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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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Thank you for your many helpful suggestions. We address each one below with our responses in bold.

Both reviewers asked us to clarify which variables in our parametric model were averaged over 40S-40N and why. The variables were temperature, water vapor and ozone column, because the tropics and subtropics are where these variables have the most influence on tropospheric OH and methane lifetime. One reviewer also pointed out that global averages are more often reported by GCMs and in the literature, and asked how our future projections would change if they were based

on global averages. We agree that global quantities are more easily available. In response to both comments, we have revised our parametric model calculations to use global-mean air temperature and water vapor mixing ratio. All text, figures and tables are updated accordingly. Interannual changes in temperature and water vapor are dominated by tropical variability, so $\tau_{\mathrm{CH_4 \times OH}}$ changes in the parametric model are robust against choosing averages over 40S-40N or globally. For ozone columns, however, polar regions cause major interannual changes in global-mean ozone column, but contribute little to $\tau_{\mathrm{CH_4 \times OH}}$ variability since most OH production from tropospheric O3 photolysis occurs at low latitudes. Therefore, we continue to average ozone columns over 40S-40N. All our results, including projections of future methane are robust against changing averaging regions for temperature and water vapor. For example, $\tau_{\mathrm{CH_4 \times OH}}$ increases 12.9 ± 10.8 % by 2100, compared to 13.3 ± 10 % in our ACPD manuscript (using 40S-40N averages). We added the following paragraph to Sect. 3.3 describing how choice of averaging region affect the parametric model performance.

"We find that 85% of methane oxidation by tropospheric OH occurs between 40S and 40N and this region also controls the interannual variability of $\tau_{\mathrm{CH_4 \times OH}}$ in the CTMs. The same latitudes also dominate the variability of global-mean temperature, water vapor, lightning NOx and biomass burning, so the parametric model performs nearly as well if these input variables are averaged over 40S to 40N instead of globally. Stratospheric ozone exerts the greatest influence on $\tau_{\mathrm{CH_4 \times OH}}$ over the tropics and subtropics, where UV photolysis of tropospheric ozone is an important primary source of tropospheric OH and where the quasi-biennial oscillation is the dominant source of stratospheric ozone variability. However, global-mean ozone columns are strongly influenced by variability in the springtime polar regions, so using global averages significantly degrades the parametric model correlation with the CTMs ($R^2 \approx 0.75$)."

In addition to changes suggested by reviewers, after submitting our manuscript

to ACPD, we discovered a bug in the aerosol heterogeneous chemistry in one of the models (Oslo CTM3). We repeated all simulations after fixing the problem and found that the bug had negligible effect on our results, although it changed the final rounding digit of some numbers. However, in the process we found a typo in the Oslo CTM3 sensitivity to biomass burning in Table 2. Using the correct sensitivity parameter dramatically improves the parametric model reconstruction of the CTM3 methane lifetime during 1997-2009 (R^2 = 0.94 now vs. 0.48 previously; Fig. 1), which strengthens one of the main arguments of our manuscript: that a simple parametric approach can reproduce the main features of global methane lifetime simulated in many CTMs. We have updated the text in Sections 3.2 and 3.3 to reflect the changes, including a few sentences to explain why Oslo CTM3 is less sensitive to biomass burning than the other CTMs.

The manuscript is a timely, important contribution. It presents a new parametric approach to projecting changes in atmospheric methane lifetime including several sources of uncertainty, such as changes in anthropogenic emissions, meteorology, and climate-chemistry feedbacks. This approach allows for projections to be made and most importantly an uncertainty estimate, without requiring large ensembles with full climate-chemistry models. Important findings include the identification of the most important variables leading to inter-annual variability in hydroxyl radical and thus methane lifetime.

GENERAL COMMENTS: The text needs clarification in a few places but is generally well-written with clear figures and tables. The general comments from referee #1 will improve the flow of the paper. I recommend publication once those and the points below are addressed.

It is not clear the extent to which the authors tested the role of feedbacks from natural sources, notably isoprene and wetlands. Section 3.2 states that the simulations neglect biogenic VOC variability but Section 5 asserts that they were found to have a minor impact on present-day inter-annual variability. This should be clarified in Section 2.

How would inclusion of dust or wetland methane emissions change the conclusions?

Feedbacks on natural methane emissions are included as an explicit uncertainty term in the future projections. This is now explicit in Sect. 6: "The scenario specifies future anthropogenic methane emissions and we assume natural emissions could change $\pm 20\,\%$ (1σ) by 2100 due to climate feedbacks, which is about twice the change in wetland emissions since the preindustrial era (Houweling et al., 2000)."

