

We thank the reviewer for the helpful comments. All of them are addressed and answered below.

Comment: This study presents a multi-model comparison of the distribution and trends in hydroxyl (OH) radical and an analysis of how the different OH fields influence atmospheric methane trends over the 2000-2016 period. This paper is very relevant to the current debate in the scientific literature about the drivers of recent methane increase in general and specifically the role of OH in this renewed methane growth since 2007. Generally, the paper is well-organized and addresses scientific questions within the scope of ACP.

The authors have analyzed the free-running coupled chemistry-climate model simulations derived from the CCMI project as well as from three additional models not participating in CCMI. Like the ACCMIP models, the models considered here also produce a wide range of OH distributions and agree on the sign of change in OH over the 1980 to the present (2010) time period. The authors largely attribute the reasons for intermodel diversity in the simulated mean OH distributions to differences in the representation of nitrogen oxide, and natural emissions of NMVOCs and their chemistry. I felt that the analysis could be a bit deeper, especially the role of differences in lightning NO_x and stratospheric ozone could be investigated. The authors also do not evaluate the models against proxy observational constraints (such as ozone, water vapor, CO, NO_x column, methane lifetime) to assess their skill in simulating OH levels. How do we know which model is closer to reality? I acknowledge that this is a difficult question to answer but feel that some effort is needed to evaluate the models. Finally, the authors attribute the increasing trend in tropical OH over 2000 to 2010 to increasing NO_x emissions, again without an in-depth quantitative analysis. I have highlighted these and other issues in my specific comments below. I recommend the publication of this paper after it has been revised to include additional evaluation and analysis.

Response:

In this paper our aim is to estimate the impact of OH distribution in space and time on methane changes since 2000 using an ensemble of state-of-the-art atmospheric models. We acknowledge that the depth of analysis of the root causes of what we find here can be increased the lack of evaluation of these models in our paper. This is because finding the “best model” is very difficult, regarding the multiple criteria to take into account even when only looking at OH, that we use such an

ensemble. Doing a full evaluation of these models is beyond the scope of our study but to better explain what we find and strengthen a bit model evaluation, we now compare ozone simulated by the CCM1 models with TOMS/SBUV observations (Fig. S4) and we have calculated tropospheric chemical lifetime in table 4. In addition, the CO column and tropospheric O₃ column have already been evaluated by Strode et al. (2016) and Revell et al. (2019), we also cite these two references in the text.

We have added in the text:

” The tropospheric chemical CH₄ lifetime of the models that provided CH₄ chemical loss data are 8.7 ± 1.1 yr. Both the multi-model mean and the (large) range of [OH] as well as tropospheric CH₄ chemical lifetime are consistent with previous multi-model results given by the ACCMIP project (Naik et al., 2013; Voulgarakis et al., 2013), as well as with inversions based on MCF observations (Bousquet et al., 2005; Rigby et al., 2017).”

“Previous studies have attributed the inconsistency between the simulated and the observed OH N/S ratios to a model overestimation of O₃ and underestimation of CO over the Northern Hemisphere (Naik et al., 2013; Young et al., 2013; Strode et al., 2015), which also have been reported for CCM1 models (Strode et al., 2016; Revell et al., 2018), ...”

All of other more specific comments have been addressed in the revised manuscript. Please see out itemized responses below.

Specific Comments:

Comments: L70: Levy (1972) is the wrong reference here. The correct reference should be Levy (1971).

Response: Thanks for pointing it out. The reference is now corrected.

Comments: L71: Atomic excited oxygen is denoted as O(1D). Please revise O^{1D} throughout the manuscript.

Response: Changed as suggested

L99-103: The tropospheric methane chemical lifetime against OH loss from ACCMIP models was calculated as the global annual mean **atmospheric** methane burden divided by annual mean methane tropospheric loss by OH. Please revise.

Response: Changed as suggested

Comments: L112-114: “The precise scenario...” sounds odd. Please revise to “The precise reasons for stagnation and renewed growth of methane still remain unclear...”

Response: Changed as suggested

Comments: L127-130: With their chemistry model, Dalsoren et al (2016) actually simulate an ~8% increase in OH over the 1970 to 2012 time period. Not only is this trend as large as that inferred by Turner et al and Rigby et al, but is also in a completely opposite direction and is also contrary to the no OH trend deduced by Nicely et al (2018). I think the point that there is a mismatch in the OH trends inferred from observations and simulated by global models should be explicitly highlighted.

