focused on characterizing the instrument detection limit and measurement uncertainty, and justifying some of the conclusions of the paper before it is accepted for publication.

Reply: First of all, we appreciate the reviewer's positive comments on our manuscript. In response to the reviewer's comments and suggestions, major revisions have been made in the revised manuscript. Listed below are our responses and the corresponding changes made to the manuscript according to the suggestions given by the reviewer.

Major Comments:

The main focus of the paper is providing measurements over a data scarce remote background region. However, the paper lacks discussion of the instrument detection limit and measurement uncertainty. Proper characterization of the detection limit and measurement uncertainty is very important so that the data presented in the paper are properly utilized in the future. Please include discussion of the instrument detection limit and measurement uncertainty. Based on the presented RMS values, most of the measurement appears to be close to or below the detection limits of the instrument. Please comment on the frequency of measurement at or below the detection limits, and how this impacts the reported background values for NO2 and HCHO of 4 x 1014 and 2.27 x 1015 molecule/cm2 respectively. Uncertainty due to geometric approximation also needs to be better characterized with some radiative transfer calculations and using measurements at different EA. Right now measurements at different EA are only being used to filter data. Absolute difference in VCDs between 15 and 20 EA of 1 x 1015 molecule/cm2 for NO2 and 2 x 1015 molecule/cm2 for HCHO is used as one of the filtering criterias. This is a factor of 1-2 higher than the mean background value so the measured VCDs could have error >100%.

Reply: Many thanks for your comments. Per your suggestions, we have added the discussion of instrument detection limit and measurement uncertainty in Section 3.1 of the revised manuscript. Based on the spectral fit errors corresponding to filtered NO₂ and HCHO DSCDs, twice the medians of the spectral fit errors were estimated as the instrument detection limits for NO₂ and HCHO, which are 0.68×10^{15} and 2.11×10^{15} molec cm⁻², respectively. According to the DSCD detection limits divided by the DAMF for 15 ° elevation angle, the VCD detection limits were estimated to be about 0.24×10^{15} molec cm⁻² for NO₂ and 0.74×10^{15} molec cm⁻² for HCHO, respectively. These values are very similar to the estimation of the background levels for NO₂ and HCHO VCDs estimated by the maximum frequency method: the half widths at half maximum of the fitted curves were estimated to be their uncertainties ($\pm 0.23 \times 10^{15}$ molec cm⁻² for NO₂ and $\pm 0.96 \times 10^{15}$ molec cm⁻² for HCHO), respectively (Section 4.1).

There are 17% and 15% of the retrieved NO_2 and HCHO DSCDs below the detection limits, respectively. Based on the spectral fit errors, we can also calculate the relative errors for each NO_2 and HCHO DSCD. Then the mean relative errors (uncertainties) of NO_2 and HCHO DSCDs were about 21% and 12%, respectively.

It is a good suggestion to compare the geometric approximation and the atmospheric radiative transfer simulation. However, we lack necessary data during the field campaign to simulate the correct NO_2 and HCHO AMFs. For example, the varying azimuth angle and shelter situation in these viewing direction are not known exactly along the driving routes. Thus we compare and filter NO_2 and HCHO VCDs at different elevation angles, referring to previous study (Brinksma et al., 2008). In

this study, we used both absolute difference and relative difference as the filters, and data would be kept if at least one of both filters was fulfilled. The reason for choosing the absolute difference is to avoid to skip many measurements with low VCDs. With the condition of using two filters in this study, the means of the absolute differences and relative differences in the VCDs between 15° and 20° elevation angles are 5.48×10^{13} molec cm⁻² and 11% for NO₂ and 3.02×10^{14} molec cm⁻² and 7% for HCHO respectively.

We also tested the applicability of the geometric approximation method by radiative transfer simulations using typical parameters. According to the AODs from the AERONET website (<u>https://aeronet.gsfc.nasa.gov</u>, last access: 2 December 2022) at three sites (Mt_WLG, NAM_CO, QOMS_CAS) over the Tibetan Plateau, we estimate the AODs around 0.1 during our field campaign. But similar results are also found for AODs of 0.05 and 0.2. The simulation scheme is as the following.

