Review of Atmos. Chem. Phys. Manuscript (#ACP-2019-931) "Constraining remote oxidation capacity with ATom observations" by Travis et al.

General Comments

In this work, measurements of OH and OH reactivity (OHR) from two Atom field deployments were used to evaluate the oxidation capacity over the remote oceans and its representation in the GEOS-Chem model. Good model-measurement agreement was obtained for OH and its precursors with over estimation of NOy to be attributed to insufficient or missing loss processes. The measured OHR below 3 km is greater than the sum OHR calculated from the measured OH reactants and the OHR in GEOS-Chem. The underestimate of acetaldehyde and peroxyacetic acid in GEOS-Chem and the reconcile of model-measurement agreement call for further work on the OH loss and production processes over land. In general, the paper is well written and within the scope of ACP. Evaluation of a transport chemical model against observations in a global scale are important and rare. I would recommend accepting it for publication after the authors address the following special comments in their revision.

Special Comments

- 1. P.2, L.1, maybe change "Organic aerosol is ..." to "The production of organic aerosols is ..." or something like that.
- 2. P.2, L.21-22: I would suggest adding "In the remote atmosphere" before "OH is primarily produced by the photolysis of ozone (O₃) in the presence of water vapor." as in polluted environments other OH sources like the photolysis of HONO could be dominant at certain times of day.
- 3. In Section 2.1: a brief discussion of the GEOS-Chem model uncertainties in simulating OHR and OH as well other important species like acetaldehyde and PAA and should be included. Coupled with the measurement uncertainties in Table 2, the combined measurement-model uncertainties can help to understand if the discrepancies in the model-measurement comparisons (Figures 3-8 & 10-14) are significant or not.
- 4. P.5, L.13 and Figure 1(a): define the altitude range of the surface layer.
- 5. P.5, L.14-15: How are three quarters and 40% calculated? Are they numerical ratios or somehow weighted? In my opinion, spatially integrated and maybe air mass weighted cOHR values (over oceans versus over land and below 3 km versus above 3 km) should be considered in terms of global oxidation capacity.
- 6. P.5, L.24: Noticed both Fig. and Figure are used. Not sure which one is required by ACP to use, but please be consistent.
- P.6, L.11: "accuracy of 1.35" is given for OH measurement: what are the units for 1.35? In Table 2, "factor of 1.35" is given for OH and HO₂ detection limit and precision but no accuracy is given. Please clarify it.
- 8. P.6, L.14: the minimal bias of <1% seems too good to be true considering the spatiotemporal variabilities in both model and measurements (see e.g., Figures 3 and S1) as well as uncertainties and discrepancies (e.g., Fig. 6-7) in the measurement and model.
- 9. P.6, L.15-16: please include standard deviations for these concentrations.

- 10. Fig. 3-7: not sure if it's going to be too messy, but it is also important to see the percentiles in the model predictions. Maybe use whiskers to show the percentiles with different altitude bins for the measurement and model so that the whiskers will not be overlapped?
- 11. P.7 L.2: change VOC to VOCs
- 12. P.7, L.15-16 and Fig. 5: the units for $i(O_3)$ should be 10^{-5} s⁻¹. Also please point out this is for the photolysis reaction of $O_3 + h\nu \rightarrow O(^1D) + O_2$ (sometimes it's called $jO(^1D)$, not the other one: $O_3 + h\nu \rightarrow O(^3P) + O_2$. I pointed this out in the initial review before the ACPD publication, but it seemed the message didn't get through to the authors.
- 13. P.7, L.17-26: there is a discrepancy as large as 30 ppb at high altitudes for winter in ATom-1 and for winter and summer in Atom-2. A discrepancy of 20-30 ppb doesn't seem "unbiased" to me. Any explanation of this overestimate at high altitudes should be briefly discussed here. This also makes me believe the above <1% bias likely to be coincident or the right answers for the wrong reasons.</p>
- 14. P.9, L.28-29: note in Table 2 an accuracy of 0.8 s⁻¹ and a detection limit/precision of 0.3 s⁻¹, which are comparable to the overall differences here. The authors should mention this to remind readers the uncertainty in the measure.
- 15. P.9 bottom and P.10 top: both r and r^2 are used for correlation. Please be consistent. In my opinion, r^2 should be used. A scatter plot of the missing OH reactivity against acetaldehyde should be included in the SI to support the strongest relationship.
- 16. P.10, L.4: ...oxygenated VOCs (OVOCs)
- 17. P.10, L.11: VOCs. There are many cases where "VOC" should be really "VOCs" and "aerosol" should be really 'aerosols". Please check this out through the manuscript.
- 18. P.10: Fig. S3 and 9: any explanation why there is a belt of enhancement over the ocean in the mid-latitude of southern hemisphere?
- 19. P.11, L. 24: change OF to of.
- 20. Table 2: in the reference for CH₄, remove AMT
- 21. Note 1 of Table 2: H₂ was measured but was set to 0.5 ppm. How does this value compare to the observations? If the difference is large, maybe use the measured value (e.g., mean) to constrain the model. From Figures 10 & 11, H₂ contributed about 5-8% of observed and modeled OHR, which is not very small.
- 22. In Supplemental Information: both ethane and propane are underestimated in GEOS-Chem (Figures S6 and S7). Is this because of unaccounted emission sources like fracking?