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 #1

This is a valuable paper analyzing what controls the atmospheric lifetime of methane (CH4) from pre-industrial (1860) to present and into the future (2100, based on the RCP scenarios). Its strengths are a thorough analysis of the GFDL climate-chemistry simulations for the current IPCC/CMIP5 experiments - this is great because it is a consistent comparison in terms of model formulation and different diagnostics, but it is also a weakness because it applies to only one model. The strengths clearly outweigh the weaknesses in terms of being useful to the community. The model experiments and basic analysis are adequate for publication, and no new runs are needed. We do need more details in some places and a better, cleaner layout in others so that the paper is easier to follow.
A detailed critique and suggestions follow the paper:

Response:
Thanks for recognizing the value of this work.

Abst. The aerosol interactions in this model are confusing and need to be stated clearly up front. To say the AIE plays a significant role climate and . . . needs a more explicit statement - i.e. through what? The aerosols influence chemistry through direct reactions, photolysis, and so forth. This does come out later, but make the caveats up front.

Response:
We have expanded this sentence in the abstract to be more explicit with regard to the role of the aerosol indirect effect in the revised manuscript (page 1, lines 26-28; page 2, line 1).
"Sensitivity simulations with CM3 suggest that the aerosol indirect effect (aerosol-cloud interactions) plays a significant role in cooling the CM3 climate. The projected decline in aerosols under all RCPs contributes to climate warming over the next century, which influences the future evolution of OH and τ_{CH4_OH}***."***

The percentages on CH4 lifetime change in the abstract are welcome, but be consistent - e.g., start with the % change for each scenario, then discuss cause. It is not clear what is meant by "only well mixed GHG" since these gases all affect the chemistry directly, especially the stratosphere and is this included? Or do you mean that the "well mixed GHG forcing" is held constant?

Response:
We have changed this section of the abstract to first list percent change and then cause, and also to indicate only well-mixed greenhouse gases for radiative forcing are held constant in the RCP4.5* experiment (page 2, lines 1-7).
"Projected changes in τ_{CH4_OH} from 2006 to 2100 range from -13% to +4%. The only projected increase occurs in the most extreme warming case (RCP8.5) due to the near-doubling of the CH_4 abundance, reflecting a positive feedback on the climate system. The largest decrease occurs in the RCP4.5 scenario due to changes in short-lived climate forcing agents which reinforce climate warming and enhance OH. This decrease is more-than-halved in a sensitivity simulation in which only well-mixed greenhouse gas radiative forcing changes along the RCP4.5 scenario (5% vs. 13%)."

The abstract is not the place to discuss future work, leave that for discussion section.

Response:
This sentence has been removed in the revised manuscript.

p.3-4. The budgets for CH4 based on observations really should be up front here. You give some references, but need the latest Montzka, Krol et al Science paper on OH and the CH3CCl3 decay and the Prather et al 2012 GRL paper on using this data to estimate the CH4 budget with uncertainties. Both papers should be much better values for the CH4 lifetime than those quoted here. The second paper clearly lays out the components in the CH4 lifetime (strat, soils, trop-Cl) that are often forgotten in this paper. To avoid confusion and having people pull the wrong numbers from this paper, you really must say something like "CH4 lifetime against trop OH loss" in every paragraph where you quote CH4 numbers. Otherwise people will pull some of your numbers (e.g., 8.44 y vs 7.82 yr on p.17) and think that these are reasonable absolute numbers compared with the total CH4 lifetime of 9 yr in Prather et al. 2012, but in reality these are OH-lifetimes and should be compared with 11 yr based on the CH3CCl3 decay. I am not dinging the GFDL model for the short OH-lifetime, most models suffer from some large bias in CH4 lifetime that we do not fully understand yet, but at least the values should be carefully labeled.

Response:
We have changed the manuscript title to "Climate versus emission drivers of methane lifetime against loss by tropospheric OH from 1860-2100" and the notation of methane lifetime against loss by tropospheric OH to be τ_{CH4_OH}. In addition, the introduction (pages 2-5) has been reorganized and we reference the recent Prather et al. (GRL, 2012) tropospheric methane lifetime (page 3, lines 5-7).

p.4. Why discuss Staffelbach at all, since the polar regions are really irrelevant to the CH4 lifetime?