The CTM simulations in this work neglect biogenic VOC and anthropgenic emission variability. The confusion arises because we compare our results to a prior GEOS-Chem simulation that includes these additional sources of variability. We now make this more clear in Sect. 3.3: "We compare our GEOS-Chem/MERRA simulation to an available GEOS-Chem simulation (M. Mu, personal communication, 2012) that includes these trends and variability in emissions, uses identical meteorology and resolution, and has minor other changes (version 9-01-01). The two model configurations simulate very similar $\tau_{\mathrm{CH_4 \times OH}}$ ($R^2 = 0.98$) over 1999–2009, meaning that year-to-year changes in biogenic and anthropogenic emissions contribute little additional $\tau_{\mathrm{CH_4 \times OH}}$ variability and do not degrade the parametric model performance."

In Sect. 5, we now repeat that the minor importance of biogenic VOC variability is based on this one model: "Neglected processes — including shifting emission locations, biogenic VOC emissions, stratosphere-troposphere exchange, and aerosol interactions with photolysis and chemistry — might cause additional systematic prediction errors, but we have found that including these processes in one model (GEOS-Chem/MERRA) has little impact on present-day $\tau_{\mathrm{CH_4 \times OH}}$ variability (Sect. 3.3)."

This distinction between inter-annual variability versus long-term trends is made several times in the paper but could be more clearly framed early on. The underlying assumption is that the variables responsible for driving inter-annual variability in these models over the past few decades will remain important over the next century, as well as long-term trends in anthropogenic emissions. It seems possible that other drivers, for example, aerosols, clouds, convection, natural emissions, could contribute to trends over the next century. Is the uncertainty estimated here sufficiently large as to cover those possibilities?

We now introduce the distinction between interannual variability and long-term trends in the introduction: "The parametric model includes climate and emission factors that control the interannual variability of methane lifetime in the CTMs. These factors are likely important on decadal time scales as well. The parametric model also includes anthropogenic emissions that can drive decadal trends in methane lifetime, but contribute little to interannual variability. Uncertainties in atmospheric chemistry are included in the parametric factors based on the range of perturbation responses across the CTMs. We evaluate the parametric model against 13-yr CTM simulations of methane lifetime, and against observed variability in tropospheric OH, as measured by the decay of methyl chloroform. Assuming that the same climate and emission processes will remain dominant drivers of methane lifetime throughout the 21st century, we use this parametric model with uncertainties to make new projections of methane abundance and its uncertainties through 2100."

The conclusions, and possibly the abstract, should place the GWP in the context of previous estimates since this is a significant change. It should be noted that this new GWP estimate primarily reflects a new estimate for the stratospheric ozone response to methane based upon results from one model. This contrasts with the other findings that are based on three models and in several cases estimates from the literature. How well does the Oslo CTM3 model represent stratospheric ozone chemistry?

The abstract now points out that our estimate is larger than past ones: "We estimate the 100-yr GWP to be 32, which is 25% larger than past assessments."

We add a statement in Sect. 6 to support the model performance in the stratosphere: "Although these stratospheric effects have greater uncertainty because they are assessed from a single model, the stratospheric chemistry mechanism in Oslo CTM3 is able to reproduce recent stratospheric ozone variability (Isaksen et al., 2012)."

These suggestions are also integrated into the conclusions: "We also provide a new estimate of the indirect components of methane RF. Based on three CTMs, tropospheric ozone contributes 30–50 % of the direct methane RF, compared to 25 % that has been used in previous IPCC assessments (Forster et al., 2007). Using one model with stratospheric chemistry (Oslo CTM3), we calculate the stratospheric chemical effects of methane, including those mediated by water vapor. Combining the troposphere and stratosphere results, the total methane-induced ozone RF is 50 % of the direct methane RF. Based on these data, the 100-yr methane GWP is 32, which is higher than several previous estimates around 25.

SPECIFIC COMMENTS:

Section 2. Consider a Table to compare the CTMs more easily.

We added Table S1 for this purpose and refer to it at the beginning of Sect. 2.

Section 2.4 The assertion that biomass burning is a major cause of OH variability should be supported by reference(s) or clarified that this is a finding from this work. Do the GEOS-Chem lightning scale factors vary each year?

We now cite Duncan et al., 2003; Manning et al., 2005; Dalsoren et al., 2006 who have shown that biomass burning causes OH variability. GEOS-Chem lightning scale factors do not change each year. We believe the existing reference to the work by Sauvage et al. (2007), which developed the scaling, is sufficient.

