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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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Response to Referee 1

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 $\tau_{CH_4 \times OH}$ 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 $\tau_{CH_4 \times OH}$ 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, $\tau_{CH_4 \times OH}$ increases 12.9 $\pm$ 10.8 % by 2100, compared to 13.3 $\pm$ 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 $\tau_{CH_4 \times OH}$ 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 $\tau_{CH_4 \times OH}$ 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 by Holmes et al. examines future OH, methane and their uncertainties using a parametric model constructed based on simulations with multiple detailed global chemistry-transport models. The work presented will be of great interest to the atmospheric chemistry and climate community, as it approaches several current topics such as the interannual variability, recent trends and future evolution of tropospheric composition, as well as the global warming potential of important radiative forcing agents. The study is also expected to have implications for more applied research in the future. The manuscript is well written and the topic is certainly suitable for Atmospheric Chemistry and Physics. Therefore, I recommend publication following some minor revisions described below.

GENERAL COMMENTS:

1) A general comment that I have has to do with the structure of the paper. It feels like sections 3.3 and 3.4, though useful, are interrupting the flow of the paper as it evolves from presenting the parametric model (Sect. 3.2) to employing it for the historical and future analysis (Sect 4, 5). I would suggest having the MCF section earlier (possibly

shortly after the beginning of Sect. 3), and the GWP section later (perhaps just before the Conclusions).

We have reorganized the sections as suggested and slightly modified the text of the affected subsections to improve the flow in the new organization

2) The work presented includes a variety of approximations and assumptions, which is inevitable, as it is an ambitious effort to bring this range of models and methods together in one study. These approximations and assumptions are even more evident when looking at table footnotes and at the supplementary material in detail. However, the fact that they are mentioned at scattered parts of the text does not help the reader to have a good idea on which assumptions are the most important ones. I would recommend including a "caveats paragraph" in the Conclusions, which would help the reader understand what could be improved in future efforts building on the approaches presented here.

Since the parametric model now uses global averages for all quantities except ozone column, we think that there are now fewer approximations and assumptions that need to be explained. Nevertheless, we have added several sentences to the introduction and conclusions to point out key assumptions in this work. We did not feel that this required an entire standalone paragraph.

From 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... 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."

In the conclusions, we now repeat many caveats that are given throughout the paper: "Our decrease is the same direction of change as previous CTM studies and consistent with methyl chloroform observations, but smaller magnitude than many past estimates. We also use the parametric model to project future methane abundance, which implicitly assumes that the emission and climate processes that controlled methane lifetime in the recent past will remain dominant throughout the 21st century."

Later in the concluding paragraph, we mention one piece of evidence that the parametric model is reasonable despite these caveats: "Our projected change in methane lifetime is consistent with an ensemble of GCMs containing atmospheric chemistry, providing further evidence that the parametric model includes the key processes."

SPECIFIC COMMENTS:

Page 20933, Line 7: Suggested rephrasing: ". . .the largest atmospheric methane sink, due to anthropogenic emissions of CO, nitrogen. . ." to ". . .the largest atmospheric methane sink: anthropogenic emissions of CO, nitrogen. . ." **See next comment.**

Page 20933, Lines 4-12: In the first part the authors refer to factors affecting OH (CO, NOx, VOCs, CH4 feedback), while in the second they refer to factors that affect methane. Please correct to make consistent.

We restructured these sentences to focus only on methane sinks, while adopting the reviewer's suggested phrasing. The new text is, "In the IPCC Third Assessment Report (TAR), 4 parameters accounted for changes in the largest atmospheric methane sink, oxidation by tropospheric OH: anthropogenic emissions of CO, nitrogen oxides (NO_x), and volatile organic compounds (VOCs) and the negative feedback between methane abundance and tropospheric OH (Prather et al., 2001). Other sinks, which include oxidation in the stratosphere, oxidation by tropospheric chlorine, and uptake into soil, were assessed but assumed not

to change during the 21st century projections."

