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Comment on acp-2021-820

Anonymous Referee #2

Referee comment on "Source and variability of formaldehyde (HCHO) at northern high latitudes: an integrated satellite, aircraft, and model study" by Tianlang Zhao et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-820-RC2, 2021

This paper presents model evaluations of HCHO against a combination of satellite, ground, and aircraft observations in a very sensitive area but rarely studied in terms of atmospheric composition. It is in general well written and fits well within the scope of ACP. It will add important insights regarding HCHO source and variability at high latitudes. I'd recommend it for publication. A few concerns and comments are listed below for considerations for clarification or potential improvement:

1. Isoprene (and monoterpenes) may not be good tracers to directly evaluate their emissions given its short lifetime. Do MVK+MACR observations available during Atom? They would provide a more regionally representative signal for isoprene emissions.

2. For model evaluation with Atom-1: why was 1 hour averaged model output used? The model was running using 10 min /20 min time steps? Would a higher time resolution comparison better help resolve the vertical profiles?

3. Figure 1: The model shows a somewhat large underestimate of HCHO in the free troposphere and the boundary layer? It seems so too for isoprene and monoterpenes? It would be worth emphasizing as they reflect some knowledge gaps that might be the first time shown in the literature.

4. Figure 2: why were the model results for the regional average used? Any justification that it should be this way? Or is it better than using the model output for the grid cell containing the station? Given the high-resolution model, I don't quite understand why such a regional average is needed.

5. a) Figure 2 shows some interesting features of enhancements captured by the model. MAX-DOAS however seems quite a noise although it is hourly data. Can the comparison be done more quantitatively while still being able to factor in MAX-DOAS instrument uncertainty? How does the model perform in non-fire conditions vs fire influence conditions? Can any quantitative results be interpreted here? Would the MAX-DOAS be useful to compare to the TROPOMI HCHO products directly?

5. b) Line 385: if the detection limit of MAX-DOAS is 1e15 molecules cm-2, then there'd be only a few data points above the detection limit in Figure 2? Am I interpreting it correctly?

5. c) Figure 2: some panels lack y scale.

6. Fire influence in the model: Is that the fire influence in the nested domain, or is that global? Fire smoke in other regions may transport to AK and affect 2019 summer? Depending on how this sensitivity was set up (does it reflect the fire influence within the AK domain, or globally), it may be the reason why the fire VOC emission within AK is only a factor 1 to 2 higher than biogenic emissions, but $dVCD_{GC,\ Fire}$ is 10 X higher than $dVCD_{GC,\ Bio}$? i.e., Lines 510-513

7. From the comparison of TROPOMI HCHO VCD with the GEOS-Chem HCHO VCD, it seems the model is predicting HCHO well and there is no significant knowledge gap regarding HCHO from biogenic VOCs or fire smoke in Alaska? But from the comparison with Atom observations, the model seems underpredicting HCHO, while the MAX-DAOS comparison may not be too quantitative? How would these be reconciled, particularly regarding Atom and TROPOMI evaluations? Overall, I was hoping to see those evaluations could be done more quantitatively. How exactly does the model HCHO compare to observations? Does the model underpredict HCHO at the surface or throughout the troposphere, which seems to be the case when compared to Atom?

8. Lines 460-465: Here and a few other places claim the VCD is mostly driven by wildfire direct emission, rather than secondary production during fire conditions, but it is according to model sensitivity tests. The more quantitative comparison between model and observation may show the model is underpredicting HCHO vertical distribution (Item 7), and the satellite data comparison approach may be biased since it uses the model information for reprocessing (Item 9). I wonder if the observations and the model evaluations have any evidence to support that the direct fire emission of HCHO drives its VCD, rather than secondary productions.

9. a) I am a bit confused about the reprocessed TROPOMI HCHO VCD. My understanding is that it also uses information from GEOS-Chem (for a priori, background column, and AMF), and later the paper compares this reprocessed product with GEOS-Chem. Wouldn't that model information used to reprocess TROPOMI VCD cause some internal biases to the new data, so that the reprocessed product would be essentially similar to and dependent on the model? Can authors explain how it would or would not be the case, and would it

affect the interpretation of HCHO VCD evaluation? In other words, is it a fair and independent comparison? The authors seem to agree with that by stating the TROPOMI products are a 'semi-quantitative tool' to constrain fire emission, which should be further clarified

9. b) Some common practices of evaluating satellite retrievals include smoothing the model with satellite averaging kernels so that they have the same vertical sensitivity, or reprocessing the satellite data with a certain priori profile so that they reflect the measurements, rather than a priori information. It seems the model and satellite data in the work both use the same a priori and the AMF. Am I understanding it correctly? If so, how often the a priori is updated in the reprocessed product? Overall, it would be great if the method for reprocessed data can be further clarified, i.e., the exact difference between the reference sector correction of this study and the default.

9. c) The model seems to underpredict the HCHO vertical distribution relative to Atom field data, while the model is used to reprocess TROPOMI HCHO VCD. How does the HCHO underprediction relative to Atom affect the reprocessed VCD?

10. Line 100. The paper cites Liu et al. 2017 for HCHO EF. There are some new studies from recent aircraft campaigns and they seem to support around 2 g/kg for HCHO EF (i.e., WE-CAN VOC emission paper Permar et al 2021 and recent FIREX-AQ AGU conference talks?). Would EF HCHO used in the model/GFED be consistent with those recent studies? It may be able to support that the fire emission in the model is simulated well?

11. Lines 550 -555: I am not entirely sure how the uncertainties of reprocessed VCD were calculated by reading this part. Can the authors clarify it?

12. Section 2.2: how many vertical profiles were used from ATom. How could AWAS do 3-5 minutes average for isoprene or monoterpenes. That's be lots of samples for AWAS? A bit of clarification would be good.

13. Line 523: ATBD not defined until next page.