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Discussions

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Comment on acp-2022-556

Anonymous Referee #1

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-RC1, 2022

This is a very interesting study, presenting a relatively simple approach to correct 3D CTM generated tracer fields using a box model and satellite observed photochemical trace gases, leading to global mean OH estimates and interhemispheric OH differences that are closer to those derived from MCF. This is a very encouraging finding, suggesting that our understanding of photochemistry and methyl chloroform are good enough to allow a reduction in the uncertainty of OH. That is, if the two selected models are representative of that uncertainty, which is limited by n=2, meaning that the convergence between model and MCF derived constraints on OH might still arise from a fortunate coincidence. Nevertheless, the results look promising enough to investigate further.

The paper is very well written and with a logical story line and results that can rather easily be understood. In part this is due to the choice for a level of detail that keeps the focus on the main findings. This is good, however, some important details are missing that would be needed for someone to be able to repeat what was done. In addition, the validity of some assumptions should either be tested or discussed in further detail as explained below. With those issues addressed, which will at most call for moderate revisions, the paper should be acceptable for publication.

Scientific comments

Line 130-132: What is missing here is the use of chemical data assimilation, which is trying to achieve the same as this study, but through a more formal data assimilation procedure. A brief discussion is required of the relation between such methods and the method proposed here. The results should also be put in perspective of what has been achieved, or is achievable, using such methods.

Line 135: Why were CESM1-CAM4chem and GEOSCCM chosen from the CCMI-1 ensemble? What makes them representative members?

Line 150: Data availability is less relevant than the time window of the data that was actually used. Only towards the end it became clear that only the year 2010 was used for the observation-based box model calculations. Does that mean that only 2010 O3 data were used? This should be clear for other compounds also.

Line 152: How is the troposphere defined in the model? How about the vertical O3 gradient within the troposphere in the application of the box model. Is the tropospheric mean applied to all tropospheric levels? Is there any use of averaging kernels? If not, how consistent is the observational adjustment of vertical profiles?

Line 152: How are the constraints from total column O3 and tropospheric O3 combined in a box at a given level in the troposphere?

Line 164: How is the planetary boundary layer defined in the analysis? Since the sensitivity of the NO2 retrieval does not stop abruptly at the top of the PBL, to which altitudes is it applied and how is the sensitivity of satellite retrieved NO2 to the free troposphere accounted for?

Line 221: This assumes that the photochemistry is in diurnal steady state at the time when satellites measure the atmosphere. What supports this assumption?

Line 231: Why are monthly means chosen if the satellite sampling is restricted to daytime satellite overpasses? How can these two be compared?

Line 235: How are satellite data that represent sub-column averages with variable vertical sensitivities regridded in the vertical? What happens if the set of observations that is imposed to the box model (as I understand it) is inconsistent with the photochemistry scheme? Is there some nudging involved, or how do you prevent that non-observed compounds do not end up in an unstable solution?

Equation 2: This equation assumes that the full 3D OH_model for 2010 that is supposed to be represented by OH_DSMACC_REF_MODEL indeed match each other on the monthly mean time scale for 2010. I did not find any evidence that this is the case, or the extent to which this requirement is satisfied.

Line 258: Does ‘I’ run over the troposphere or the entire atmosphere? Equation 4 suggests the troposphere, but equation 7 the whole atmosphere (for the global CH4 burden). This should be clarified.

Line 283: In the TRANSCOM-CH4 experiment a scaling factor of 0.92 was applied to the Spivakovsky fields based on a MCF inversion by Krol et al.

Figure 1: How realistic are the OH holes over tropical rainforests given what is known about radical recycling under low NOx conditions?

Line 341: This is a surprising finding, especially since there must be correlated regional adjusments in for e.g NOx and CO. The reason could be that the adjustments are small enough. The size of regional adjustments is not shown, but could be quite substantial. The statement that the non-linearity of photochemistry is negligible globally should be backed up by a test that it is significant regionally, which we know it is. If it is not, then I wonder what is going wrong.

Line 486: Here the reader should be reminded that this holds for the period 2000-2009.

Technical corrections

Line 390: “northern China” i.o. “North China”

Line 420: “the” i.o. “such as”

Line 453: “limited’ i.o. “a few”

Line 481: “in the previous” i.o. “in previous”

Line 541: “molec cm-3” i.o. “moelc cm-3”

Line 545: “krol” i.o. “korl”