Parameters	NO ₂	НСНО
Wavelength (nm)	440	340
Layer height (km)	0-1; 0-2	0-1; 0-2
Aerosol height (km)	same as trace gases	
AOD	0; 0.05; 0.1; 0.2	
SZA()	20, 40, 60, 70, 80	
RAA()	10, 30, 60, 90, 180	
Elevation angle ()	15	
Terrain height (km)	2, 3, 4, 5	

The VCD ratios of the RTM simulations and the geometric approximation for 15 ° elevation angle under the condition of AOD=0.1 can be obtained for NO₂ and HCHO, respectively. The DAMF ratios' means and standard deviations for all geometries (blue symbols) and RAA=10 °, SZA=60 ° and RAA=10 °, SZA=70 ° excluded (red symbols, for these rare measurement scenarios the strongest errors occur) are shown below. The main findings are that the typical errors of the geometric approximation are <20% for NO₂ and HCHO.



Figure S1 in the revised supplement

References:

Brinksma, E. J., Pinardi, G., Volten, H., Braak, R., Richter, A., Schönhardt, A., van Roozendael, M., Fayt, C., Hermans, C., Dirksen, R. J., Vlemmix, T., Berkhout, A. J. C., Swart, D. P. J., Oetjen, H., Wittrock, F., Wagner, T., Ibrahim, O. W., de Leeuw, G., Moerman, M., Curier, R. L., Celarier, E. A., Cede, A., Knap, W. H., Veefkind, J. P., Eskes, H. J., Allaart, M., Rothe, R., Piters, A. J. M., and Levelt, P. F.: The 2005 and 2006 DANDELIONS NO₂ and aerosol intercomparison campaigns, Journal of Geophysical Research, 113, D16S46, 10.1029/2007jd008808, 2008.

A large portion of the manuscript is dedicated to characterizing the temporal variation of the NO2 and HCHO even though the mobile MAX-DOAS drives covered large spatial region. The temporal variation analysis assumes there is a little to no spatial variation in these species along the drive track. However, this is not the case as shown in the spatial analysis plots. There is spatial variability as well as day to day variability along the drive track. Assuming the drives started at around the same time each day and reached the same locations around similar time during the drives, the diurnal variation presented here represents spatial variation and not temporal variation. This is also the likely reason for the W shape in HCHO diurnal variation, and U shape in NO2. So, it might be best not to include the temporal variation section in the paper or may be present it as spatial variation.

Reply: Many thanks for your suggestions. Yes, the NO_2 and HCHO VCDs, measured by mobile MAX-DOAS, reflected their combined spatio-temporal variations. From the findings in this study, we infer that the variable diurnal patterns of HCHO were connected with the secondary photochemical formation of active VOCs at different locations. We conclude that the 'U' shape of the NO_2 diurnal variations were primarily caused by enhanced pollution in the morning and evening when the mobile observation vehicle was located in or close to the cities or county town. An additional effect on the NO_2 diurnal variation is probably caused by the enhanced NO_2 photolysis around noon.

Therefore, as you suggested, we combined the Section 4.2 (Temporal variation) and 4.3 (Spatial distribution) in the original manuscript together, which were reorganized into Section 4.2 (Spatio-temporal variation) in the revised manuscript. This change is not marked up using revision track in order to keep the manuscript clear to read.

The paper talks about making measurements at high spatial resolution, but there is no information about the spatial resolution of the measurements in the paper. The only spatial information about the data is presented at 0.25 degree resolution. Please include the spatial resolution of the data. Further TROPOMI data is also gridded into 0.25 x 0.25 degree cells (~ 25 km x 25 km). This suggests that the mobile MAX-DOAS data is only available at 25 km spatial resolution. Please include reasoning for not averaging the MAX-DOAS data to the TROPOMI grid (~ 5km)?

Reply: It spent about 8 min for measurements at two adjacent 15 °elevation angle.

The corresponding spatial resolution can be estimated as ~8 km at a speed of ~60 km/h of the mobile vehicle. Assuming that the trace gas is located in the lowest 1000m above the surface, we can also estimate the horizontal extent of the line of sight through that layer. For measurements at 15 ° elevation angle, this extent is about 4km.

