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Interactive comment

Interactive comment on "Influences of hydroxyl radicals (OH) on top-down estimates of the global and regional methane budgets" by Yuanhong Zhao et al.

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This study provides important evidence to the importance of improving our estimates of the tropospheric OH sink in other to accurate quantify the CH_4 budget. However, I believe that there are three main aspects this study that could make this study have a much stronger impact:

1. The study found the largest absolute OH induced differences for Inv1 over northern South America, South Asia and China and at gridcell level over South America, Central Africa, East and South Asia, and mainly for wetlands, and agriculture and waste. While, it is already explained that the distribution of sampling stations





is one of the reasons for this. This and further reasons for the larger uncertainty in the Tropics were discussed in detail in Bousquet et al., 2011 (another paper from this groups which I think should be reference at that this point in the paper). Furthermore, there are not only less sensitivity to observations in the Tropics, but also larger uncertainty in the fluxes. As a consequence, the inversion fits in everything in the Tropics which is too costly to accommodate elsewhere. Unfortunately, the Tropics is also the region where most of the OH reaction occurs. Therefore, it is very difficult to make conclusions on how the estimation of Tropical fluxes is affected by the OH assumptions on a regional level. I believe the study is missing either one more scenario where the uncertainties for each source are uniform globally, e.g. 5 nmol m⁻² s⁻¹ for wetlands (if there are wetland emissions in the gridcell), and/or include the analysis of the uncertainty reduction and posterior correlations, to determine how well resolved are these regions.

2. The main goal of using an inversion is to find the fluxes that best explain the observations. However, we do not get to see how well the observations are fitted by the inversions with the different OH fields. Therefore, we cannot evaluate which features of the different OH distributions are realistic. By knowing for example the spatial distribution of the residuals, or of the correlations between posterior mixing ratio and observations, we can evaluate if certain spatial patterns are realistic. Also the use of aircraft profiles for validation, e.g. over the Amazon (Miller et al., 2007, Beck et al., 2012, Gatti et al., 2015, Basso et al., 2016), Asia (Brenninkmeijer et al., 2007, Baker et al., 2012, Schuck et al., 2010) or across latitudinal transects (e.g. Wofsy, 2011 and Schuck et al., 2012) could provide information on the realism of the vertical distribution. During the period of the simulation, there were two satellites sensors available SCIAMACHY and IASI with distinctly different sensitivities. SCIAMACHY is more sensitive to the surface, while IASI to the upper troposphere. Using this, it may be possible to say something about how realistic is both the horizontal and vertical distribution

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of OH fields. The validation with methylchloroform measurements (CH³CCl³) as an anthropogenic tracer is also very important, in order to know which magnitude of OH makes sense and which N/S ratio.

3. Link to the validation, there is little discussion on the features of the OH fields provided by the models. For example, Patra et al., (2015) determine that observations of CH³CCl³ support a N/S gradient of ~ 1, so more should be done to explain how probable bias in the modeled OH distributions affects the CH₄ estimations. Also many of the different features in the spatial distributions OH are caused by known biases in the climate chemistry models, e.g. the NMVOC levels, the CO burden, CO biases, O3 biases (e.g. Naik et al. 2013, Shindell et al., 2006). Here, it would be very interesting to see, for example, if there is a relationship between the N/S ratio of the OH distributions and the N/S ratio of the posterior fluxes (similar to figure 2). Also, why are SOCOL3 and MOCAGE such outliers?

Some additional comments,

- We are shown inversions with and without interannual variability in the OH fields. However, due to the increase of tropospheric temperatures, even in the simulations with fixed OH or the fields distributed in the TRANSCOM-CH4 experiment, the lifetime of CH₄ will decrease. This effect is not quantified in the paper unless I missed it.
- As stated in the study, the transport model uncertainty is very large. This means that the distribution of CH₄ is model dependent. Therefore, there could be a large uncertainty in the global OH means weighted by the CH₄ reaction. I believe an airmass or volume weighted OH means should be at least provided in the supplement and that the comparison with box models or with other models should be done with air mass or volume weighted means, including the relationship in figure 2.

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• On which basis did you choose only 7 of the 20 CCMI simulations?

Minor edits and information display.

- You mentioned the TRANSCOM-2011 project. However, this was actually known as the TRANSCOM-CH4 (Patra et al., 2011), since there have been several TRANSCOM projects mainly with CO₂. It might be useful to mention, that in Patra et al., (2011), the OH fields from Spivakovsky (2000) were scaled to match the CH³CCl³ decay by Marteen Kroll in the TM5 model.
- Table 1 and table 2 are missing the units
- Could you specify which convection parameterization is used? In Locatelli et al., (2015) three parameterizations are used.
- I think that at least in the supplements you should include the maps of the mean differences between the scenarios, e.g. $E_{c}hange_{a}ll, E_{c}hange_{f}ixoh, E_{c}hange_{v}aroh.$
- In figures 4 and 5, would it be possible to show the a priori uncertainties as error bars? In general I find the double axes confusing and maybe a single axis with absolute emissions would be better.

