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Interactive comment on "Effects of heterogeneous reactions on global tropospheric chemistry" *by* Phuc T. M. Ha et al.

Anonymous Referee #2

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This manuscript presents a global model-based study of the impacts of various heterogeneous uptake processes on tropospheric composition. The CHASER model is validated against a number of ground-, ship-, aircraft-, and satellite-based observations of relevant species (NOx, PM, O3, CO, OH, and total column O3) and parameters (cloud fraction). A variety of simulations are designed to probe aerosol, cloud droplet, and ice particle uptake of N2O5, HO2, and RO2, with individual species and uptake pathways (cloud versus aerosol) turned off in turn. Total and spatially-/temporally-resolved changes in methane lifetime, NOx, O3, and CO are assessed. Finally, a sensitivity simulation is conducted to evaluate the impacts to atmospheric composition with variations in the examined heterogeneous loss rates.

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Discussion paper

The study presents a well-rounded analysis of heterogeneous uptake from a global model perspective. The model is thoroughly assessed against a reasonable number of available observations. The thoroughness of sensitivity simulations, both turning on/off all and individual heterogeneous uptake reactions as well as varying the magnitude of the first-order loss rate applied, addresses in a methodical way how this chemistry might impact global composition. I think there may be a missed opportunity in this manuscript to more thoroughly discuss the mechanisms through which these impacts manifest, but diagnostics necessary to perform such assessments may be lacking, and this should not preclude publication of this work. There is also a concern that model biases (such as the overestimation in cloud fraction in the northern Pacific lower troposphere) will introduce model-dependent errors in the results, though I regard the acknowledgment of this issue in the text as sufficient. I would consider this work as suitable for publication in this journal following the incorporation of the suggestions noted below, primarily concerned with clarifications and organization.

Major Comments

In addition to collective improvements to figure clarity and organization, noted below under minor comments, my only other major request would be to expand on discussions of the mechanisms underlying some of the changes exhibited in the presented results. For instance, the description of effects on PAN production and transport, ∼L492, could be more explicit and re-emphasized in the Conclusions. The reasons for the large increases in NOx near the surface in the Arctic during JJA due to HR(RO2) are still unclear to me – if there were a large source of NOx here, the reduction in PAN formation may make sense, but large sources at these high latitudes seems unlikely. Similarly, why are there increases in NOx during DJF due to HR(RO2) in the high latitude southern oceans, just offshore?

Similarly, one impact attributed to RO2 uptake is a decrease in CO (e.g., L257). I'm curious about the mechanism, and not aware of any discussion regarding this. I would assume that the CO decrease is due simply to reductions in secondary production –

functionalized C-containing RO2 species that would otherwise be oxidized to form CO are instead taken up on aerosols/cloud droplets. Do the authors know if this is the case, or if there is another mechanism at play?

I understand that diagnosing these kinds of questions from global model output, especially from lengthy and numerous simulations, may be difficult, given limitations on how much output can be generated. While model evidence to further describe these mechanistic questions would be ideal, hypotheses supported from prior literature or simpler, logical arguments would suffice.

Minor Comments

L120: Please add a reference for the MAC reanalysis biomass burning emissions, or else provide more detail

∼L175: Somewhere in Section 2, the timeframe of the simulations should be clearly stated. I gather from some of the time series figures that model output is available for at least 2010-2018 – were all sensitivity simulations run for this entire period?

∼L255: The discussion surrounding Fig. 3 refers to differences between the various model sensitivity runs, but it is very difficult to make out the different colored lines representing the different simulations in the figure. Perhaps an inset that shows a "zoomed in" view of a representative portion of each panel, or else plotting in different coordinates, like % difference compared to obs versus time, would help remedy the issue. This applies to Figures 4 and 5 as well.

L286: The suggestion of insufficient downward mixing of stratospheric air in the model while CO is underestimated by the model seems counterintuitive to me. Stratospheric air should be depleted in CO, so I would expect higher observed CO would point to something other than stratospheric influence, especially at the surface, as the ship-based observations are. I'm unfamiliar with Kanaya et al.; do they provide some other rationale to explain this apparent discrepancy?

L334: It is unclear to me why the “ground layer” is defined differently for all flights (> 800 hPa) versus for the N. Pacific region (> 700 hPa) in Table 8 – could the authors include a brief explanation?

L365: I feel that the TCO plots in Fig. 7 would be more easily understood by plotting Model – OMI differences, for both the STD and noHR simulations. As is, the differences between the model runs and OMI stand out far more than the differences between the STD and noHR runs. One really has to focus on small details to see where the model is improving with respect to OMI.

L466: The statement that “recent O3 increases can be attributed to reduced HO2 uptake under aerosol (PM) decreases brought about by the new Chinese Air Pollution policy,” is, I think, too strongly worded without a quantitative accounting of the observed O3 changes. Other effects, such as the non-linearity in O3 production with NOx concentration, could also be contributing. Qualifying the statement as “can be attributed in part to reduced HO2. . .” or similar, would be sufficient.

L548: “magnitude of HRs” is vague; could you clarify if this sensitivity test is meant to probe uncertainties in the first-order loss rate, possible non-linearities in the uptake, etc.?

L589: I’d suggest staying consistent with the number of significant figures reported in the % changes here, in the abstract (L13), and elsewhere. Sometimes one digit is reported after the decimal place, sometimes two, and sometimes none.

Figs. 10 and 14: x-axis labels would be helpful, for anyone who may miss the (%) in the title.

Technical corrections

L130: “uncertainties” should be “uncertain”

L293: “undervalues” is a slightly out-of-place word choice; “underestimates” may be better

L297: The use of “extends” here suggests that model underestimates are extending in time/space instead of getting worse. “worsens” or “exaggerates” may better reflect the intended meaning.

L301: A verb is needed in this sentence; “Ocean is mostly dominated. . .”

L325: “However, for the. . .” the “for” is not needed.

L326: Here and elsewhere, I’d suggest the authors check for consistency in how “ATom-1” is capitalized and punctuated.

L364: The order of appearance of the Supplemental figures should match the order in which they are mentioned in the main text.

L380: “surface aerosol density” should be “surface area density”

L415: “preferably onto” confuses the meaning of this sentence; I suggest “rather than onto” to emphasize the importance of aerosol uptake over cloud uptake

L503: “glob” should be “globe”

L502: Fig. 14 is introduced here before Fig. 13 is discussed; I’d suggest switching the two.

L536: The phrase “N2O5 uptake on aerosols are mostly ascribed” would be more easily understood as “N2O5 uptake is mostly ascribed to aerosols”

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