However, for the comparison with TROPOMI observations, we use the means of typically 2-3 data points (approximate 25 km) at a specific grid in order to reduce the uncertainties of the VCDs from both mobile MAX-DOAS and TROPOMI. In addition, the chosen grid cell matches the finest mesh of the ERA5 meteorological data. The spatial resolution of measurements and the reason for using the 0.25 °×0.25 ° grid have been added in the Section 3.2 and 4.3 of revised manuscript.

We also tried to match the individual MAX-DOAS measurement with TROPOMI Level-2 NO₂ and HCHO products (5.5 km ×3.5 km). Both data sets are combined under the conditions of Δ longitude<0.05° & Δ latitude<0.05° & Δ Time < 1.5h (or on the same day), which was referred to ' Δ T_{1.5}' ('All') in the figure below. The finer (0.05 °×0.05 °) spatial distributions of TROPOMI NO₂ and HCHO are also shown in the following. From the results with fine (0.05 °×0.05 °) and coarse (0.25 °×0.25 °) spatial resolutions, the conclusions drawn in this paper were almost the same. Thus we did not change the comparison results in the paper.







It is not clear how the authors came to the conclusion that "TROPOMI can't identify the fine scale spatial variability in the tropospheric NO2 VCDs in the background atmosphere over the Tibetan Plateau". TROPOMI data is not even being used at its native resolution for this analysis. Same with HCHO fine-scale and temporal variations.

Reply: Many thanks for your comments. Originally, we want to say: 'The elevated trends of the tropospheric NO_2 VCDs around the counties, which are clearly observed by the mobile MAX-DOAS, are nearly not captured by TROPOMI'. This conclusion has been deleted in revised manuscript.

I don't think regression analysis is appropriate tool for evaluating the TROPOMI satellite products over remote background region where the dynamic range of the data is very small. It is not clear how linear regression helps validate fine-scale variability in tropospheric NO2 and HCHO VCDs? The lower TROPOMI NO2 VCDs over the cities might be related to the difference in timing of the measurements between mobile MAX-DOAS measurements and TROPOMI measurements. Mobile MAX-DOAS generally measured higher NO2 over the cities in the morning during the start of the drive, whereas TROPOMI make measurements at mid-day when NO2 abundances are lowest. I suggest the authors focus on bias in TROPOMI data in the background region rather than correlation.

Reply: Although the regression analysis is limited by the dynamic range of NO₂ and HCHO levels, it is still a common method to compare ground-based observation with satellite products and correlation coefficients are usually small at a remote site (Vigouroux et al., 2020; Verhoelst et al., 2021). For NO₂, both mobile MAX-DOAS and TROPOMI present clear differences between Xining city and the main area of the Three Rivers' Source, although there are differences in measurement time in Xining city between the two methods. However, the elevated trends of the tropospheric NO₂ VCDs around the counties over the main area of the Three Rivers' Source, which are clearly observed by the mobile MAX-DOAS, are nearly not captured by TROPOMI. These differences of the NO₂ spatial distribution are reflected in the different correlation coefficients by the regression analysis for the situations of 'All' and ' $\Delta T_{1.5}$ ', respectively. The regression analysis for the HCHO VCDs also partly showed the similarities and differences of the HCHO spatio-temporal variations between mobile MAX-DOAS and TROPOMI. For example, the higher TROPOMI HCHO VCDs appear more in the second circling journey and the lower HCHO VCDs in the third and fourth circling journey, consistent with the results derived from mobile MAX-DOAS. Therefore, we find it very useful to retain the results of the regression analyses. Of course, comparing the bias between two data sets, which is significant over the background region, has been added in revised manuscript.

References:

Vigouroux, C., Langerock, B., Aquino, C. A. B., Blumenstock, T., Cheng, Z., Mazière, M. D., Smedt, I. D., Grutter, M., Hannigan, J. W., Jones, N., Kivi, R., Loyola, D., Lutsch, E., Mahieu, E., Makarova, M., Metzger, J.-M., Morino, I., Murata,

I., Nagahama, T., Notholt, J., Ortega, I., Palm, M., Pinardi, G., Röhling, A., Smale, D., Stremme, W., Strong, K., Sussmann, R., Té, Y., Roozendael, M. v., Wang, P., and Winkler, H.: TROPOMI–Sentinel-5 Precursor formaldehyde validation using an extensive network of ground-based Fourier-transform infrared stations, Atmos. Meas. Tech., 13, 3751–3767, 10.5194/amt-13-3751-2020, 2020.

