Authors' responses to referees' comments on: John et al., Climate versus emission drivers of methane lifetime from 1860-2100, Atmos. Chem. Phys. Discuss., 12, 18067-18105, 2012.

We are very grateful to the referee for providing thorough and constructive reviews. All comments and suggestions been taken into full consideration in producing the revised version of the manuscript. Responses to these comments are provided below.

Anonymous Referee #2

This study explores the variation in methane lifetime and the factors affecting it over the past century and possible realizations of the coming century. In particular, it focuses on the role of climate changes and changes in emissions of short-lived species in driving these changes. As the climate impacts of methane are effectively dependent on its lifetime as well as its abundance (a fact which could be brought out more strongly in the opening paragraph of the introduction), this is an important topic worthy of detailed study. This is the first study to tackle this issue in a consistent way, and is therefore very useful. Overall the paper is well written and clearly presented, and is appropriate for publication with only minor revisions.

Response:
We are very grateful to the referee for recognizing the value of this work.

We have revised the introduction (page 2, lines 16-21).
"Quantifying climate impacts requires an estimate of the CH_4 lifetime, which is largely determined by its loss via reaction with the hydroxyl radical (OH, Levy et al., 1971) in the troposphere, (τ_{CH4_OH}). The OH distribution, in turn, is strongly influenced by changes in both climate and emissions. Furthermore, since methane is an important sink for OH, any change in the methane abundance is amplified via a positive feedback on τ_{CH4_OH} and thereby on the climate system (Prather, 1994)."

The introduction states that the paper examines "the role of changes in emissions versus climate on atmospheric methane lifetime" (p.18069,l.9) While this abbreviated statement is suitable for the title and abstract, a clearer, more precise statement is required in the text. Does "emissions" refer to methane, short-lived species, or both? Does "climate" refer to influences on emissions, atmospheric photochemistry, or both? This only becomes apparent after careful reading of section 2. Given the chemical feedback of methane on its own lifetime and the climate feedback on natural emissions, it would help the reader to be clear at the outset about which factors are being compared.

Response:
The introduction has been reorganized to explicitly indicate the climate and emission factors under consideration in the revised manuscript (page 2, lines 21-28).

"We investigate here the relative importance of changes in climate factors (temperature, OH, water vapor (H_2O), lightning NO_x (LNO_x), photolysis rates) versus anthropogenic emissions (CH_4 abundance, carbon monoxide (CO) and nitrous oxide (NO_x)) in contributing to changes in the τ_{CH4_OH} from 1860 to 2100 in the context of the new set of historical and future emission scenarios (Lamarque et al., 2010; Meinshausen et al., 2011; van Vuuren et al., 2011) developed for the fifth phase of the Coupled Model Intercomparison Project (CMIP5) (Taylor et al., 2012), in support of the Intergovernmental Panel on Climate Change (IPCC) Fifth Assessment (AR5)."

The analysis in section 4 is very interesting, but is somewhat speculative in places, particularly in sect 4.3. The attribution aspects here need to be more firmly grounded. Given the sensitivity studies performed, it should be possible to provide a more quantitative attribution of the effects of the different emission and climate changes over this period. I appreciate that many of the relationships are not linear, and therefore that this would only be approximate, but an important goal for the paper should be to resolve some of the uncertainty in the trends and attribution that are apparent from previous studies so neatly summarized in Table 2.

Response:
To our knowledge, the best information for diagnosing the sensitivity of OH to the individual driving factors comes from the recommendations in Holmes et al. (ACPD, 2012). For the sensitivities they report, we've combined our percentage changes (Table S1 below) but we find that summing the individual estimates for HIST leads to no change (0.05% decrease) from pre-industrial to present whereas CM3 estimates a 5% decrease. This suggests that the sensitivities in our model differ from those recommended by Holmes et al. (ACPD, 2012) or that non-linearities are confounding this approach. Isolating the individual factors in CM3 involves targeted (expensive) sensitivity simulations that are beyond the scope of this first analysis. We have revised the discussion in 4.3 (page 12) to better place our results in the context of recent work. We agree that recent trends deserve attention in future work but prefer to retain our focus on the full historical to future period.