Response: We add in the text “Meanwhile, not only the OH trend calculated by atmospheric chemistry models cannot reach consensus, but it can also be different from the OH trend inferred by top-down approaches from observations. Indeed, Dalsøren et al. (2016) simulated ~ 8% increase in OH during 1970 to 2012, while other models mostly calculated only a small increase of [OH] (decrease in CH₄ lifetime) or no trend in [OH] from 1980s to 2000s (e.g. Voulgarakis et al., 2013; Nicely et al., 2018). Top-down observation-constrained approaches (e.g. Rigby et al., 2017) tend to find flat to decreasing OH trend over this period but with larger year-to-year variations than models.”

Comments: L141-144: The configuration of ACCMIP simulations was not ideal for assessing interannual variability in OH due primarily to the fact that these simulations were not performed continuously from year to year but were timeslices with emissions as well meteorology characteristic of the decade being simulated. So in that sense, the ACCMIP simulations did allow one to assess decadal (or multi decadal) but not year to year variability. I think this sentence should be revised to something like this for clarity: “Year-to-year integrations of CCMI and INCA models driven by time-varying emissions and meteorology facilitate the investigation of interannual variability in OH which was not possible using the ACCMIP time-slice simulations”

Response: “Changed as suggested”

Comments: L158-159: From the perspective of understanding the influence of OH changes on methane, I think the analysis of REF-C1SD is more relevant because it will likely reproduce (or be close to) observed changes in various climate related factors that influence OH (e.g., humidity, temperature) as well as chemical composition since it is nudged to observed meteorology. I would highly encourage the analysis of REF-C1SD simulations if results from at least 5 of the models analyzed here are available.

Response:

We add in the main text:” The models of REF-C1SD experiment are nudged towards reanalysis datasets. The REF-C1SD experiment is not analyzed in the main text since it has been conducted by only part of the models and covers a shorter time period. A comparison of spatial and vertical distributions of OH fields from REF-C1 experiment with that from REF-C1SD reveals only small latitudinal differences (<10%, see Section S1).”

We have added Section S1 with table S1, S2, and Fig. S1 in the supplemental:

“S1 OH fields from CCM1 REF-C1 experiments.

We compare spatial and vertical distributions of OH fields from REF-C1 (main text) with that from REF-C1SD to assess influences from dynamic biases. Of CCM1 models included in this study, 7 models conducted REF-C1SD experiments (EMAC offers fields at two different model resolutions). Fig. S1 shows the spatial distributions of the volume-weighted tropospheric mean [OH] averaged from 2000 to 2010 simulated by REF-C1SD experiments, Table S1 summarizes their inter-hemispheric ratios and mean values over four latitudinal bands. The volume-weighted mean [OH] averaged over the troposphere and over three pressure latitudinal intervals are calculated in Table S2. By comparing Fig. S1, table S1, and table S2 with Fig. 2, table 3, and table 4, respectively, we find that OH fields from REF-C1 and REF-C1SD experiments show similar spatial and vertical distributions. Only CESM and MOCAGE simulated recognizable different N/S ratios (small differences within 0.1-0.2) by REF-C1 and REF-C1SD experiments, and the differences in mean OH over four latitudinal bands and latitudinal intervals are within 10%.”

Table S1. Inter-hemispheric ratios (N/S) of hemispheric mean OH and volume-weighted tropospheric mean [OH] for four latitude bands (in $10^5 \text{ molec.cm}^{-3}$) averaged over the years 2000 to 2010 from CCM1

REF-C1SD experiment.

OH fields	N/S ratio	90°S-30°S	30°S-0°	0°-30°N	30°N-90°N
		(10 ⁵ molec.cm ⁻³)			
CESM1-CAM4Chem	1.3	6.3	13.3	15.9	8.7
CESM1-WACCM	1.2	6.6	13.3	15.9	9
CMAM	1.2	5.8	12.8	13.7	8.1
EMAC-L47MA	1.2	6.4	14.1	15.6	8.5
EMAC-L90MA	1.2	6.2	13.5	15.1	8.4
MOCAGE	1.3	6.1	12.1	14.5	8.9
MRI-ESM1r1	1.2	4.7	14.2	15.7	6.9
UMUKCA-UCAM	1.3	5.6	13.9	15.2	10.1

Table S2. Global mean [OH] averaged over the troposphere and three vertical pressure levels (in 10⁵ molec cm⁻³) over the years 2000 to 2010 from CCM1 REF-C1SD experiment.