Response:
We have revised the introduction in the amended manuscript (page 4, lines 24-26).
"Fewer constraints exist for the change in OH since the pre-industrial period, with estimates ranging from a 33% decrease to a 7% increase (cf. Table 1), although a recent multi-model study suggests little change (Naik et al., 2012b)."

p.5. "Here we investigate. . ." This section is key to what is in the paper - please move it to the number 1 or 2 paragraph of the Intro.

Response:
We have reorganized the introduction in the revised manuscript (page 2, lines 21-28).

p.6. Lightning NOx (LNOx) is one of the major problems in projecting tropospheric chemistry. This paper is not going to solve that problem, but I think it is vital (and beneficial to all) that a critical analysis of what LNOx did/does to the CH4 OH lifetime IN THIS MODEL is presented. For one, how much does the NOx source change, latitudinal effects, and what effect different LNOx has on the CH4 lifetime.

Response:
Labrador et al. (GRL, 2004; ACP, 2005) demonstrated that global mean OH is sensitive to the magnitude and vertical distribution of LNO_x, and we agree that this is an important question to address. However we do not have simulations on hand with the CM3 model that isolate the effects of LNO_x on τ_{CH4_OH} in CM3, and thus this request is beyond the scope of our current study. Figure S1 (below) shows that the tropical to global LNO_x ratio is relatively stable throughout HIST, fluctuating between 0.876-0.896, and over recent decades has declined back to a ratio similar to the pre-industrial, so we do not see much evidence for latitudinal effects in LNO_x driving the overall change compared to the large influence from anthropogenic emissions. If we combine the sensitivities from Table 2 of Holmes et al. (ACPD, 2012) with our percentage changes in Table 5, in all scenarios we conclude that there is a larger influence on τ_{CH4_OH} from water vapor than from LNO_x, both of which are responding to changes in temperature (we include our calculations in Table S1 below). We include this point in the text on page 16, lines 15-19.

p.7/25 - Note these RCP forcings are only "nominal" as the scenarios cannot and do not specify the RF for aerosols and ozone.

Response:
The IIASA website states that the "the radiative forcing estimates are based on the forcing of greenhouse gases and other forcing agents - but does not include direct impacts of land use (albedo) or the forcing of mineral dust." The radiative forcing (RF) values reported for the RCPs therefore attempt to include the RF for aerosols and ozone though this is very low in 2100 due to the large reductions across all RCPs (van Vuuren et al.,

Clim. Change, 2011). We have added "nominal" to the text as RF values computed from model results may differ somewhat to the RCP RF estimates (page 7, line 7).
"The scenarios are labeled according to the year 2100 nominal radiative forcing (RF), which ranges from 2.6 to 8.5 Wm^{-2} (Moss et al., 2010)."

p.8/eqn 1 - the notation for this lifetime should make it clear that it is the OH-only.

Response:
The notation has been changed to τ_{CH4_OH} throughout the revised manuscript.

p.9/3-7. This 8.1 yr is OK and within the range, but it is much lower that recent estimates of 11 yr.

Response:
The Martinerie et al. (1995), Unger et al. (2009) and Sövde et al. (2011) values listed in Table 1 and in text (page 8, lines 5-6) are all for *pre-industrial* tropospheric or chemical methane lifetime. Our model's *pre-industrial (1860)* value for methane lifetime against OH loss is within the range of these studies (page 8, line 4).

p.11/6-21. Can you start by listing what effects are or are not included for aerosols in this model. The use of AEROSOL and AEROSOL INDIRECT is confusing here, especially as the issues of chemistry are more complex as to what is included. J's are not, but cloud changes are? The current tables did not help me here.

Response:
We have amended Section 2 of the manuscript to more thoroughly describe the effects included for aerosols in the model (page 5, lines 18-23; page 6, lines17-24).

p.11/23. Use the same pair of words consistently (increase/decrease) to describe changes in the lifetime, bringing in "shortened: is confusing. It helps the reader.

Response:
We have amended the revised manuscript (page 10, line 22).
"Indeed, τ_{CH4_OH} is considerably decreased in WMGGO3 …"

p.11/29. It is important to have a quantitative understanding of just how important a 4.6% change in LNOx is, since this is only 0.025 Tg-N ?? Trivial compared with surface sources.

