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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Constraining remote oxidation capacity with ATom observations" *by* Katherine R. Travis et al.

**Anonymous Referee #2**

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General Comments

The article "Constraining remote oxidation capacity with ATom observations" by Travis et al., submitted for publication in Atmospheric Chemistry and Physics, presents an evaluation of the hydroxyl radical (OH) and OH reactivity (OHR) measured during the Atmospheric Tomography Mission (ATom) against the GEOS-Chem global chemical transport model. While OH is generally well-modeled in the remote regions sampled by ATom, notable biases occur in wintertime Northern Hemisphere NOy and in OHR in the lowest 3 km. Multiple explanations for these discrepancies are discussed through literature review and explored through model sensitivity simulations.

The submitted manuscript provides a wide-ranging examination of the complex, timely topic of tropospheric oxidizing capacity. While the study does not provide any strong



evidence for solutions to the outstanding questions regarding NOyÂň and OHR biases, it presents a helpful survey of all the issues and demonstrates how a number of them may be further explored in a global modeling framework. Also, the conclusion that GEOS-Chem does "not show systematic bias in the simulation of OH or the drivers of remote OH production" is an important one, suggesting that persistent biases in models' globally integrated methane lifetime must instead be driven by simulated OH abundances over land, as the authors point out. Because the topic is highly relevant, the conclusions are sound but not overstated, and the improvements I would suggest are only moderate, I would suggest publication in ACP once the items noted below are addressed.

Specific Comments

The Introduction does not acknowledge the Thames et al. (ACPD, 2019) manuscript, cited later on Pg. 11, which has a likelihood of being published prior to the finalization of this submission. That paper also discusses the ATom OHR measurements, albeit with a focus on the MBL instead of global oxidation capacity. Since the Thames et al. paper will be so closely related to this one by Travis et al., a discussion of its findings and how Travis et al. will complement Thames et al. is warranted here.

Pg. 2 L. 29: The authors acknowledge "the persistent CO underestimate in models" yet do not go on to evaluate this large sink of OH. A figure analogous to Fig. 3, showing CO comparisons between model and observations should be included and discussed. Does the reasonably accurate OH field within this GEOS-Chem simulation translate to similarly well-simulated fields of CO over the oceans? Or does longer-lived CO have the imprint of biased continental OH, to which the authors refer?

Pg. 4 L. 3: Could you specify whether the methane concentration boundary condition varies with latitude and/or longitude? And, since they derive from monthly observations, is it correct to assume that the boundary condition changes from month to month?

Pg. 4 L. 30: Please describe how exactly the tropospheric mean OH is being calculated. As Lawrence et al., 2001 explain, there are multiple ways to weight this calculation, and, for the purposes of facilitating comparisons of these values between studies, an explicit definition of this metric should be included in each paper that discusses it.

Figures 3-8, 12-14: Please consider trying to visualize not only the 25th/75th percentiles for the observed median profiles, but also for the modeled profiles. How well the spread of each of these quantities agrees can be instructive as well.

On organization: I found, reading through the paper, that the topics of various results/discussion sections (Sections 3-6) jumped around quite a bit. For instance:

-Some discussion of the literature on acetaldehyde is initiated in Section 5 (mention of Read et al., 2012 on Pg. 10, L.4), mentioned again farther down on the page (Pg. 10, L. 29), and continued throughout Section 6. I would suggest consolidating the discussion of the acetaldehyde literature in one place, and perhaps making Section 6 a subsection of Section 5.

-Similarly, the discussion of NOy as a proxy for OH secondary source NOx is understandable, given the issues with measured NO2, but the discussion necessarily turns to HNO3 evaluation, all under Section 4: Constraints on the remote source of OH. Generally, HNO3 is viewed as a sink for OH, so this further contributes to the feeling of "jumping around" between topics.

Additional subsections and improvements in framing the discussion should help to give a more logical structure to these sections .

Technical Corrections

Pg. 2 L. 1: The sentence starting "Comparisons" is a run-on; either include a comma between "aerosol but" or separate into two sentences

Pg. 2 L. 5: Run-on sentence; place comma between "sources and" or split to two sentences.



Pg. 2 L. 20: Run-on sentence; place comma between "atmosphere and" or split.

Pg. 4 L. 1: Place comma after "Sherwen et al."

Pg. 4 L. 25: MCM v3.3.1 has an additional reference, regarding the update from v3.2: Jenkin et al., 2015

Pg. 4 L. 29: Figures should generally be numbered in the order that they appear in the text, even Supplemental figures. Fig. S8 should be moved to S1. Same with Tables (S4 and S5 appear before S1), and Fig. S9 (appears before S5).

Pg. 5 L. 13: Could the authors please state the number of species that are listed in Table S1?

Pg. 6 L. 6: "attitude" should be "altitude"

Pg. 6 L. 11: Should there be units for the accuracy value provided here (molec cm–3)?

Pg. 6 L. 17: Please specify if Fig. S1 shows in situ OH concentrations of column averaged. If it is column averaged, please use the OHcol notation in the text at this location and in the figure.

Pg. 6 L. 22: Instead of "successful" and "success" here, simulation should be described as having "good agreement" or similar wording.

Pg. 7 L. 10 & 15: Replace "successfully"

Pg. 7 L. 30: Run-on sentence; place comma between "2018) and"

Pg. 8 L. 10: Anderson et al. 2014 also indicated a bias in the anthropogenic NOx inventory; please cite that paper here as well.

Pg. 8 L. 18: "higher larger ozone" seems redundant

Pg. 8 L. 21: "free tropospheric" should be "free tropospheric bias"?

Pg. 9 L. 21: "We compare OHR. . ." I would suggest explicitly stating here that "OHR"

refers to directly measured OHR, to avoid confusion.

Pg. 11 L. 4: Thames et al. (2019) does not appear in the reference list.

Pg. 11 L. 17: ". . .when the lifetime of CO is long." I would consider this circular reasoning; the reason the lifetime of CO is long in the wintertime is because OH concentrations are low.

Pg. 11 L. 24: "OF" should be "of"

Pg. 11 L. 30: Nicely et al. (2016) also recognized the importance of acetaldehyde in explaining model vs. measurement-constrained OH differences, could be cited here.

Pg. 12 L. 23: It would be helpful to state, quantitatively, how large the model bias in PAA is.

Pg. 13 L. 9: It is unclear what the percentage values provided in parentheses refer to– are they percent increases in acetaldehyde from corrections to model ethane/propane, or are they percent yields of acetaldehyde per molecule of ethane/propane oxidized?

Table 2: Please number the superscripts in the order they appear in the table.

Figure 5: Units for jO3 should be 10–5 s–1 instead of 105. Would also be helpful to specify whether this is j(O3 –> O1D + O2) or j(O3 –> O3P + O2).

Fig. 11: I appreciate the difficulty of finding unique color choices for a figure like this, but I find the two shades of green, representing MHP and HCHO, practically indistinguishable on my computer screen (so the problem is likely worse in hard copy). Please adjust one of the two.

References

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