Verhoelst, T., Compernolle, S., Pinardi, G., Lambert, J.-C., Eskes, H. J., Eichmann, K.-U., Fjæraa, A. M., Granville, J., Niemeijer, S., Cede, A., Tiefengraber, M., Hendrick, F., Pazmiño, A., Bais, A., Bazureau, A., Boersma, K. F., Bognar, K., Dehn, A., Donner, S., Elokhov, A., Gebetsberger, M., Goutail, F., Grutter de la Mora, M., Gruzdev, A., Gratsea, M., Hansen, G. H., Irie, H., Jepsen, N., Kanaya, Y., Karagkiozidis, D., Kivi, R., Kreher, K., Levelt, P. F., Liu, C., Müller, M., Navarro Comas, M., Piters, A. J. M., Pommereau, J.-P., Portafaix, T., Prados-Roman, C., Puentedura, O., Querel, R., Remmers, J., Richter, A., Rimmer, J., Rivera Cárdenas, C., Saavedra de Miguel, L., Sinyakov, V. P., Stremme, W., Strong, K., Van Roozendael, M., Veefkind, J. P., Wagner, T., Wittrock, F., Yela Gonz alez, M., and Zehner, C.: Ground-based validation of the Copernicus Sentinel-5P TROPOMI NO₂ measurements with the NDACC ZSL-DOAS, MAX-DOAS and Pandonia global networks, Atmospheric Measurement Techniques, 14, 481-510, 10.5194/amt-14-481-2021, 2021.

The spectral analysis or the method section has no references other than Platt and Stutz (2008). There are lots of papers describing MAX-DOAS measurements, retrievals, pros and cons of fixed vs sequential FRS. So, please add references to appropriate paper.

Reply: Many thanks for your kind suggestions. Several references have been added in the revised manuscript.

Minor Comments:

It would be great to show the VCD profiles as mixing ratio profiles. Temperature and pressure from the ERA5 reanalysis could be used for this purpose as well. This would very likely provide greater utility of the dataset. It would also be very useful to have the vertical profiles that does not include anthropogenic influenced values from Xining.

Reply: Many thanks for your suggestions. We have amended the profiles of the NO_2 and HCHO VCDs along the driving route altitude (Figure 6b in revised manuscript), which were used to derive the profiles of the NO_2 and HCHO mixing

ratios (VMRs, Figure S2 in the revised supplement, also shown below). During the calculation process of the NO₂ and HCHO VMRs, we firstly obtained the differences of the VCDs (Δ VCD) between two adjacent vertical intervals. Then we utilized hourly surface air pressure and temperature at 2 m above the land surface from ERA5 (with the 0.25 °×0.25 ° resolution) to calculate the mean temperatures and pressures at each altitude interval. In addition, it should be noted that the conversion of the Δ VCDs to VMRs was based on an assumption, i.e. the abundances of the trace gases at the same altitude are independent on measurement location. But this assumption may be not quite fulfilled. Therefore, the profiles of NO₂ and HCHO VMRs were just approximate estimation values and we tend to put them in the supplement.

The data points at the grid cell of 2000-2500 m represent the conditions around Xining. We prefer to keep the measurement point at this grid cell in order to clearly show the differences between Xining city and background region.



Figure 6b in the revised manuscript



Figure S2 in the revised supplement

How is ERA5 reanalysis temperature and solar radiation at the surface (SSRD) used to calculate daily temperature and SSRD? Do you interpolate it to each VCD measurements and then calculate the average for each day?

Reply: Yes, the air temperature at 2 m above the land surface and the downward solar radiation at the surface (SSRD) from ERA5 are firstly matched with each measured HCHO VCD. Then the daily averages of temperature and SSRD were calculated. The description has been made more clear in the revised manuscript.

It is not clear how relatively large noise in the TROPOMI satellite product results in small correlation coefficient between two data sets. Is the uncertainty in measurements included in the regression analysis?