Section 3.1. P 20939 Line 24-25 Labrador et al. (2005) http://www.atmos-chem-phys.net/5/1815/2005/acp-5-1815-2005.html could be included in the references listed.

A paper by Labrador et al. (2004, Geophys Res Lett) is more relevant, so we add references to it here and in Table 2.

P20940 Line 19-20 is awkward since the models seem to agree fairly well in the interannual variability of the sensitivity.

We now describe the models as having "moderate agreement in mean sensitivity."

P20940 Line 24-25 suggests future climate will be more El-Nino like but this is not a robust finding, see Collins et al. Nature Geoscience 3, 391 - 397 (2010).

For this reason, and because of the dependence of biomass burning sensitivity on emission altitude, we now say, "Future El Niño activity cannot be predicted robustly (Collins et al., 2010) and injection altitude distributions remain uncertain so, despite some CTM consensus on present-day biomass burning sensitivity, we adopt a broad uncertainty range for future sensitivity."

P20941 f=1.34 but 1.33 on P20946. Do the adopted values include the new results plus previous findings or changes with the parameter?

The correct value is f=1.34 and we have fixed this. Adopted values consider previous findings as well as new results. This is already stated in a footnote to Table 2: "Adopted values are the mean of CTMs, except for VOC, and aviation NO_x, which come from literature. Uncertainties are 1-σ values based on CTM spread and expert assessment of literature."

Section 3.2 p 20942 needs to define the sensitivity parameter. Lines 15-20 are confusing.

The sensitivity parameter was defined earlier (new Sect. 3.2). To avoid confusion, we promote the definition of α to a numbered equation and refer to it immediately

after the point of confusion in Sect. 3.3: "α_i is the sensitivity of the CTM to that forcing variable (Eq. (2))"

Section 3.3. This section is very important as it is the only evaluation in the paper and some important complications in interpreting the measurements are found which seem worthy of inclusion in the abstract and/or conclusions, though this needs to be properly placed in context of the strengths and value of the measurements as pointed out in M. Krol's comment. The correlation between the two networks does not look very strong and thus it could be of interest to show the other collocated sites in the supplemental information.

The abstract now mentions the observational comparison: "We simulate methane lifetime in three chemical transport models (CTMs) — UCI CTM, GEOS-Chem, and Oslo CTM3 — over the period 1997–2009 and compare the models' year-to-year variability against constraints from global methyl chloroform observations."

The conclusions now add: "The simulated $\tau_{CH_4 \times OH}$ anomalies generally lie within the constraints provided by global methyl chloroform observations, but correlate poorly with year-to-year changes inferred from the NOAA and AGAGE networks. However, while both networks provide strong and mutually consistent estimates of the long-term mean $\tau_{CH_4 \times OH}$, their year-to-year anomalies differ from each other by as much as they differ from the CTMs. These differences in global $\tau_{CH_4 \times OH}$ anomalies can partially be explained by small (~ 0.1 ppt) discrepancies in the monthly mean methyl chloroform concentrations at collocated measurement sites."

Supplemental Figure S2 now includes comparisons of AGAGE and NOAA monthly means at all 4 collocated sites (Cape Grim, American Samoa, Trinidad Head, and Mace Head).

Section 3.4 Was this simulation with 5% more methane used to determine the sensitivity to CH4 abundance in Table 2? Somewhere it should be explained how that parameter was determined.

The revised text makes this clear in Section 3.2, where the sensitivity to methane abundance is first discussed: "The sensitivity of $\tau_{CH_4 \times OH}$ to methane abundance is closely related to the methane feedback factor, f, which is the ratio of methane perturbation lifetime to total budget lifetime (Prather et al., 2001). We calculate these terms from a 5 % perturbation to methane abundance for 1997–2009 in the CTMs."

Section 4. The choice not to include uncertainties to CO and VOC emissions seems arbitrary since the other reference listed includes uncertainties. Some justification is needed.

We initially neglected uncertainty in these terms because we did not have constraints on its magnitude. We now estimate the uncertainty from new perturbation simulations for anthropogenic CO (now in Table 2) and from literature values for VOC. Our updated CH_4 historical reconstruction and future projections change only slightly (i.e. second significant digit) as a result.