Page 20933, Lines 16-20: And surface albedo. **CHANGED**

Page 20934, Lines 6: Please change "previous approaches" to "previous parametric approaches" or "previous approaches of this kind". **CHANGED.**

Page 20935, Lines 24-26: What about overhead ozone column and aerosols? Do they affect photolysis in the model?

Photolysis rates in CTM3 use overhead ozone columns calculated in the model. Aerosols are not included in the photolysis calculations. The text now makes this clear: "Photolysis rates required in the chemistry mechanism are calculated online using the Fast-JX method (Neu et al., 2007), with cloud distributions from ECMWF meteorology and ozone concentrations calculated in the CTM. Aerosol effects on photolysis are neglected except for a small contribution from black carbon (Sovde et al., 2012), which increases OH and biases $\tau_{CH_4 \times OH}$ low by about 10 % (Bian et al., 2003; Martin et al., 2003)."

To avoid repetition, we have shortened the description of aerosols in the UCI CTM: "Aerosol effects on chemistry are neglected, which biases $\tau_{CH_4 \times OH}$ low by up to 5 % in addition to the aerosol-induced photolysis bias described above for Oslo CTM3 (Martin et al., 2003; Macintyre and Evans, 2010)."

Page 20936, Lines 5: Oslo CTM3 is sometimes referred as CTM3 and sometimes with its full name. It would be better to use a consistent name throughout the text. **CHANGED. "Oslo CTM3" is now used throughout.**

Page 20936, Lines 4-14: Again, please briefly mention whether time-varying overhead ozone column and clouds are used in photolysis calculations.

The text now makes clear that we use time-varying ozone and clouds in the UCI CTM: "As in Oslo CTM3, clouds from ECMWF meteorology are used for photolysis. Simplified stratospheric O_3 chemistry is simulated with Linoz (version 2,

Hsu and Prather, 2009) and used for photolysis calculations and stratosphere-troposphere exchange."

Page 20936, Lines 13-14: References should be chronological. **FIXED, here and elsewhere.**

Page 20938, Line 19: More up-to-date present-day values from ACCMIP: 9.7±1.5 yr (Naik et al., in prep.); 9.8±1.6 yr (Voulgarakis et al., 2012, ACPD).

Thank you for the updated numbers, which we have inserted in our paper. At the time we submitted to ACPD, we were working from an earlier draft of the manuscripts by Naik et al. and Voulgarakis et al.

Page 20938, Lines 20-25: Any ideas why GEOS-Chem shows larger variability?

The parameter sensitivities explain the larger variability. We now point out the different magnitudes of variability and foreshadow the explanation at the start of Section 3: "Oslo CTM3 has the least variability (0.65 % for σ/mean), while GEOS-Chem with MERRA meteorology has the most (1.1%). The common features, as well as differences in their magnitude, are explained below by a small number of processes (Sect. 3.3)."

Page 20939, Line 6: Is there any rationale behind the exact choice of years.

This was based on the years of available meteorology. To make this clear, we now explain, "Perturbations are applied to 1997–1999 for Oslo CTM3 and the UCI CTM, and, because of meteorological data availability, to 2004–2006 for GEOS-Chem with GEOS-5."

Page 20939, Line 10: Table 2: Please note next to footnote (a) that this labeling as "major cause" is based also on the actual interannual changes, not only on the sensitivities.

DONE. The footnote text now says, "...major causes of interannual $\tau_{CH_4 \times OH}$

changes (based on sensitivity and interannual changes in the variable)..."

Also, please note that Krol and VanWeele (1997) found a 2.8% decrease in methane lifetime due to a 10% decrease in the ozone column, while Voulgarakis et al. (2009) found a 5.1% decrease in methane lifetime for a 20% decrease in the ozone column. Note though that in the former the ozone column perturbation was applied only to the extratropics, while in the latter it was applied globally.

The results of Krol and van Weele and (1997) and Voulgarakis et al. (2009) are not strictly comparable to ours because they perturbed ozone columns in the extratropics and globally, respectively, whereas we limit our perturbation to 40S-40N. Nevertheless, we added both references to Table 2, with notes about how the perturbation latitudes.

