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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "A 3D-model inversion of methyl chloroform to constrain the atmospheric oxidative capacity" *by* Stijn Naus et al.

Anonymous Referee #2

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The paper presents an estimate of methyl chloroform (MCF) tropospheric loss and emissions using a global 3D model and a 4DVAR data assimilation system. The subject is important, because MCF has been used extensively, mostly in 2D box models, to estimate the magnitude and variability of global mean concentrations of the hydroxyl radical (OH), which is responsible for removing gases such as methane from the atmosphere.

The work is timely and it is important, given the current uncertainty surrounding the global methane budget and the fate of other atmospheric constituents. I was very much hoping that this paper would provide a valuable new constraint on this complex problem, because the model and inverse approach is well established. There is a lot to like in this paper, and in many respects, the authors have done a very thorough

investigation into the solution they have found.

However, I am concerned about the stated lack of convergence in the main results. I believe that this substantially reduces the confidence we can have in the results and conclusions and implies that the main results are not reproducible. This lack of convergence is investigated in Section 3.5, and it is stated that of the three main inversions exhibited a different level of convergence. The presented results are therefore somewhere between the prior and posterior solution, but we don't know where. For some of the inversions, the "main" results are substantially different from the converged posterior, as shown for a subset of years in Figure S6. Put another way, the solutions presented are not consistent with the data, the cost function and the prior assumptions. Of course, if my understanding is correct (and please clarify it not), this would imply that the results of this paper would not be fully reproducible, even if I used the same data, model, cost function and priors.

Of further concern, the reason given for presenting a partially converged solution is that the fully converged solution does not look physically realistic. I think there are at least two problems with this reasoning. Firstly, it is not clearly articulated why these solutions are necessarily unphysical (large changes to the priors are identified, but it is not stated why these cannot occur). Secondly, it is surely not a valid approach to prematurely stop your inversion midway down its descent because you don't like what you see at the bottom. Wouldn't the preferable approach be to interrogate the physical model, the observations or the uncertainty assumptions, to understand why the converged solution is the way it is and try to formulate an alternative which provides a more physically reasonable result?

The authors do present a set of solutions which have converged, but only for a subset of years (10 years, as shown in Figure S6). If no further runs of the model are possible for the full 20 year period, I suggest the paper be refocused only on those years where a converged solution is found. To me, it would seem preferable to present a converged solution over a shorter time period, than a longer solution that could is not consistent with the data, model and uncertainty assumptions.

As I said, there is a lot to like in this paper, and I hope the authors can fix these convergence issues and resubmit.

I have some other general points, which I hope are helpful:

1. The importance of using ocean fluxes that account for absorption and reemission, compared to a 1st order loss has been well known for almost 20 years, at least for the overall MCF trend, and particularly during the period where emissions were changing rapidly. This article presents a nice demonstration of the influence of different ocean flux parameterizations on the meridional gradient. However, given that it is well established, I'm puzzled as to why the more realistic ocean fluxes weren't used in the main inversions? It would surely be preferable to rerun the inversions using a more accurate ocean flux estimate, precisely because, as the authors show, they can influence the solution in important ways. Further, as an aside, the type of ocean flux parameterization used in the inversions (i.e. 1st order loss) does not seem to be explained in the main text but needs to be specified.

2. A main conclusion of the paper is that the variation in oxidation magnitude is small (< 3% per year). This does indeed seem to be the case from the point of view of the standard deviation in the solution. However, some year-to-year changes in fact seem to be very large. For example, sometime around 2010 – 2012, the REF inversion shows a change from ∼-5% to ∼+5% compared to the prior (Figure 1). Wouldn't a change in tropospheric oxidation of 10% over 2 years actually be considered quite substantial, and have major impacts on, for example, the global methane budget?

An additional point: converged solutions in Figure S6 seem to show, in general, more variation than the unconverged "main" results. So, again, it would be important to investigate more fully how sensitive this main conclusion is to the lack of convergence in the main results.

3. If emissions are being derived in the main inversions, why was it necessary to "pre-optimize" the emissions, assuming constant loss? What happens if you don't do this? If this changes the result substantially, I'd be very concerned, as you're essentially using the observations twice, and, in the first step, you're fixing one of the parameters that you are trying to infer in the second pass. If it doesn't change the results substantially, then wouldn't this step be unnecessary?

Minor comments

L7: "... better reproduce..." (than what?)

L18: "the signals are small". This is too vague. Need to specify what quantities are being referred to.

L19: "... better match the global MCF observations..." (than what?)

L33 – 35: citation needed.

L48: What does the phrase: "but artifact-free sampling has become more difficult" mean?

L91 and throughout: phrases like "such as OH" need to specify that it is OH concentration that is being referred to. Perhaps define "[OH]".

L155: Can you be more specific than saying that model error was "proportional to the 3D spatial gradients". I.e. define what you mean by spatial gradients and let us know if there was a constant of proportionality.

L180: "... correlation with the REF inversion" (need to state what the correlation is with respect to)

L195: I'm not sure what you mean by "would be hard to exclude from a bottom-up perspective". Can you be more explicit?

Figure 4: There seems to be a consistent increase in the mismatch in the IH gradient

around 2013. Any idea what causes this? Seems like a potentially interesting feature. Could this point to a sudden increase in emissions?

Figure 5 and throughout: For the reader not familiar with these site codes, it would be useful to clarify where these stations are (i.e. their latitude is particularly important for the discussion of Figure 5).

L374-375: It would seem important to include the results of this test (scaling global OH) in the supplement.

L379: “These results indicate substantial robustness of the derived OH variations”. I’m wondering if this can really be stated so strongly, given that the three main inversion show different OH variations?

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