Section 5. Does the 35% decrease in biomass burning apply specifically to NOx, or is it uniformly occurring for all fires such that CO/NO remains constant? Are the 3D water vapor and temperature fields not available from CMIP5? Some typos in referring to Figures 6 and 7. It seems important that the uncertainty range for the projections here do not overlap zero, a robust change of sign, whereas the range from ACCMIP does. Should we place more confidence in the results of this study than the multi-model ensemble? Another new study in ACPD with only one climate-chemistry model (John et al. : http://www.atmos-chem-phys-discuss.net/12/18067/2012/acpd-12-18067-2012.html) reports changes in methane lifetimes under the RCP8.5 scenario and is consistent with the findings here.

The 35% reduction in biomass burning applies to all gases and aerosols emitted by fires. This is now stated in the text. We use surface temperature from CMIP5, rather than 3-D water vapor and temperature fields, because this makes our results more easily reproducible (without downloading terabytes of CMIP5 data) and makes it easier to apply the parametric model to other climate scenarios. This is now stated in the text. We learned of the study by John et al. after submitting our original manuscript. We now cite it elsewhere.

We think the parametric model and GCM ensembles are complementary rather than competing approaches for estimating future methane change. To clarify this we have slightly expanded the comparison of our results to ACCMIP: "Although we do not think future lightning estimates from GCMs are robust (see above), assuming an equally large lightning change in the parametric model would lower $\tau_{CH_4 \times OH}$ in 2100 by about 5 %, after including the methane feedback. The similar central estimates and ranges of $\tau_{CH_4 \times OH}$ change in this work and ACCMIP, after accounting for different lightning assumptions, demonstrates that our simple parametric model represents the major climate-chemistry interactions and uncertainties affecting methane in current GCMs. This supports using the parametric model for climate scenarios where a large ensemble of GCMs with chemistry is not available and too costly to generate."

Throughout the text, and in Table 2, it should be clarified as to whether the temperature and water vapor are only averaged below 400 hPa and 40N-40S. Are the stratospheric columns only 40N-40S in Table 2?

In our revised manuscript all temperature and water vapor averages are global, which will avoid this confusion. Ozone columns are always averaged over 40S-40N. Where the perturbations are introduced in Sect. 3.2, we now explain, "Perturbations apply globally, except for stratospheric ozone, which is only perturbed over 40S–40N, where tropospheric OH production from UV photolysis of tropospheric O_3 occurs."

In Figure 1, the parametric model developed from the Oslo CTM3, which represents more climate-chemistry interactions than the other CTMs, captures the least amount of variance. Does this imply that these interactions, not included in the other models, may be important in driving lifetime variability?

There was an error in the parametric model (biomass burning factor) for the Oslo CTM3, which we have now corrected. After this change, the parametric model explains over 90% of the variance in all CTMs, so we don't think this is evidence for other important processes.

Figure 6. Are these single-year differences? If so, probably better to average at least 5-year periods at the beginning and end.

The updated figure uses 5-year averages for 1980-4 and 2001-5. Differences from the first manuscript are unnoticable, except for the CH4 term, which has small changes, because of the plateau in the CH4 abundances in the early 2000s. Since there is no short-term variability in the future projection, we do not need to do a multi-year average.

Table 2. The sensitivities for convective mass flux and water clouds are similar to biomass burning. Why are they deemed less important? While the variables are not exactly comparable, some of these sensitivities could be compared to those reported in Spivakovsky et al., 2000, their Table 6.

As the main text now says, "Convective fluxes and cloud optical depths for water and ice clouds, as diagnosed in ECMWF meteorology, vary annually by 2 % and have small sensitivity (-0.03 to $+0.01$), so these factors have very little impact on $\tau_{\mathrm{CH_4 \times OH}}$ ($\ll 1\%$)." We now remind the reader of this with a footnote to Table 2 for the important variables: "Major cause of interannual $\tau_{\mathrm{CH_4}} \times \mathrm{OH}$ changes (based on sensitivity and interannual changes in the variable)."

Spivakovsky et al. (2000) report sensitivities of mass-weighted, tropospheric-mean OH concentration to various factors. These are not directly comparable to our results in Table 2, which are the sensitivities of methane lifetime. The OH and methane lifetime sensitivities can differ because of changes in the vertical distribution of OH. Therefore, we do not include the Spivakovsky results in our Table 2, but we now reference them in the text.

Table 4. Why the difference between the CH4 abundance change reported in Table 4 and Figure 7 in 2100?

Fixed. This was a typo in Table 4. We have rechecked all entries in Table 4.

Figure S6 Consider using the same y-axis scale to aid the reader in determining which variables are most important.

DONE.

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