Response:

We agree with the referee that this is negligible compared to the surface sources. Assuming a τ_{CH4_OH} sensitivity of -0.16 for LNO_x as in Table 2, Holmes et al. (ACPD, 2012), the 4.6% change in LNO_x in WMGGO3 would account for less that 1% variation in τ_{CH4_OH} over the historical period (Table S1). We have removed this from the text in the revised manuscript (page 10, lines 25-28).
"In this ensemble, the changes in CH_4 burden, CO and NO_x emissions are the same as in HIST and ANTHRO (Tables 3, 5), but rising temperatures (~2K by 2005) and the associated rise in water vapor increase OH and thus drive τ_{CH4_OH} down in WMGGO3 (Table 5), a negative feedback on climate."

p.12/10. Pinatubo also had impact on J's - what did that do?

Response:
Aerosols (including volcanic aerosols) do not have a direct radiative effect on photolysis rates in our model, but the changes in stratospheric ozone column induced by the volcanic aerosols do affect the J's. Photolysis decreased after Krakatoa and Agung (prior to CFCs), but increased after El Chichon and Pinatubo (Fig. 4c). Any influence from the decrease in $J(O^1D)$ after Pinatubo is clearly offset by the climate responses to the cooler temperatures since the methane lifetime decreases.

p.12/5. I an not sure why you want the correlations listed in Table 4, but the reasoning here as to why there is no correlation in this case (trends?) makes no sense to me, can you make the logic more simple. The only obvious reason to me is that other forces of variability are confounding the obvious correlation that should exist.

Response:
We have attempted to clarify. Table 4 indicates a lack of correlation of OH below 500hPa with τ_{CH4_OH} in WMGGO3, even though these should be well-correlated. Fig. S2 (below) shows a scatterplot of OH below 500hPa versus τ_{CH4_OH} for the WMGGO3 ensemble mean. Colored dots indicate different time periods within the simulation: 1860-1920 (red), 1921-1970 (green), 1971-2005 (blue). While these different periods give good correlations of τ_{CH4_OH} against OH, the correlation over the entire simulation period is negligible (page 11, lines 12-21).
"In WMGGO3, temperature, together with water vapor, rises gradually before the 1960s and more sharply after. From 1921-1970, increasing CO emissions, CH_4 burden and decreasing photolysis rates drive OH down, counterbalancing the competing influences of rising temperature, water vapor, NO_x and LNO_x. In the late 20th century, τ_{CH4_OH} falls continuously as temperature and water vapor continue to rise even more rapidly (Fig. 3a, 4b). τ_{CH4_OH} is well correlated with OH within time periods (1860-1920, 1921-1970, and 1971-2005), but the different rates of change of temperature and OH within these time periods results in the lack of correlation over the entire length of the simulation (Table 4). As noted in Section 3, τ_{CH4_OH} in WMGGO3 is driven mostly by temperature through changes in the rate constant, and associated water vapor."

p.12/25. Section 4.3 brings up a serious problem as there is no such decline in the CH3CCL3 observed lifetime (Montzka 2011) - this needs to be a discussion point. I admit that most models get this, but why ? and why is it wrong?

Response:
We have revised this portion of the manuscript to look at the period from 1980-2000 instead and find a 3% increase in OH, which is consistent with a recent multi-model modeling study (Naik et al., ACPD, submitted). Montzka et al. (Science, 2011) suggests that observational estimates of CH_3CCl_3 prior to 1998 may have been overestimated and obtain an interannual variability of 2.3 ± 1.5% from 1998-2007 (page 12, lines 5-12).
"We find a small (~3%) increase in modeled OH over the 1980-2000 period, which is consistent with the multi-model ensemble value of 3.5 ± 2.2% reported by Naik et al. (2012b) This small positive OH trend in recent decades is consistent with the current generation of models (Table 2), but disagrees with the negative OH trends inferred from observations of CH_3CCl_3 and inversion studies (e.g., Prinn et al., 2001, Bousquet et al., 2005). More recently, however, Montzka et al. (2011) suggest that uncertainties in measurements of CH_3CCl_3 prior to 1998 may have contributed to the wide OH variability reported in previous studies; they find a small observationally-derived OH interannual variability of 2.3 ± 1.5% from 1998-2007.

p.13/4. Do not aerosols affect clouds? I thought the AIE was through this.

Response:
We have revised this sentence to be more explicit in the revised manuscript (page 11, lines 26-28, page 12, line 1).
"Since the 1970s, we also note an increase in $J(O^1D)$ - presumably driven by depletion of stratospheric ozone (Fig. 4c, d) as aerosol changes do not directly affect photolysis rates in these simulations, except through aerosol-induced cloud changes. The increase in $J(O^1D)$ also enhances OH formation and shortens τ_{CH4_OH} (Wang et al., 2004) "

p.13/10. As noted earlier, the OH lifetime of 9 yr here is much lower than the current best numbers noted above. I am not sure that some of these comparisons are for total lifetime. Why quote a trend that is too small to be meaningful.

