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Comment on acp-2022-629

Anonymous Referee #2

Referee comment on "Mobile MAX-DOAS observations of tropospheric NO_2 and HCHO during summer over the Three Rivers' Source region in China" by Siyang Cheng et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-629-RC2, 2022

This paper presents mobile MAX-DOAS measurement of tropospheric nitrogen dioxide (NO_2) 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 NO_2 and HCHO VCDs in the Tibetan plateau. Using the terrain altitude of the drive track, it also presents the vertical profile of NO_2 and HCHO VCDs over this remote background region. Finally, the measured NO_2 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.

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 NO_2 and HCHO of 4×10^{14} and 2.27×10^{15} molecule/cm^2 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 1×10^{15} molecule/cm^2 for NO_2 and 2×10^{15} molecule/cm^2 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%.

A large portion of the manuscript is dedicated to characterizing the temporal variation of the NO_2 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 NO_2. So, it might be best not to include the temporal variation section in the paper or may be present it as spatial variation.

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)?

It is not clear how the authors came to the conclusion that "TROPOMI can't identify the fine scale spatial variability in the tropospheric NO_2 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.

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 NO_2 and HCHO VCDs? The lower TROPOMI NO_2 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 NO_2 over the cities in the morning during the start of the drive, whereas TROPOMI make measurements at mid-day when NO_2 abundances are lowest. I suggest the authors focus on bias in TROPOMI data in the background region rather than correlation.

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.

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.

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?

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?

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.

Specific Comments:

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

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?

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

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

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

Line 146: Is there a reference 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?

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

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?

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

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

Line 256: add polluted environment.

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

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

Line 445: Lower NO_2 for TROPOMI is likely due to difference in time of measurements.

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

Figure 5: Please remove square from measured data for clarity. RMS looks higher than listed in the figure legend especially for NO_2. Please consider including fit residual plot as well.

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.

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

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?

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

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