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Comment on acp-2021-815

Anonymous Referee #1

Referee comment on "MAX-DOAS observations of formaldehyde and nitrogen dioxide at three sites in Asia and comparison with the global chemistry transport model CHASER" by Hossain M. S. Hoque et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-815-RC1, 2021

This paper provides an analysis of HCHO and NO2 across three diverse sites. While the data themselves are very interesting and important, the analysis lacks insight and the model used to aid interpretation is inappropriate leading to a paper that is fundamentally flawed in several respects. These include, but are not limited to, the following:

1) Authors posit a change in NO2 behavior at Phimai between this dataset and a previously published data set by the same authors (Hoque et al. 2018) with the difference being that the current dataset shows high NO2 concentrations in the wet season while the previous 2015-2016 data did not. Frankly, I do not see this difference. Looking at figure 3 from Hoque et al. 2018, the 2015-2016 NO2 data are greater than the more recent 2017-2018 data for all months. The trends are also similar, with even higher NO2 during wet season in the earlier 2015-2016 data. This leads to a somewhat unnecessary and lengthy discussion of soil moisture and associated NOx emissions that is not compelling. Even the model simulations show that month-to-month soil emissions only range from 18-24%, such that, even if soil emissions maximize in July, it is only a 5% effect overall compared to the annual average.

2) The treatment of the HCHO to NO2 ratio is not well posed. Looking at Figure 4, it is clear that the vertical gradient in NO2 and HCHO between 0-1 and 1-2 km are quite different. This difference in gradient with much larger decreases in NO2 with altitude fundamentally undermines the use of column values of the ratio as outlined in Schroeder et al. (2017; https://doi.org/10.1002/2017JD026781). Rather than following an old and flawed recipe from the Martin et al. and Duncan et al. papers, the authors would be better served to ask how their data challenges the use of the fixed range of values (VOC-limited for <1 and NOx limited for >2) that Schroeder clearly shows are not viable when more detailed information on vertical gradients is known.

3) The use of the CHASER model, given its resolution on 2.8x2.8 degrees is entirely

inappropriate for this analysis. While model suitability for all three sites is problematic, it is greatest for Pantnagar, which is located near the Himalayan foothills where large changes in elevation occur well within the local grid resolution of CHASER. While the problem is obvious from the start, the authors go through an awkward analysis of all possibilities for why the model fails, only to come to the conclusion that problems “are expected to decrease with high resolution (improved spatial resolution) simulations.” This was already a foregone conclusion. In the end, the comparison between CHASER and the MAX-DOAS data yields no useful insights in my opinion.

4) There are red flags in the analysis with regard to proper methods. One example is the comparisons between CHASER model results compared to CHASER when smoothed by application of the averaging kernel. This smoothing should not generate wholesale shifts in the data. For example, in Figure 10, the Phimai (top left) seasonal average NO2 columns from CHASER not only change by as much as a factor of three when smoothed, they also show anticorrelation in the seasonal trend. I don’t know how this is possible, and I have to assume that this is a mistake. Likewise, in Figure 8, sometimes the smoothing has almost no effect (e.g., Post-Monsoon and Winter in Pantnagar) and at other times there are dramatic changes (e.g., Winter in Chiba).

5) Finally, the language issues in the paper are simply too comprehensive to recount here. This goes beyond grammatical issues and affects clarity, especially when trying to understand how data was treated statistically. I would suggest that the authors invite a colleague to help with this as a simple language editor would almost certainly miss the nuanced language problems.

While I would normally provide a thorough line-by-line review, such detail is not warranted given the major flaws in the paper. Given the importance of the data, I suggest that the authors go back and reconsider their choice of model and approach to investigating metrics like HCHO:NO2 ratios. This data set could well challenge current thought on the value of such metrics.