Reply: The rather low signal-to-noise ratio of the TROPOMI HCHO VCDs will result in a partly random distribution, which will lead to a smaller correlation coefficient of the HCHO VCDs between mobile MAX-DOAS and TROPOMI. However, the noise level of the TROPOMI VCDs is difficult to estimate, because it not only depends on the spectral analysis, but also on the applied tropospheric AMF, which is in particular influenced by partial clouds and varying surface elevation. Therefore, the measurement uncertainties were not included during the process of the regression analysis in Figure 15. This information has been amended in the revised

manuscript.

I wonder why the authors decided not to simply do zenith DOAS measurements. It would remove some of the challenges associated with MAX-DOAS measurement such as signal blockage. I assume this is to leverage the air mass factor. This needs to be made clear. Since data from all EAs are not used, I also suggest the authors include some discussion on need for different EA measurements. This will be very helpful for future mobile MAX-DOAS measurements.

Reply: Many thanks for your comments and suggestions. The tropospheric light path is shorter at the zenith view, compared with the off-axis view. Especially for the background atmosphere with typically low concentration levels, the weak absorptions of the trace gases can be enhanced using longer light paths in the troposphere in order to enhance the detection limit. In addition, because this is the first practice of mobile MAX-DOAS observations over the Tibetan Plateau, we didn't know which elevation angle was the best for measuring the tropospheric VCDs of trace gases in the background atmosphere over mountain terrain before this campaign. Therefore, we made the telescope scanning at 7 elevation angles. In future studies on observations of tropospheric NO₂ and HCHO VCDs by mobile MAX-DOAS, we suggest to measure at 15°, 20°, 90° elevation angles. There are at least two reasons: (1) The larger elevation angles are less influenced by the road tilt and obstructions; (2) The measurements at 15° and 20° elevation angles with longer light path have a higher sensitivity (by about a factor of 2.9 for 20°, and 3.8 for 15° elevation angle, respectively) for tropospheric trace gases in the background atmosphere. We have added this information to the text at the end of Section 3.2 in revised manuscript from our experiences in this study.

Specific Comments:

Line 48: "grinding environment" – not sure what you mean by that. May be replace with complex terrain?

Reply: The description of "grinding environment" has been deleted in revised

manuscript.

Line 49: What do you mean by the sparseness of effective techniques and methods? Do you mean most methods don't work due to high altitude environment?

Reply: Yes. We mean that some instruments don't work due to special environment (such as lower pressure) over the Tibetan Plateau.

Line 93-96: Please provide more information about the methods. Right now it is very vague.

Reply: Many thanks for your suggestions. More information about the sampling observation methods and surface in-situ measurement methods has been added in the revised manuscript.

Line 135: Are there any other measurements on the vehicle?

Reply: Yes. The other measurements for the purpose of improving weather forecasts were performed by other groups and will be reported in another study.

Line 142: consider replacing "complex" with "difficult"

Reply: Agreed. Done.

Line 146: Is there a reference for the Tube MAX-DOAS instrument?

Reply: Yes. We have added some references for the Tube MAX-DOAS instrument.

Line 157: DC and OS are collected at night. Does the instrument need to be operating at night every day or can you use single DC and OS files?

Reply: The instrument recorded spectra of dark current (DC) and electronic offset (OS) at each night automatically. But we use single DC and OS files, which were

sufficient because the Tube MAX-DOAS worked at a stable detector temperature of 15 ± 0.1 °C.

Line 168: remove "in the field measurement area". I don't think it is needed.

Reply: Agreed. Done.

Line 226: For EA 15 degree measurements, SZA > 75 would result in the sun below the measurement geometry. Under such a condition, geometric approximation is likely not well suited? Should the SZA cutoff be 75 degree instead?

Reply: It should be noted that the ground-based MAX-DOAS observations utilize scattered illustrated below sun light. As in the sketch (http://iup.uni-bremen.de/doas/maxdoas instrument.htm, accessed date: 23 November 2022), if the scattering point is above the layer of the target species then the geometric approximation is applicable. In this study, SZA $< 80^{\circ}$ is used as one of the filters during the post processing of NO₂ and HCHO DSCDs. According to the aforementioned results about the applicability of the geometric approximation method by radiative transfer simulations, the geometric approximation is still well suited for measured data in the range of 75 SZA < 80 $^{\circ}$ without additional significant errors. We prefer to select the threshold of SZA<80° to balance the quality of the results and skipping not too many data.