	Tp ¹	750	500	250
CESM1-CAM4Chem	11.1	12.1	13.1	11.5
CESM1-WACCM	11.2	12.3	13.4	11.8
CMAM	10.1	14.3	10.9	10.8
EMAC-L47MA	11.2	12.4	12.4	11.1
EMAC-L90MA	10.9	12.3	12.1	10.2
MOCAGE	10.4	19.2	15	7.3
MRI-ESM1r1	10.5	12.4	10.8	9.7
UMUKCA-UCAM	11.2	16.0	12.4	10.6

¹ Tp refers to the volume-weighted tropospheric mean [OH], 750 refers to the volume-weighted average from the surface to 750hPa, 500 refers to the volume-weighted average from 750hPa to 500 hPa, and 250 refers to the volume-weighted average from 500 to 250hPa.

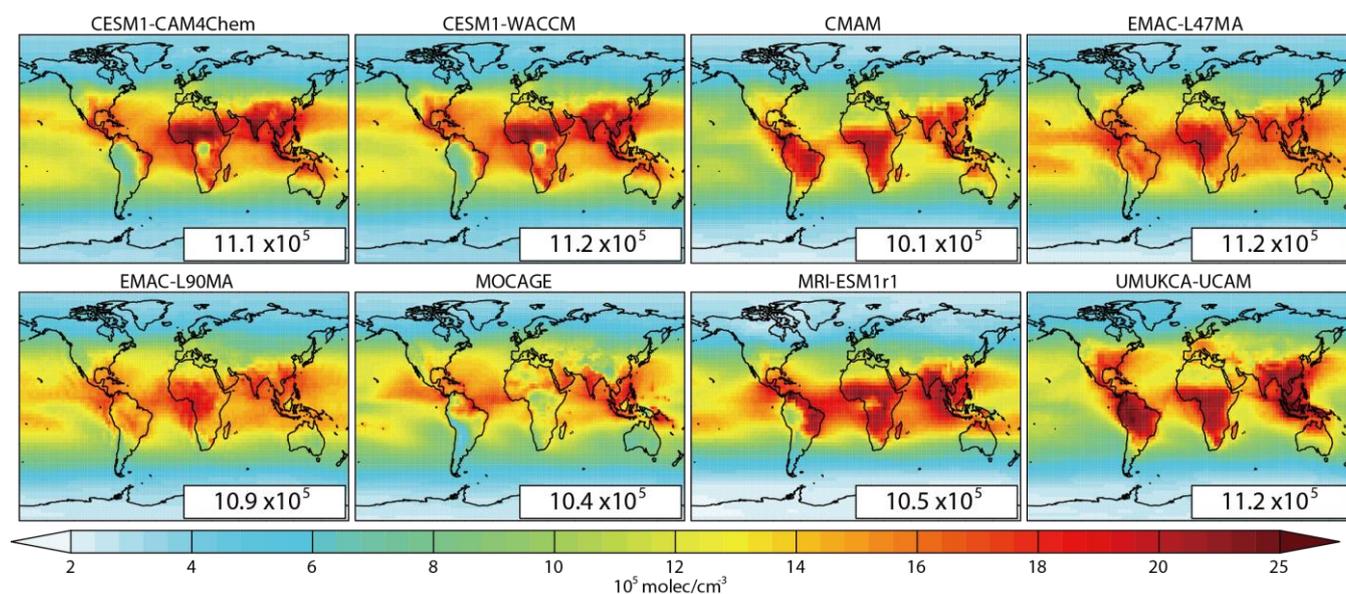


Figure S1. The spatial distributions of volume-weighted tropospheric mean OH fields CCM1 REF-C1SD experiments averaged for 2000-2010. Global mean values (10⁵ molec cm⁻³) are shown as insets.

Comments: L168-169: Please clarify what is tropospheric ozone chemistry coupled to - stratospheric chemistry, physical climate?

Response: We change “coupled” to “detailed”

Comments: L172-173: Define the chemical names before using them HCHO (formaldehyde) and C₅H₈ (isoprene).

Response: Thanks for pointing it out. We add the chemical names as suggested.

Comments: L177-178: I think it would be helpful to clarify that after year 2000, the MACCcity emissions (therefore REF-C1 simulation) follow the RCP8.5 scenario. It would also be helpful to clarify whether biomass burning emissions are also from MACCcity and if they vary from year to year.

Response: We have clarified this by adding :

L185: “...(which follow the RCP8.5 inventory after 2000),...” after the REF-C1 experiment continued to use the MACCcity inventory.

