Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-624-RC1, 2020

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 #1

Received and published: 5 September 2020

General comments: This paper conducted a 3D model inversion of global MCF emissions and OH concentrations for 1998-2018. The inversion results show that the interannual variations of OH are small and there is no significant long-term trend in OH. If the main conclusion is correct, it can be an important contribution to our understanding of the global CH4 budget. Overall, the paper is addressing important questions and within the scope of ACP. However, some questions need to be clarified. I would like to recommend the publication of the manuscript if the following questions are addressed.

Major comments: 1) The inversion set-up. Unlike previous studies (e.g. Bousquet et al. (2005)) that together optimized MCF destructions by OH and the ocean sink, this study only optimized OH and applied the first-order ocean flux. Thus the inversion results may be largely impacted by the uncertainties in the prescribed ocean flux. Before inver-

sion, the prior emissions were pre-optimized to fit the global mean MCF mole fractions. It is not clear why the emission needs to be pre-optimized. Will the pre-optimization reduce the OH variation estimated by the inversions since the MCF emissions already fit the observations? The study assumed a 50% error for MCF emissions. The MCF emissions become small after 1997. In Turner et al. (2017), the error for the MCF emission in the northern hemisphere is set to no less than 1.5 Gg/y. Will the assumption that prior MCF emission error proportional to emissions lead to underestimation of the prior error?

2) The optimized OH is not well presented in the results. Only the interannual variations weighted by temperature were shown in Figure 1 and Figure 2. For the posterior OH, what are the global tropospheric mean OH concentrations and the corresponding CH4 lifetime? Are the latitudinal distributions consistent with previous studies? What is the N/S ratio? In the inversion TM5OH, the prior OH field shows higher concentrations over the northern hemisphere. Is the inversion using two different prior OH distribution estimated similar posterior latitudinal OH distributions? I think these values are also worth discussing.

3) The inversion results show small OH interannual variability and no significant OH trend. This is different from previous top-down studies using the two-box model inversion (Rigby et al., 2017; Turner et al., 2017). The author shows that the interannual variation can be supported by the negative correlation with the ENSO cycle. The large negative OH anomaly during 1998 (El Niño year) has been proven by several previous studies (Bousquet et al., 2006; Butler et al., 2005; Nguyen et al., 2020; Zhao et al., 2020). However, here the inversion shows a large positive anomaly in 1998. Besides, how to explain is a large positive anomaly in 2012?

Other comments: L75: What is meant by "the most promising period in its measured history"?

L110: Are the interannual variations of stratospheric photolysis considered over the

inversion time period?

L118: The 10% error in latitudinal OH may be underestimated. Usually, the error for global annual mean OH is given by ∼10%. But for the monthly mean OH averaged for 5° latitude, the error can be much larger (e.g. Naik et al., 2013; Zhao et al., 2019).

L105-115: How you get the initial conditions for MCF? Are the initial conditions also optimized?

L128-L133: Are the MCF emissions also pre-optimized in POP and TM5OH?

L175: The results during the spin-up and spin-down period are not significant, I suggest the author remove the results of the corresponding period.

L196: The top-down estimated emissions and OH variations also depend on the variations of observed atmospheric MCF concentrations and the reaction rates (temperature) of MCF with OH. It is not clear for me why the small correlation coefficient between OH and MCF variations can reflect the OH and MCF are independently derived.

Figure 4: The MCF emissions are pre-optimized to reproduce the global mean MCF mole fractions, why there are still very large mismatches between the model simulated and observed MCF mixing ratios (dash line)?

In Figure 3, Figure 4, and Figure 5, the line color corresponding to each inversion experiment is different. I suggest using the same color for each experiment in different figures.

L254-L255: Is the bias in OH vertical distributions also contribute to the model-measurement mismatch?

L277: Why MEI should lead by one year? Is this mean that the OH should show a negative anomaly one year after the El Niño? This is not consistent with the explanation in L283-L286 and previous studies (Nguyen et al., 2020; Zhao et al., 2020), which show negative OH anomaly during El Niño years.

L349: What are the criteria for stopping the iteration? Are the 3 inversion experiments reach a similar value of cost function J (and the gradient of the cost function) in the last iteration?

L352: Why further convergence will result in less realistic OH variations?

L372: “Firstly...we generally identified similar tendencies in each.” Figure 1 has shown the variations of OH estimated by three inversions are quite different.