Line 249: change "more exact, but depends on…" to "more accurate, but requires information on…"

Reply: Agreed. Done.

Line 250: change "less correct, but relatively simple …" to "simpler and assumes trace gases are uniformly distributed in the lower troposphere"

Reply: Agreed. Done.

Line 256: add polluted environment.

Reply: Done.

Line 265: Error due to pitch and roll distribution could be easily quantified. Consider quantifying this error rather than saying "errors will cancel out".

Reply: Many thanks for your kind suggestion. we estimate the error of the elevation angle to be about 2.3 °, based on the median of the mobile platform attitude angle during the effective MAX-DOAS measurement period. The corresponding error of an individual measurement will be up to about 21%. However, it should be noted that on average the positive and negative deviations of the elevation angle will almost cancel each other. Thus the errors of individual measurements will be usually much smaller (except for measurements on continuous strong slopes). For averages of several measurements the errors of the elevation angles lead to much smaller VCD errors with a magnitude smaller than 1% when using geometric approximation method (equation 6):

 $\alpha = 15^{\circ} - 2.3^{\circ} = 12.7^{\circ}, VCD=0.2818 \times DSCD;$ $\alpha = 15^{\circ}, VCD=0.3492 \times DSCD;$ $\alpha = 15^{\circ} + 2.3^{\circ} = 17.7^{\circ}, VCD=0.4232 \times DSCD;$ [(0.2818 × DSCD+0.4232 × DSCD) ÷2-0.3492DSCD] ÷(0.3492 × DSCD) × 100 = 1%

Line 269: change ~ to –. Also in line 362.

Reply: Agreed. Done.

Line 445: Lower NO2 for TROPOMI is likely due to difference in time of measurements.

Reply: Yes. Considering the NO_2 diurnal variations in cities (such as Xining), the lower NO_2 VCDs for TROPOMI in the 'All' situation in Figure 15 were also affected by the differences in time between two measurement methods. This cause has been added to the revised manuscript.

Figure 2: Elevation map of the drive track would be very useful.

Reply: Many thanks for your suggestion. The altitudes along the driving routes have been added in the Figure 2 of the revised manuscript.

Figure 5: Please remove square from measured data for clarity. RMS looks higher than listed in the figure legend especially for NO2. Please consider including fit residual plot as well.

Reply: Per your suggestions, we have amended the figure 5 about the spectral fitting for NO_2 and HCHO.

Figure 6: This information is probably better presented in table 3. Scatter plot between different EA VCDs will be more informative than what is currently presented in Figure 6. With the mean value dominated by few large values, I suggest including bias as well.

Reply: Many thanks for your suggestions. The Figure 6 has been removed in the revised manuscript and the corresponding information has been added to the amended Table 3.

Figure 11-14: It would be better to combine Figures 11 and 13 and Figures 12 and 14.

Reply: Many thanks for your suggestions. However, we prefer not to combine Figures 11 and 13 (Figures 12 and 14), because they include many kinds of colors and a lot of information besides the spatial distributions of the tropospheric NO₂ VCDs, respectively. For example, the colors of rivers and lakes, roads, and administrative boundaries on the background street map (https://map.baidu.com/, last access: 16 June 2022) in Figure 11 (Figure 12) cannot be modified and will be indistinguishable when we plot all the information of Figures 11 and 13 (Figures 12 and 14) together.

Figure 11: Is the gridded VCDs, mean VCDs over that grid or the median? Since we are dealing with background values, median VCDs might be better for comparison. Also approximately, how many data points are there in each grid?

Reply: We use the mean VCDs for the gridded VCDs. There are no significant differences in the VCD patterns during this campaign. There are typically 2-3 data points in each grid approximately.

Figure 15: Why do you have negative VCDs? Should they be excluded?

Reply: It is possible that there are negative VCDs for the background atmosphere, where the trace gas absorptions are very low. The negative NO_2 and HCHO VCDs are mainly caused by the scatter of the data in this study, which are related to the measurement noises. It is important to keep these negative VCDs. Otherwise we would introduce an artificial positive bias.

Figure S1: Consider including scatterplot between HCHO VCD and temperature in the paper rather than in SI.

Reply: Agreed. Done.