L188: “Biomass burning emissions used in REF-C1 are RETRO inventory (Schultz et al. 2008) before 1996 and GFEDv3 inventory (van der Werf et al., 2010) for 1997-2010 with interannual variability.”

References:

” Schultz, M. G., Heil, A., Hoelzemann, J. J., Spessa, A., Thonicke, K., Goldammer, J. G., Held, A. C., Pereira, J. M. C., and van het Bolscher, M.: Global wildland fire emissions from 1960 to 2000, *Global Biogeochemical Cycles*, 22, 10.1029/2007gb003031, 2008.”

“van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), *Atmos. Chem. Phys.*, 10, 11707-11735, 10.5194/acp-10-11707-2010, 2010.”

Comments: L190: According to the description in Morgenstern et al (2017), methane concentrations prescribed in the CCMi models vary in time following the RCP6.0 contrary to the specification (global

mean of ~1750ppbv averaged from over 2000-2010) described here. Please clarify.

Response: We add “vary in time” in the text.

Comments: Section 2.1: It would be helpful if the authors could clarify how tropopause is defined (in order to calculate tropospheric OH from the models) somewhere in this section.

Response: We add in the L233 “We analyzed spatial distributions and annual variations of OH fields by calculating volume-weighted tropospheric mean [OH] with tropopause pressure calculated using WMO tropopause definition on 3D temperature for each model (World Meteorological Organization, 1957).”

Reference:” World Meteorological Organization: Definition of the tropopause, Bulletin of the World Meteorological Organization, 6, 136–137, 1957.”

Comments: Section 2.2.1: The motivation for using this model and simulations became clear to me only after I read section 2.2.2. I think the motivation for using this model with methane emissions should be clarified up front in this section.

Response:

We clarify by changing L143-146:

“Using this ensemble of OH fields, our aim is to estimate a range for the contribution of changes in [OH] to the atmospheric CH₄ variations since 2000, and to relate this contribution to characteristics of the different OH fields.” to “We then conducted an ensemble of CH₄ simulations with different OH fields using the LMDz chemistry-transport model to estimate a range for the contribution of changes in [OH] to the atmospheric [CH₄] changes since 2000, and to relate this contribution to spatio-temporal characteristics of the different OH fields.”

Comments: L232: access should be replaced with assess.

Response: Thanks for pointing it out. The typo is now corrected.

Comments: L238-239: It is mentioned above that the offline LMDz5B is run with OH field from CCMI models. Does it also use O(1D) fields from the CCMI models?

Response: We state in the text L246: “To assess the influences of OH only, all LMDz simulations used the same O(1D) fields generated by INCA model simulations.”

Comments: L245-248: Please clarify which chemistry module is used in the setup here.

Response: We now state: “Chemistry module applied here is the simplified chemistry module SACS (Pison et al., 2009)”

Comments: L301, L552: Please confirm if this is indeed 1‰ yr⁻¹ or 1% yr⁻¹.

Response: To make it clear, we change all “1‰” to “0.1%”.

Comments: L313-314: Revise “...which overestimation of [OH]...”

Response: We revise this sentence to “SOCOL, which overestimation of [OH] have been reported by Staehelin et al. (2017), simulated the highest [OH].”

Comments: L317: IPCC (2011) is missing from the reference list. Also suggest citing the IPCC chapter (though I do not think there is a 2011 IPCC report relevant here) rather than the whole report.

Response: Thank you for pointing it out, we remove the reference IPCC 2001 here.

Tables 3, 4, 5, and 7: It would be helpful to provide a quantitative measure of model spread (e.g., coefficient of variation or range) at the end of the columns which would make it easier for the reader to quickly get an estimate of intermodel diversity.

Response: we now present Multi-model means with their standard deviations in table 3, 4, 5, and 7 by adding lines of “Mean ± stand. dev.” in the corresponding tables.

Comments: L336-347: In addition to individual model maps, it would be helpful to have a map of the standard deviation of OH concentrations across the models considered here to clearly see the regions of high/low spread. Also, could variability in lightning NO_x emissions across models be a cause of the model spread in OH? What about differences in simulated stratospheric ozone across the models; relevant for southern hemisphere OH differences? I think a deeper analysis of the factors that influence OH is needed to assess the reasons for the spread in OH fields.