Page 20939, Line 17-18: Why aren't the emissions variables also averaged over 40S-40N (Fig. 2)? Please explain.

In our revised manuscript all emissions and climate variables are averaged globally, except stratospheric ozone column. Immediately after the perturbations are introduced, the text now explains, "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."

Page 20945, Line 4: Why is the perturbation 5%? Is it totally random?

We now explain that, "A small perturbation is used to satisfy the linearity assumption in the methane feedback factor derivation (Sect. 3.2)."

Page 20945, Line 20: Why does Table 5 come before Table 3 and 4?

The order is fixed.

Page 20946, Lines 5-6: Suggested rephrasing: ". . .tropospheric ozone RF is 30–50% of the direct methane RF . . ." to ". . .methane RF through its impacts on tropospheric

ozone is 30-50% of the direct methane RF. . ."

We adopted this suggestion.

Page 20947, Line 15: Please change the title of Table 3 to "Datasets used for calculation of historical. . ."

We updated the title to "Datasets used to calculate historical . . . "

Page 20948, Lines 8-9: State more clearly why this is true (effects on UV and photolysis), as it may not be intuitive for all readers.

The revised sentence now explains, "$\tau_{CH_4 \times OH}$ is also depressed through much of the 1990s when stratospheric ozone was low, due to the solar cycle and Mt. Pinatubo, enhancing UV penetration and photolysis in the troposphere"

Page 20950, Line 2: The region over which the averaging is performed for meteorological variables (40S-40N and up to 400hPa) is indeed very relevant when it comes to methane oxidation, but arguably quantities averaged over such a region are not typically reported by GCMs as global metrics. It would be a good idea, at least in future work (but possibly also here, if it does not require a large amount of effort), to test whether the results from the parametric model using global tropospheric mean meteorological metrics (which are more commonly reported) are similar to the ones using the regional values. I would expect that the difference would not be large, at least for scenarios like RCP8.5 which feature large global temperature and humidity changes. The ozone case would be less straightforward, obviously, but still worth examining.

See our first response. We revised the parametric model projections to use global averages of temperature and water vapor. This required us to recalculate the historical regression between surface temperature and atmospheric temperature and water vapor, using global averages for all (Figs. S3 and S4). We apply the updated relations to future surface temperatures from CMIP5 models (Fig. S5). With these updates, we project atmospheric temperature will increase 3.6

$\pm$ 0.9 K in 2100 in RCP 8.5 (compared to 3.7 $\pm$ 0.9 K in our initial manuscript) and water vapor will increase 43.2 $\pm$ 9.5% (compared to 38.5 $\pm$ 9.2% in our initial manuscript). Our projected CH4 in 2100 dropped less than 50 ppb (out of 3900 $\pm$ 320 ppb).

Another comment that I have here is that although the perturbations to temperature/humidity (Supplement, Table 1) for calculating sensitivities were global, the values used in the parametric models are regional (40S-40N). Isn't there an inconsistency introduced? Please comment.

Since the parametric model now uses global averages for temperature and water vapor, there is no inconsistency.

Page 20951, Line 9: It is interesting that the parametric model and MAGICC reveal a similar evolution, even though the parametric model includes more factors. Could you comment on this?

This is best explained after showing the main components driving the parametric model results. At the end of the section, we now explain, "MAGICC predicts similar $\tau_{CH_4 \times OH}$ changes to the parametric model because its temperature response is similar to the combined effects of temperature and water vapor, and sensitivities to the other dominant terms—land NO_x and methane feedback—have changed little since the IPCC TAR, on which MAGICC is based. The parametric model and MAGICC will differ in socioeconomic and climate scenarios where other emissions, ozone, or lightning drive $\tau_{CH_4 \times OH}$ changes"

Page 20951, Line 11: More up-to-date value from ACPD manuscript: +8.5$\pm$10.4 yr.

We now use the updated ACCMIP value (+8.5 $\pm$ 10.4 %, note percent units), which reduces the discrepancy between our parametric model and the ACCMIP ensemble.

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