There needs to be a discussion in the conclusions (or perhaps the end of Section 4) on the influence of the factors neglected in this study. What are the expected effects of interactive vegetation, soil and fire emissions, or aerosol interactions with photolysis? While detailed analysis may not be possible here, any evidence on the likely magnitude of these effects and their influence on the results would be useful.

Response:
We now explicitly point out the need for future work to address these other processes, in a framework that allows for a consistent comparison with the changes diagnosed here. (see pages 16-17).

p.18069,l.21: Add uncertainty terms on these emission estimates to quantify "fairly well known".

Response:
We have added uncertainty estimates in the revised manuscript (page 3, line 23-24).
"Overall, annual source and sink strengths are estimated at 582 ± 87 Tg CH_4 and 581 ± 87 Tg CH_4 respectively (Denman et al., 2007)."

p.18070,l.9: Better estimates of methane lifetime are now available from Prather et al., GRL 2012.

Response:
The revised manuscript now includes the Prather estimate (page 3, lines 5-7).
"Prather et al. (2012) have recently estimated an observationally-derived present-day τ_{CH4_OH} of 11.2 ± 1.3 yr, which includes updated best estimates, with uncertainties, for the factors contributing to the atmospheric methane budget."

p.18070,l.18: Note here that methane integrates the OH abundance over a wide range of different environments due to its long lifetime. Localized OH changes may differ greatly in magnitude and sign in different places.

Response:
We have noted the large spatial variability in OH in the revised manuscript (page 4, lines 8-9).
"Furthermore, the relative importance of individual OH sources and sinks varies widely by region (e.g., Lelieveld et al., 2004)."

p.18072,l.14: Some comment is required on the impacts of the "restoration" (relaxation?) process applied to CH4. The CH4 emissions aren¿t modeled, and this should be explicitly stated; the effective emissions are therefore driven by the model vertical transport processes as well as just the applied boundary conditions. I assume that the boundary conditions are taken from the earlier CMIP5 runs (or observations?) but it would be good to refer to figure 5 here.

Response:
We have addressed this in the revised manuscript (page 5, lines 24-28).
"Emissions of CH_4, nitrous oxide (N_2O), and ozone-depleting substances (ODS) are not simulated in CM3. For CH_4 and N_2O, concentrations for chemistry below 800hPa are restored to historical or RCP values (Meinshausen et al., 2011), with a timestep of 1 day. For ODS, volume mixing ratios are prescribed as a lower boundary condition, with the source of reactive chlorine and bromine in the stratosphere parameterized following Austin and Wilson (2010)."

p.18072,l.19: "likely alter the findings". It would be useful to know by how much.

Response:
We have eliminated this sentence.

p.18073,l.9: The ensemble naming is nearly self-explanatory, but not quite; while the full specifications are provided in Table 3, it takes some effort to decipher the differences between ensembles. Please state in the text (briefly) what the individual simulations are designed to target.

Response:
The revised manuscript includes a short discussion of the individual simulations in Section 2 (page 6, lines17-27).
"The AEROSOL simulations include both the direct (radiative) effect and warm-cloud aerosol indirect effect. In AEROSOL INDIRECT, aerosol and aerosol precursor emissions vary as in HIST, but an 1860 climatology taken from years 1-20 of CONTROL is used for radiation. Thus, AEROSOL INDIRECT includes the warm-cloud aerosol indirect effect on clouds (and the effect of aerosol changes on heterogeneous reaction rates), but does not include direct (radiative) forcing changes by aerosols. The difference between the AEROSOL and AEROSOL INDIRECT simulations provides an estimate of the aerosol direct effect. In ANTHRO, only anthropogenic forcings (viz., aerosols, ozone, greenhouse gases) vary over the historical period while natural forcings are held constant. In NATURAL, only solar and volcanic forcings are allowed to change. The WMGGO3 simulation isolates the influences from changing only well-mixed greenhouse gases and ozone."

p.18075,l.1: Please state how this dynamical tracer tropopause definition is applied; are the calculations done online, or is this based on monthly or annual output? Given that the thermal tropopause is likely to change under climate change simulations and that the tracer tropopause is dependent on tropospheric chemistry, how sensitive are the calculated lifetime changes to this tropopause definition?