References

Baker, A. K., Schuck, T. J., Brenninkmeijer, C. A. M., Rauthe-Schöch, A., Slemr, F., van Velthoven, P. F. J., Lelieveld, J. (2012). Estimating the contribution of monsoon-related biogenic production to methane emissions from South Asia using CARIBIC observations. Geophysical Research Letters, 39(10), n/a–n/a. http://doi.org/10.1029/2012GL051756



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Basso, L. S., Gatti, L. V., Gloor, M., Miller, J. B., Domingues, L. G., Correia, C. S. C., Borges, V. F. (2016). Seasonality and interannual variability of CH4 fluxes from the eastern Amazon Basin inferred from atmospheric mole fraction profiles. Journal of Geophysical Research-Atmospheres, 121(1), 168–184. http://doi.org/10.1002/2015JD023874

Beck, V., Chen, H., Gerbig, C., Bergamaschi, P., Bruhwiler, L. M. P., Houweling, S., et al. (2012). Methane airborne measurements and comparison to global models during BARCA. Journal of Geophysical Research-Atmospheres, 117(D). http://doi.org/10.1029/2011JD017345

Brenninkmeijer, C. A. M., Crutzen, P. J., Boumard, F., Dauer, T., Dix, B., Ebinghaus, R., et al. (2007). Civil Aircraft for the regular investigation of the atmosphere based on an instrumented container: The new CARIBIC system. Atmospheric Chemistry and Physics, 7(18), 4953–4976.

Bousquet, P., Ringeval, B., Pison, I., Dlugokencky, E. J., Brunke, E. G., Carouge, C., et al. (2011). Source attribution of the changes in atmospheric methane for 2006-2008. Atmospheric Chemistry and Physics, 11(8), 3689–3700. http://doi.org/10.5194/acp-11-3689-2011

Gatti, L. V., Gloor, M., Miller, J. B., Doughty, C. E., Malhi, Y., Domingues, L. G., et al. (2015). Drought sensitivity of Amazonian carbon balance revealed by atmospheric measurements. Nature, 506(7486), 76–80. http://doi.org/10.1038/nature12957

Locatelli, R., Bousquet, P., Saunois, M., Chevallier, F., Cressot, C. (2015). Sensitivity of the recent methane budget to LMDz sub-grid-scale physical parameterizations. Atmospheric Chemistry and Physics, 15(17), 9765–9780. http://doi.org/10.5194/acp-15-9765-2015

Miller, J. B., Gatti, L. V., d'Amelio, M. T. S., Crotwell, A. M., Dlugokencky, E. J., Bakwin, P. S., et al. (2007). Airborne measurements indicate large methane emissions

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from the eastern Amazon basin. Geophysical Research Letters, 34(10), L10809–. http://doi.org/10.1029/2006GL029213

Naik, V., Voulgarakis, A., Fiore, A. M., Horowitz, L. W., Lamarque, J. F., Lin, M., et al. (2013). 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(1), 5277–5298. http://doi.org/10.5194/acp-13-5277-2013

Patra, P. K., Houweling, S., Krol, M., Bousquet, P., Belikov, D., Bergmann, D., et al. (2011). TransCom model simulations of CH4 and related species: linking transport, surface flux and chemical loss with CH4 variability in the troposphere and lower stratosphere. Atmospheric Chemistry and Physics, 11(24), 12813–12837. http://doi.org/10.5194/acp-11-12813-2011

Patra, P. K., Krol, M. C., Montzka, S. A., Arnold, T., Atlas, E. L., Lintner, B. R., et al. (2015). Observational evidence for interhemispheric hydroxyl-radical parity. Nature, 513(7517), 219–223. http://doi.org/10.1038/nature13721

Shindell, D. T., Faluvegi, G., Stevenson, D. S., Krol, M. C., Emmons, L. K., Lamarque, J. F., et al. (2006). Multimodel simulations of carbon monoxide: Comparison with observations and projected near-future changes. Journal of Geophysical Research-Atmospheres, 111(D), D19306. http://doi.org/10.1029/2006JD007100

Schuck, T. J., Brenninkmeijer, C. A. M., Baker, A. K., Slemr, F., Velthoven, von, P. F. J., Zahn, A. (2010). Greenhouse gas relationships in the Indian summer monsoon plume measured by the CARIBIC passenger aircraft. Atmospheric Chemistry and Physics, 10(8), 3965–3984.

Schuck, T. J., Ishijima, K., Patra, P. K., Baker, A. K., Machida, T., Matsueda, H., et al. (2012). Distribution of methane in the tropical upper troposphere measured by CARIBIC and CONTRAIL aircraft. Journal of Geophysical Research, 117(D19), n/a-

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n/a. http://doi.org/10.1029/2012JD018199

Wofsy, S. C. (2011). HIAPER Pole-to-Pole Observations (HIPPO): fine-grained, global-scale measurements of climatically important atmospheric gases and aerosols. Philosophical Transactions of the Royal Society a: Mathematical, 369, 2073–2086. http://doi.org/10.1098/rsta.2010.0313

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