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Comment on acp-2021-672

Anonymous Referee #1

Referee comment on "Field observational constraints on the controllers in glyoxal (CHOCHO) reactive uptake to aerosol" by Dongwook Kim et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-672-RC1, 2021

This paper uses glyoxal measurements from the KORUS-AQ aircraft campaign over S Korea to show evidence for glyoxal uptake by aerosols with reactive uptake coefficient ~0.01 and accounting for ~20% of organic aerosol formation. The authors further find that aromatics are the dominant source of glyoxal, thus representing an important anthropogenic SOA formation pathway. I found the analysis to be carefully done and the results above to be of fundamental importance to support the hypothesis of glyoxal as a significant SOA precursor. The paper is overall very well written. I strongly support publication in ACP. My only significant concern is that the authors are in my opinion over-interpreting their data to reach conclusions that are featured prominently in the abstract but are not based on sound reasoning. I urge the authors to revise or delete these components of the paper, or at a minimum to address my objections in the text.

Specific comments:

- Abstract, line 28: I suggest "is an important POTENTIAL precursor…"
- Abstract, lines 32-33: I didn't find in the text a statement or reference that the glyoxal yields from aromatics are "relatively well constrained'. In fact, different aromatic oxidation mechanisms disagree in their glyoxal yields and the MCM used here overestimates the yields in chamber data (see Bates et al., ACPD https://doi.org/10.5194/acp-2021-605, Figure 5).
- Abstract, lines 36-42: in my opinion these findings are based on unsound interpretation of the data. See comments below for specifics.
- Abstract: missing from the Abstract is the glyoxal reactive uptake coefficient inferred from the data, which I think is a robust result and will be an important reason for citing this paper.
- Line 127: the authors focus on the boundary layer below 2 km and that makes sense, but they should comment on their free tropospheric glyoxal data reported in the KORUS-AQ database that are surprisingly high with relatively little variability, and imply a background source of glyoxal missing from the models. The same problem was found over the US during SENEX (with the same instrument) by Chan Miller et al. 2017 cited here. I think that the authors need to comment on this. Are their measurements

reported in the free troposphere incorrect? It would be very useful (and a relief!) for the community to know this. If they are correct, where does this background glyoxal come from? How would it contribute to boundary layer glyoxal?

- Line 203: I think you mean equilibrium, not steady state.
- Lines 246-250: I don't see differences in patterns between glyoxal, formaldehyde, and CO in Figure 1.
- Line 265: there should be some comment on glyoxal yields from the important VOC precursors. See comment on Abstract.
- Lines 275-279: this 'test' seems circular?
- Lines 317-325: I am not convinced at all about the dependence on JNO2. The effect is small and there could be any number of confounding factors.
- Lines 326-331: I am also not convinced at all about the argument for the flattening of keff as being due to higher aerosol viscosity due to high CAN. That seems really far-fetched. Again, there could be any number of confounding factors.
- Lines 333-354: that whole subsection is based in my view on flawed interpretation. I don't think that the data provide a distinction between surface- and volume-controlled uptake, because both would show a correlation with surface area. The 'hybrid treatment' is an interesting attempt to explain the glySOA formation mechanistically but it fails miserably. This is important to note but it could be due to any number of reasons.
- Lines 383-390: these conclusions are in my view not supported by the data, as detailed in my comments above.