

Atmos. Chem. Phys. Discuss., referee comment RC2 https://doi.org/10.5194/acp-2022-556-RC2, 2022 © Author(s) 2022. This work is distributed under the Creative Commons Attribution 4.0 License.

Comment on acp-2022-556

Anonymous Referee #2

Referee comment on "Reconciling the bottom-up and top-down estimates of the methane chemical sink using multiple observations" by Yuanhong Zhao et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-556-RC2, 2022

This paper aims to improve "bottom-up" estimates of OH concentrations by constraining chemical model simulations with observations of OH precursors. The paper is thorough, novel, well-written, and tackles a very important issue in atmospheric chemistry. I recommend it for publication in ACP, subject to some relatively minor corrections.

General comments

A simplified 0D model of atmospheric chemistry is used, gridcell-by-gridcell to determine the how the OH fields from a global 3D photochemical model would be adjusted by incorporating observations on OH precursors. One thing I felt was missing from the paper was a comparison of the OH fields predicted by the simplified model to that of the "parent" 3D model (i.e., how does [OH]_DSMACC_ref_model compare to [OH]_model, using the terms from eq. 1?). It seems that this is important because large differences could lead to non-linear effects that could influence the results. Perhaps some simple comparisons could be presented in the Supplement.

- On first reading, I was confused by the definition of the term: "[OH]_trop-M". When it is first introduced, both on line 44 and line 273, it is defined as a global value. E.g. "global tropospheric mean OH concentration" on line 44. However, it is later used to show regional distributions. I think that the authors are using [OH]_trop-M to mean something like "column average airmass-weighted [OH]", which is then sometimes averaged to produce a "global mean [OH]_trop-M"? I think the terminology needs to be tightened up a little here.
- L83: "Such MCF-based top-down methods have..." rather than "method has".
- L105 107: I don't think these papers show that decreased [OH] can explain the resumed CH4 increase. Both have a high degree of uncertainty (such that no OH change is within the plausible range), and Rigby et al., 2017 has a coincident CH4 emissions increase in their maximum-likelihood estimate. I would perhaps keep it more general and say that these papers indicate that MCF-based top-down methods indicate that [OH] changes may have influenced recent CH4 trends, although with a high degree of uncertainty.
- L109: I don't think the models show a monotonic increase in [OH], do they? i.e, does the use of "continuous increase" need to be softened to "decadal trend" or similar?
- L111 125: It seems that the Nicely et al. (2017; 2018) papers would fit into the discussion here too?
- L221: "... DSMCC is/was run forward" (insert is or was)
- L223: "DAMSCC" should be changed to "DSMACC"
- L235: "observation-based", rather than "observational-based"
- L282 (and 315, 316 and 325): To improve readability, and given that it is only mentioned a couple of times, I suggest just referring to Spivakovsky et al. each time, rather than defining another term (S2000).
- L300: should this by [OH]_Trop-M, rather than [OH]?
- L307: "which is larger than that over ..." (remove "the")
- L375: "over the 15 60N region" (insert "region" or similar)
- L384: "... by 0.07, but still cannot explain..." (insert "but")
- L395: "NO2 results in a positive bias" (insert "a")
- L404: Remove "The" from the start of the second sentence, or add "model" after "CESM1 CAM4-chem"
- L481: "... loss of CH4 in the previous GCP..." (add "the")
- L522: "... respectively, dominating the bias" (dominating, rather than "dominant")
- Section 4 (Conclusions): This section could be made more concise and readable. I suggest thinking about the paragraph structure so that ideas are grouped together more concisely and start each paragraph with a sentence describing the main point of the paragraph (at present lots of paragraphs start with phrases like "In addition", or "Also", which don't help to orientate the reader).
- L526: add "major" before "global CH4 sink", to make it clear that you're referring to one of the methane sinks (i.e., you're not also investigating CI, etc.)
- L586: "Such a difference is partly attributable to..." (remove "be")
- L593: Remove "In addition"
- L627: Remove "Also"
- Figure 5: Consider making this a 2-panel plot (well, really a 6-panel plot) merged with

Figure 2.