L374-379: Here the manuscript tried to prove the robustness of the OH interannual by an additional inversion and a forward simulation. But the details of the two experiments and the results are not given. I suggest include some details in the supplements. E.g. how the one global scaling factors compare with the REF, POP, and TM5OH? Is the forward simulation use the prior or optimized MCF emissions? I cannot understand the logic here, can you clarify why the two experiments can indicate the robustness of the derived OH variations?

Reference Bousquet, P., Hauglustaine, D. A., Peylin, P., Carouge, C., and Ciais, P.: Two decades of OH variability as inferred by an inversion of atmospheric transport and chemistry of methyl chloroform, Atmos. Chem. Phys., 5, 2635-2656, 10.5194/acp-5-2635-2005, 2005. Bousquet, P., Ciais, P., Miller, J. B., Dlugokencky, E. J., Hauglustaine, D. A., Prigent, C., Van der Werf, G. R., Peylin, P., Brunke, E. G., Carouge, C., Langenfelds, R. L., Lathiere, J., Papa, F., Ramonet, M., Schmidt, M., Steele, L. P., Tyler, S. C., and White, J.: Contribution of anthropogenic and natural sources to atmospheric methane variability, Nature, 443, 439-443, 10.1038/nature05132, 2006. Butler, T. M., Rayner, P. J., Simmonds, I., and Lawrence, M. G.: Simultaneous mass balance inverse modeling of methane and carbon monoxide, Journal of Geophysical Research: Atmospheres, 110, 10.1029/2005jd006071, 2005. Rigby, M., Montzka, S. A., Prinn, R. G., White, J. W. C., Young, D., O'Doherty, S., Lunt, M. F., Ganesan, A. L., Manning, A. J., Simmonds, P. G., Salameh, P. K., Harth, C. M., Muhle, J., Weiss, R. F., Fraser, P. J., Steele, L. P., Krummel, P. B., McCulloch, A., and Park, S.: Role of atmo-

spheric oxidation in recent methane growth, Proc Natl Acad Sci U S A, 114, 5373-5377, 10.1073/pnas.1616426114, 2017. Turner, A. J., Frankenberg, C., Wennberg, P. O., and Jacob, D. J.: Ambiguity in the causes for decadal trends in atmospheric methane and hydroxyl, Proc Natl Acad Sci U S A, 114, 5367-5372, 10.1073/pnas.1616020114, 2017. Naik, V., Voulgarakis, A., Fiore, A. M., Horowitz, L. W., Lamarque, J. F., Lin, M., Prather, M. J., Young, P. J., Bergmann, D., Cameron-Smith, P. J., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R., Eyring, V., Faluvegi, G., Folberth, G. A., Josse, B., Lee, Y. H., MacKenzie, I. A., Nagashima, T., van Noije, T. P. C., Plummer, D. A., Righi, M., Rumbold, S. T., Skeie, R., Shindell, D. T., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., and Zeng, G.: Preindustrial to present-day changes in tropospheric hydroxyl radical and methane lifetime from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), Atmospheric Chemistry and Physics, 13, 5277-5298, 10.5194/acp-13-5277-2013, 2013. Nguyen, N. H., Turner, A. J., Yin, Y., Prather, M. J., and Frankenberg, C.: Effects of Chemical Feedbacks on Decadal Methane Emissions Estimates, Geophysical Research Letters, 47, e2019GL085706, 10.1029/2019gl085706, 2020. Zhao, Y., Saunois, M., Bousquet, P., Lin, X., Berchet, A., Hegglin, M. I., Canadell, J. G., Jackson, R. B., Hauglustaine, D. A., Szopa, S., Stavert, A. R., Abraham, N. L., Archibald, A. T., Bekki, S., Deushi, M., Jöckel, P., Josse, B., Kinnison, D., Kirner, O., Marécal, V., O'Connor, F. M., Plummer, D. A., Revell, L. E., Rozanov, E., Stenke, A., Strode, S., Tilmes, S., Dlugokencky, E. J., and Zheng, B.: Inter-model comparison of global hydroxyl radical (OH) distributions and their impact on atmospheric methane over the 2000–2016 period, Atmos. Chem. Phys., 19, 13701-13723, 10.5194/acp-19-13701-2019, 2019. Zhao, Y., Saunois, M., Bousquet, P., Lin, X., Berchet, A., Hegglin, M. I., Canadell, J. G., Jackson, R. B., Deushi, M., Jöckel, P., Kinnison, D., Kirner, O., Strode, S., Tilmes, S., Dlugokencky, E. J., and Zheng, B.: On the role of trend and variability of hydroxyl radical (OH) in the global methane budget, Atmos. Chem. Phys. Discuss., 2020, 1-28, 10.5194/acp-2020-308, 2020.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-624,

2020.