Response:
We have revised the manuscript to address this (page 7, lines 25-30).
"The tropopause is defined, separately for each ensemble member, as the model level in which annual mean ozone is 150 ppbv. The 1860 model tropopause is then applied to all simulation years. A comparison of tropospheric methane lifetime computed using surface to 200hPa annual CH_4 loss rates in HIST and RCP8.5 yielded values that were within 1% of those derived using the 150 ppbv O_3 tropopause, suggesting that τ_{CH4_OH} as calculated here is insensitive to the tropopause definition."

p.18075,l.4: How is the standard deviation here derived? Is this over the annual lifetimes along the full length of each ensemble member? In the historical and future simulations where there are underlying trends, have these been removed first? How

should the reader interpret this variability?

Response:
We have revised the manuscript to address this (page 8, lines 1-3, 11-13).
"For all statistics, we first average annual means over all ensemble members to obtain the ensemble mean time series. We then report the mean and standard deviation over the full length of the simulations."
"The standard deviations in Table 4 reflect trends over the time periods considered and are therefore higher than standard deviation of CONTROL which has no long term trend."

p.18076,l.14: The word "solely" is inappropriate here given that there are a range of competing factors influencing the lifetime.

Response:
This has been removed in the revised manuscript (page 9, lines 17-20).
"We therefore ascribe the increase in τ_{CH4_OH} to the doubling of the global methane burden in HIST, which offsets the decrease in τ_{CH4_OH} due to the quadrupling of surface NO_x emissions (Table 5)."

p.18077,l.25: The change in the reaction rate constant increases by a factor of 6, not the rate constant itself!

Response:
Thanks for catching this oversight. The sentence has been amended in the revised manuscript (page 10, line 23).
"Indeed, τ_{CH4_OH} is considerably decreased in WMGGO3 due to a large warming (Fig. 3a), which leads the percent change in the CH_4+OH reaction rate constant to increase by factors of about 6 and 3.3 respectively, compared to HIST and ANTHRO (4.6% vs. 0.8% and 1.4% in Table 5)."

p.18079,l.19: Several of the models shown in Table 2 show this same decrease without any change in meteorology, and therefore greater justification is needed for the attribution of the effect here to changing water vapor. In fact, earlier studies have already noted decreased methane lifetime over the past decade contrasting with the increasing lifetime over centennial timescales (e.g., Gupta et al., GRL 1998; Wild and Palmer, GRL 2008). These studies did not account for changes in climate or aerosol, and recent lifetime decreases could therefore be attributed to the geographical redistribution of emissions of short-lived species from mid-latitude regions towards the tropics. Can the contribution from this effect be estimated from the current simulations? The focus here on global-scale correlations is likely to miss the influence of this type of localization.

Response:

The referee makes a good point. Fig. S3 (below) shows the global distribution of percent change in surface OH from 1980 to 2000 where the largest percent increases occur over India and South East Asia, and Fig S4 (below) shows the ratio of tropical to global surface NO_x emissons from 1980-2005 suggesting that regional changes of OH in the tropics could be contributing to the decrease in methane lifetime in recent decades. We hope to provide additional regional analysis over the 1980-2000 period in a future study, and note the increase of tropical NO_x emissions in the revised manuscript (page 12, lines 13-15).
"Similar to HIST, τ_{CH4_OH} in ANTHRO continues to fall over the last 20 yr, likely reflecting some combination of rising water vapor and an increase in tropical NO_x emissions, particularly over India and Southeast Asia (not shown; Gupta et al., 1998; Wild and Palmer, 2008). "

p.18080,l.17: Can you provide any more specific insight into how this discrepancy might arise, given that all the model studies in Table 2 (despite different formulations) agree on a positive trend?

Response:
Montzka et al., (Science, 2011) suggest that uncertainties in CH_3CCl_3 measurements prior to 1998 may have contributed to the wide OH variability reported in previous studies. We have revised Section 4.3 to address this (page 12, lines 5-12).

p.18082,l.18: increasing temperature, and also humidity.

