Comment on acp-2021-820
Robert George Ryan (Referee)

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-RC1, 2021

Peer review of Zhao et al, (2021), Atmospheric Chemistry and Physics

Reviewer: Dr R. Ryan (UCL)

Title: Source and variability of formaldehyde (HCHO) at northern high latitude: an integrated satellite, ground/aircraft, and model study

Author(s): Tianlang Zhao et al.

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Reviewer recommendation: Minor revisions

Comments: Overall this is an interesting and well written paper addressing VOC
production and measurement in an understudied region. I think it is suitable for publication in ACP after some minor revisions. I think the primary areas that need to be addressed are:

- The MAX-DOAS section. The MAX-DOAS geometric approximation for VCDs requires that the bulk of the trace gas column be above the scattering height. At line 373 you note that the HCHO column has a large fraction above the lowest kilometres, which already brings the validity of the geometric approximation into question. Moreover, you note that that you’re looking at presenting optimally-estimated profiles in a follow-up paper. In my view, the vertical profiling capability of the MAX-DOAS is its chief advantage as a ground based measurement technique. It could provide you with some useful information to compare in this paper to ATOM results, and then to modelled profiles. I really don’t think that geometric approximated MAX-DOAS VCDs (even if valid) are adding much to your discussion. It would be better either to leave the MAX-DOAS results out and save them for your follow-up paper, or (best case scenario!) incorporate the full optimally-estimated profiles into this paper to take advantage of the MAX-DOAS’s full capability.

- The uncertainty section (4.4). This section could be tidied up and incorporated into your other results sections. You list many examples of uncertainty in different parameters from different papers, and yet I am still a little unclear on how you arrive eventually at the 90 % and 35 % uncertainty values for fire-free and fire-influenced scenarios. It would be great to spell out exactly how you incorporate each uncertainty term to calculate the final uncertainty. I also think you should do this earlier in the results section. This would aid your discussion of agreement between TROPOMI and GEOS-Chem by allowing you to specify whether/when/where you find agreement between the two less/greater than the TROPOMI uncertainty. You could help this further by including in your map plots (Figs 4,5 and 6) difference maps (GEOS-Chem minus TROPOMI or vice versa) to visually see where the agreement is below the uncertainty and/or less than the TROPOMI detection limit.

Minor corrections:

- Abstract line 1: spell out formaldehyde for the first time in the abstract too
- Lines 31-33: remove “to” in front of all the percentage ranges
- Lines 37-38: Sentence starting with “The source…” is repetition of previous information
- Line 44: “show” not “shows”
- Line 51: remove “a significant amount of”, it is subjective without quantification
- Line 58: remove “After these biogenic... ... atmosphere”, filler and not necessary for the flow of the sentence
- Line 61: LAI already defined
- Line 68: remove “been”
- Line 77-79: Reword the sentence beginning with “This high...”. It reads like you are saying, in the end, there's an important role of climate warming on climate, which is tautological.
- Line 81: HCHO already defined
- Line 88-89: Reword. It reads like “in regions where BVOC emissions are dominated by... the variation of BVOC emissions”, again, tautological.
- Line 90: Not clear how this sentence connects to previous paragraph. For example of what?
- Line 128: remove “First”, unnecessary
- Line 131: Not sure about “accuracy”. (A) accuracy is hard to verify, as opposed to precision, and (B) I think the more important point is that ground based measurements are closer to being in-situ with, and therefore more sensitive to, the trace gas source.
- Line 134-135: MAX-DOAS measurements are also really hard to interpret in cloudy and high AOD conditions. You say so yourself later when you omit MAX-DOAS measurements from the most smoke effected periods.
- Line 167: “transfer” not “transport”
- Line 213: remove “that”
- Line 222: In the methods section, I would state that the reprocessed VCD has differences to TROPOMI VCD, rather than “advantages”. Stating “advantages” starts to confuse results with methodologies.
- Line 226: Again, save this information about how your method leads to an improvement, for the results.
- Line 238: why different averaging times?
- Line 252-253: Why average to 2 hours for a 3 hour window? Why not just average all results from 12:00 to 15:00 (if you end up keeping the MAX-DOAS results in)?
- Line 257: state why you would want to choose the highest elevation.
- Line 264: I think shift this first sentence to be the second, the second sentence of the paragraph introduces the section better.
- Line 279: “have” not “has”
- Line 282: “BVOC emissions are calculated using”, not “follows” – follows sounds jargonistic
- Line 308-309: save for results
- Line 324: Guide the reader with approximate altitude ranges in the text
- Line 339: “reproduces”, not “well reproduce”
- Line 342: “mixing ratios are” not “mixing ratio is”
- Line 346: First sentence is unnecessary and emotive, rendering the second sentence repetitive.
- Line 352: This suggests a minor contribution in most of Alaska, but perhaps not everywhere?
- Line 414: Give an example to show how “high” is “high”, perhaps by comparing to other parts of the world, to some threshold, by relationship to uncertainty or the detection limit
- Line 419: “of” not “for”
- Line 420: reword to “May to August 2018”
- Line 423: remove “largely”, it is unnecessary
- Line 424-425: You say “stems from”, but all you’ve proven is that the HCHO predominantly “resides in” the lowest atmospheric layers. In fact, you highlight the
large contributions of background methane oxidation which may not necessarily stem from the lowest layer at all – methane could be transported from long range including higher atmospheric layers.

- Line 425: can you comment on the extent to which fewer plants (presumably lower BVOCs) and more long-lasting snow (higher albedo, more retrieval problems) could contribute to the lower HCHO VCD in elevated regions?
- Line 429: What causes this enhanced methane oxidation?
- Line 443: remind the reader what a negative dVCD physically represents.
- Line 450: I'm unclear on the relationship of ideas in this paragraph. How does “widespread HCHO enhancement” follow from the first sentence, then on to saying that HCHO production is actually suppressed by low NOx levels? In addition, please clarify quantitatively what you mean by low and high NOx
- Line 472: This small section is mostly repetition of ideas in the previous paragraph, it can be incorporated or removed
- Line 489: First short sentence not needed. Also, reword the next sentence to have “in the 2019 Alaskan summer.”
- Line 492: add “the” between than and TROPOMI
- Line 497: “sources” not “source”. Also, be quantitative instead of simply saying “Much lower...”
- Line 505: Be quantitative instead of simply saying “We find little change”
- Line 511-512: Why are the biogenic emissions higher by a factor of “1-2”? You have the numbers there, surely it is larger by a factor of 498/374 exactly?
- Line 534: Don’t have “etc”, be specific.
- Conclusions: I think you want to start your conclusions with positive results, what you want people to take away from this paper, not another summary of the previous literature. Imagine you get to the end of the paper, and after all that reading the first thing you see in the conclusion is “VOC emissions... remain poorly quantified...”. No – tell the reader why the work you’ve done is great! Tell them how you’ve helped close a literature gap, don’t highlight how one is still open. To achieve that, you can significantly shorten your conclusion, cutting it to the most salient points only.