Response:
We have revised Section 4.3 as noted above (page 12, lines 5-12).

p.14/3-18. Very good discussion, clear.

Response:
Thanks for the supportive comment.

p.15. avoid "shortening"

Response:
We have changed this in the revised manuscript (page 14, lines 4-5).
"In RCP4.5*, we again find a decrease in τ_{CH4_OH}, …"

p.18. It would be best to give %'s for each scenario (to nearest % only, not 4.3%), then discuss the differences. Also you need to assess the % changes in the total CH4 lifetime since that is what determine the future methane abundance. So come up with some ideas for soil, strat, trop-Cl. I would also say that "Further study is needed on LNOx" - I think as a community we have not established clear metrics and diagnostics that let us understand what is important here for future OH.

Response:
We have made appropriate changes in the revised manuscript (page 16, lines 1-6; page 17, lines 3-7).
"Relative to the simulated historical change (+5%), the projected changes in tropospheric methane lifetime span a fairly wide range over the next century: -9%, -13%, -6%, +4% for RCP2.6, RCP4.5, RCP6.0, and RCP8.5 respectively (changes are 2081-2100 average minus 2006-2025 average). Assuming soil, stratospheric and tropospheric chlorine sinks (120 yr, 150 yr, 200 yr respectively) remain constant, projected changes in total methane lifetime are -10% (RCP2.6), -15% (RCP4.5), -8% (RCP6.0) and 0.1% (RCP8.5)."

"Developing observation-based constraints on distributions and trends of LNO_x, as well as water vapor, photolysis rates, and feedbacks from the biosphere would help clarify the relative importance of these climate factors on the future evolution of τ_{CH4_OH} as compared to changes driven by anthropogenic emissions."

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. S1 Ratio of tropics (30S:30N) to global lightning NO_x emissions below 500hPa in HIST.

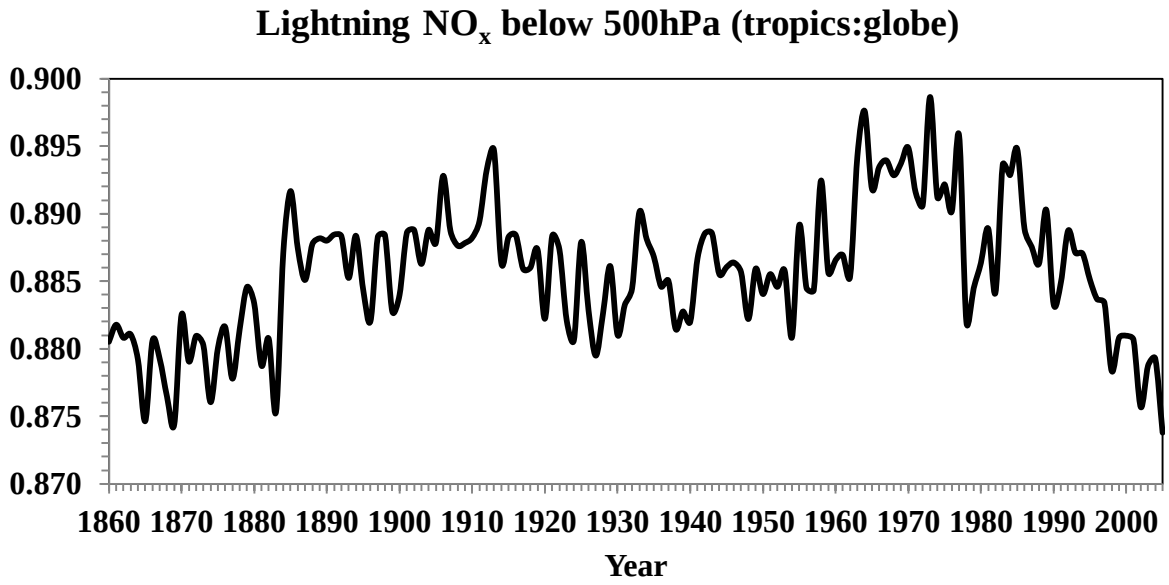

Fig. S2 Scatter plot of τ_{CH4_OH} versus OH below 500hPa for WMGGO3. Colored circles denote different time periods within the simulation: 1860-1920 (red), 1921-1970 (green), 1971-2005 (blue).