Response:
Thanks for catching this. The sentence has been amended in the revised manuscript (page 14, line 28).
"Despite the increasing temperature and humidity (Fig. 3a, 4b), which our previous analysis indicates should shorten the τ_{CH4_OH}, the tropospheric methane lifetime increases, from 8.24 yr in 2006-2025 to 8.73 yr in 2081-2100, with a peak value of 8.91 yr in 2062 (Fig. 1)."

p.18082,l.27: Previous studies have noted that the runaway effect seen in a box model does not occur in 3-D models due to the interactions of chemistry with transport processes. What is interesting here is that the runaway effect is even less likely to be seen in a GCM due to the negative feedback of methane on its lifetime through climate.

Response:
Agreed. The sentence has been revised (page 15, lines 6-7).
"In the late 21st century τ_{CH4_OH} declines slightly, presumably reflecting the negative feedback from climate."

p.18083,l.12: "due to anthropogenic emission increases in CH4", is this really what is meant here? The 5% increase is due to increased CH4 emissions offsetting increased

OH from increased NOx emissions and climate changes.

Response:
The referee is correct and the sentence has been amended in the revised manuscript (page 15, lines 18-20).
"Over the historical period, the methane lifetime increases by 5% overall due to anthropogenic emission increases in CH_4 offsetting increases in OH from increased NO_x emissions and climate change."

p.18084,l.11: This decline is not discussed in the text of section 5. Does this decline reflect greater acceleration in warming than in CH4?

Response:
We have removed this sentence.

Table 2 provides a valuable summary of previous studies, but it would be more useful if the Method column indicated which of the model studies used interannually varying meteorology and which didn't. Those which didn't (e.g., Karlsdottir et al., Dalsoren and Isaksen) may be expected to show different trends and/or attribute them to different causes.

Response:
Table 2 has been amended to show this distinction in the revised manuscript.

Figs 3 and 4: Separation of these figures seems artificial, as they show related variables from the same runs. I recommend that they are combined so that key relationships (such as correlations between temperature, humidity, and lightning) can be seen more easily.

Consider moving Fig 5 earlier, as it provides a useful summary of the emission and concentration pathways and therefore helps the reader interpret the ensemble scenarios described in Section 2.

Response:
The motivation for separating the figures was to first show the drivers of methane lifetime, viz, temperature and OH (Fig. 3). Figures 4 and 5 then separated the drivers of OH into climate drivers (lightning NO_x, water vapor, photolysis and stratospheric ozone), and emission/chemistry drivers (methane burden, NO and CO emissions, sulfate burden). We have amended the figure captions to be more explicit, and added more details to Section 3 (Approach) as to motivation for splitting the figures in the revised manuscript (page 8, lines 18-21).
"As methane lifetime is determined by temperature and OH, we further refine our analysis by examining the evolution of the sources and sinks of OH, which can be broadly separated into

emission factors (CH_4, CO, NO_x) and climate factors (rate constant, water vapor, lightning NO_x and photolysis).”

p.18071,l.6: "increase in OH" or "positive OH trend"?

Response:
The introduction has been re-organized in the revised manuscript (page 4, line 19).
“…while Dentener et al. (2003) identify water vapor as the major driver of increases in OH from 1979 to 1993 (Table 2).”

p.18073,l.2: personal communication citation not required here, as L. Horowitz is a co-author. (also p.18077,l.19)

Response:
These citations have been deleted in the revised manuscript.

p.18084, l.19: The full ACCMIP project name is repeated here unnecessarily.

Response:
The conclusion section has been re-organized (pages 15-17).

Table S1. Percent variations in τ_{CH4_OH} based on percent changes in Table 5 and sensitivities from Holmes et al., (2012), Table 2. LNO_x is below 500 hPa. We use Land NO_x sensitivity from Table 2 in Holmes et al., 2012.

