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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Future methane, hydroxyl, and their uncertainties: key climate and emission parameters for future predictions" *by* C. D. Holmes et al.

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Our comments in bold below document the revisions that we have made to the manuscript, as we promised in our earlier online response.

This comments is written also on behalf of S.A. Montzka and J. Lelieveld

This paper presents an analysis of methane lifetime variations calculated by three models. Overall, calculated variations in methane lifetime are rather small (1%, page 20944, line 13) and modeled variations can be reasonably well represented by a parametric function with only five parameters, (i) temperature (ii) water vapor (iii) column ozone (iv) biomass burning emissions, and (v) lightning NOx emissions.

The paper is very well written, and provides an interesting synthesis of the main factors that are thought to influence the modeled methane lifetime.

1 Major comments

We have an objection, however, relating to a statement made in the paper that we feel is inaccurate. In section 3.3, modeled variations in the methane lifetime are compared to observed variations in the methyl chloroform (MCF) decay rate. Since 1998, MCF emissions have been small compared to the atmospheric burden, and variations in observed MCF decay rate may be interpreted as variations in the tropospheric oxidizing capacity, and hence in the methane lifetime. However, in Fig. 4 the authors show that the detailed interannual variations in the MCF decay rate depend on the observational network.

They conclude: "Given that differences in observed MCF decay rates between the two networks are as large as their difference from CTM $\tau_{CH_4 \times OH}$ anomalies, we conclude that better understanding of the systematic differences between the observation networks is required before using them as a constraint on $\tau_{CH_4 \times OH}$ and OH interannual variability."

This statement suggests that an analysis of MCF data has no informative value with respect to mean OH concentrations, i.e. the lifetime of methane, and the interannual variability of OH. We think this sentence inaccurately reflects the value of MCF measurements for deriving information about OH. We agree that a determination of the phase of interannual OH variations from MCF data would be robust especially if the timing of those changes were consistently derived from both measurement networks, but poorly correlated variations derived from the two networks do not negate the fact that results from both networks provide a consistent estimate of mean annual OH (and, therefore, $\tau_{CH_4 \times OH}$) and of the magnitude of interannual changes in OH since 1998. We also feel that this result does not negate the main message of Montzka et al. (2011), which is that the OH-variability derived from MCF since 1998 was remarkably small

compared to that inferred in earlier years and that the tropospheric oxidizing capacity appears well buffered against perturbations.

Another point we would like to make is that modeled interannual OH-variability is likely an underestimate (1%), even when the main drivers of OH variability are accounted for. Possible reasons include unaccounted variability in VOC emissions, limited spatial resolution, and simplified chemistry schemes used in the analysis. In contrast, interannual OH-variability derived from MCF measurements (2.3%) is likely overestimated (as explicitly discussed in Montzka et al., 2011), due to potential calibration and measurement consistency issues and errors in translating measurements at a few stations to a global MCF decay rate, in addition to the potential errors associated with unknown emission variations.

We agree with the authors' statement that the differences between the two MCF measurement networks should be resolved, though this is a problem that has received attention in AGAGE and NOAA for many years. In the end, the problem may be unresolvable, since the discrepancies the authors discuss arise from differences in measured mixing ratios that are often only small fractions of a part per trillion. Even so, results of both networks are not uncorrelated as both show similar variability and a clear anomaly around 2003. Likewise, the different models agree approximately, though not perfectly. Ultimately, our goal should be to reduce errors in both models and measurements, while measurements remain imperative to evaluate models.

As we wrote in our earlier online response, we agree with Krol et al. and have now edited the section to make this clear. The following sentences have been added or edited:

"Averaged over 1998–2007, the global MCF decay rates from the 2 networks differ by less than 1 % (0.1811 a^{-1} for NOAA, 0.1796 a^{-1} for AGAGE), providing a strong constraint on the long-term global-mean OH and methane lifetime."

"In all CTMs, simulated $\tau_{\mathrm{CH_4}\times\mathrm{OH}}$ variations ($0.7-1.1$ % for σ/mean) are smaller

than the upper limit imposed by the MCF constraint (2.3 %, Montzka et al., 2011). Adding variable isoprene emissions to the GEOS-Chem simulation increases the simulated variability by only 0.1 % (not shown). While the CTM anomalies are consistently within the observational uncertainty for both observation networks (shaded region of Figure 4), the year-to-year changes in the models generally do not correlate with the MCF data."

"These differences in observed MCF decay rates between the two networks are as large as the discrepancy between CTM $\tau_{\mathrm{CH_4}\times\mathrm{OH}}$ anomalies and either set of observations. Therefore, we conclude that better understanding of systematic differences between the observation networks is required before MCF can be used to constrain $\tau_{\mathrm{CH_4}\times\mathrm{OH}}$ and OH anomalies in specific years at the precision required (1-2 %) to test CTM interannual variability."

2 Minor Comments

Page 20936, line 13: "...which increases OH and biases $\tau_{\mathrm{CH_4}\times\mathrm{OH}}$ high by about 10%....". if OH increases one would expect the methane lifetime to be biased low.

We fixed this typo.

Page 20944, line 29: "Fig. 2" should be "Fig. 4". **FIXED**

In the Supplement: WLEF tower is in Wisconsin. **FIXED**

NOAA data after Dec 2007 have been appropriately omitted from the analysis, but why are NOAA data and not AGAGE data shown past that date in Figure S2?

We inadvertently truncated AGAGE rather than NOAA data in Figure S2. This has been corrected and our conclusions are unaffected.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 20931, 2012.