	TEMP	**CH_4**	**COEMIS**	**NOEMIS**	**LNO_x (below 500hPa)**	**H_2O**
Sensitivity (Holmes et al., 2012)	*-3.0*	*0.31*	*0.11*	*-0.14*	*-0.16*	*-0.32*
HIST	-0.37	33.70	12.96	-46.10	0.40	-0.64
AEROSOL	1.08	0	0	0	1.10	1.86
AEROSOL INDIRECT	0.98	0	0	0	0.75	1.70
ANTHRO	-0.68	33.70	12.96	-46.10	0.29	-1.28
NATURAL	0.12	0	0	0	0.22	0.26
WMGGO3	-2.18	33.76	12.96	-46.10	-0.74	-4.13
RCP2.6	-1.20	-8.40	-3.42	6.51	-0.70	-2.18
RCP4.5	-2.53	-2.82	-4.69	6.31	-1.31	-4.74
RCP4.5*	-1.49	0.09	0	0	-0.38	-2.69
RCP6.0	-3.05	0.71	-1.96	6.50	-1.84	-5.82
RCP8.5	-4.99	30.13	-2.88	4.23	-2.40	-9.95

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Fig S3 Global distribution of percent change in surface OH from 1980 to 2000 relative to 1980.

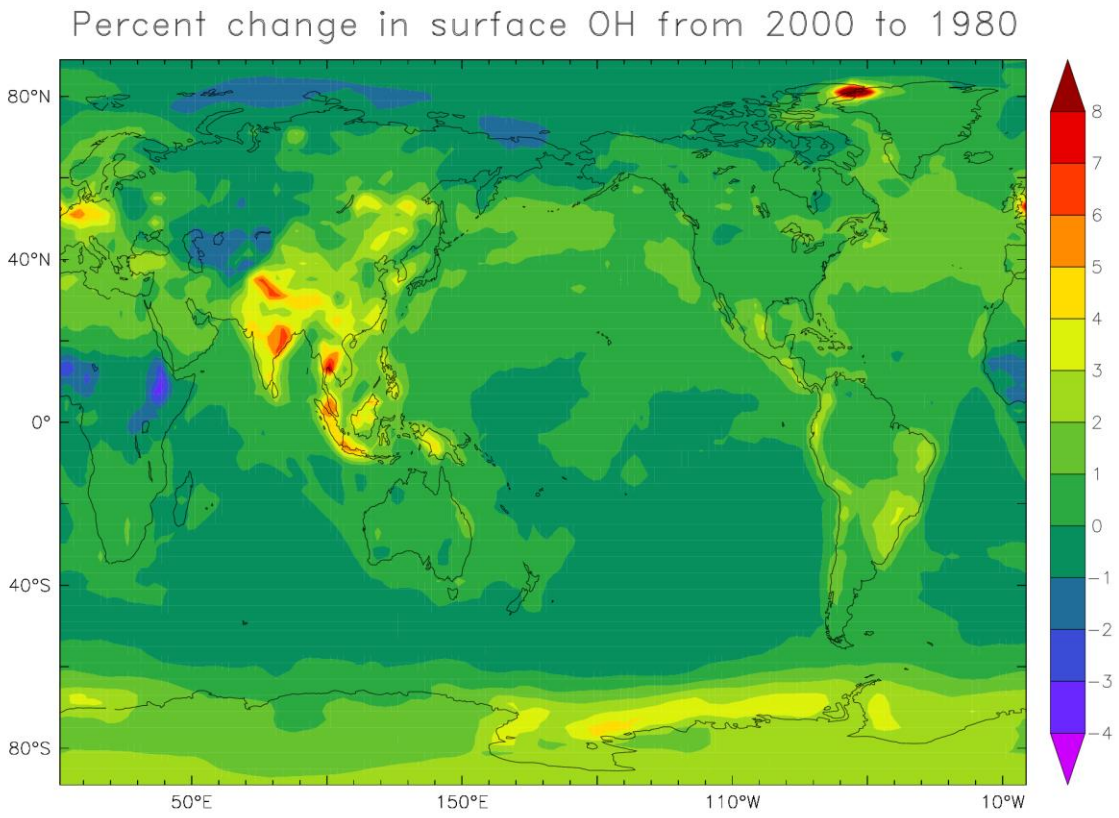

Fig S4 Ratio of tropics (30S:30N) to global surface NO_x emissions from 1980-2005 in HIST.