Response:

(1) To answer about the “map of the standard deviation of OH concentrations”, we have added **Fig.S2 in the supplement:**

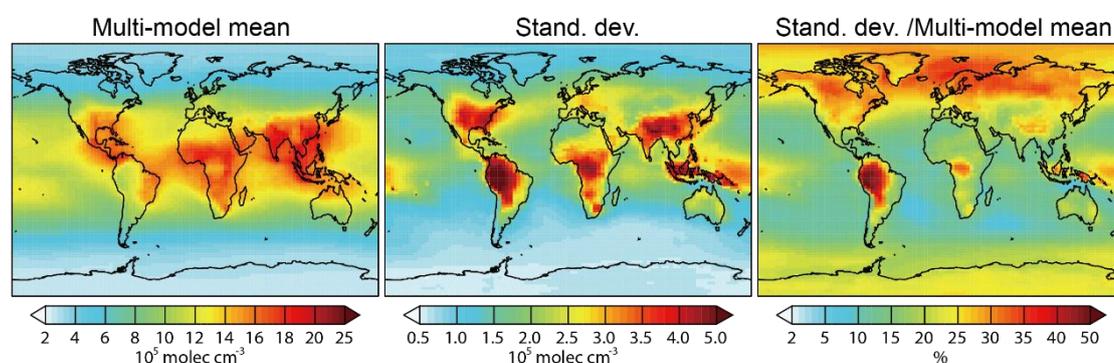


Figure S2. Multi-model mean (left), standard deviation(middle). and standard deviation relative to multi-model mean of tropospheric mean OH fields shown in figure 2.

We cite this figure by: “We further assessed the simulated OH spread by comparing the detailed spatial distributions of OH fields in Fig. 2 and Fig.S2”

We also add in the text L375:

” Tropospheric mean [OH] over the Amazon forest show large variations of $>5.0 \times 10^5$ molec cm^{-3} , representing more than 50% to the multi-model mean(Fig.S2). In a more diffuse way, high latitudes of the northern hemisphere also contribute to model spread (25-35% of the model mean, Fig. S2). Besides these, inter-model differences also exist over the open ocean (up to 25% of the model mean, Fig.S2).”

(2) To answer about the “could variability in lightning NO_x emissions across models be a cause of

the model spread in OH”, we have added table S3 in the supplement:

Table S3. Lighting NO_x emission (Tg N yr⁻¹) over three pressure altitudinal intervals and the total troposphere of CCMI models over 2000-2010.

	Surface- 750hPa	750- 500hPa	500- 250hPa	250- 100hPa	tp
CMAM	0.7	0.4	1.5	1.7	4.2
EMAC-L90MA	0.2	0.5	1.3	1.8	3.7
CESM1-WACCM	0.2	0.6	2.7	0.7	4.2
GEOSCCM	0.2	1.3	3.3	0.8	5.6
MOCAGE	0.3	1.2	2.4	1.0	4.8
MRI-ESM1r1	1.4	0.7	3.2	5.2	10.2
SOCOL3	0.2	0.8	2.1	1.4	4.4

We also add in the main text:

L431-L435: “Lighting NO_x, which are mainly emitted in the middle and upper troposphere, can contribute to inter-model differences in NO and OH distributions (Murray et al., 2013; 2014). We compare lighting NO_x emissions calculated by CCMI models in Table S3. High lighting NO_x emissions simulated by MRI-ESM1r1 above 250hPa can explain high NO mixing ratios and increasing OH with altitude over the upper troposphere for this model (Fig. 3). However, High NO in the lower troposphere simulated by MOCAGE and SOCOL3 are not corresponding to high lighting NO_x emissions in these models.”

L451 “Lighting NO_x emissions range from 3.7-10.2 Tg yr⁻¹(table S3)”

The above text about lighting NO_x emissions also response to comments on L390-410.

(3)”What about differences in simulated stratospheric ozone across the models; relevant for southern hemisphere OH differences?”

To analyse influences on southern hemisphere OH, we have added table S5, which compares stratosphere ozone and O(¹D) photolysis rate for four latitude bands, and also figure S4, which compares total ozone column with satellite observations in the supplement.

Table S5. Tropospheric mean stratosphere ozone and O(¹D) photolysis rate for four latitudinal bands averaged over 2000 to 2010. Multi-model means and standard deviations (Mean ± stand. dev.) are also shown.

	Stratosphere ozone				O(¹ D) photolysis rates (10 ⁻⁵ s ⁻¹)			
	90°S-30°S	30°S-0°	0°-30°N	30°N-90°N	90°S-30°S	30°S-0°	0°-30°N	30°N-90°N
CESM1-CAM4Chem	272	222	225	300	0.8	1.8	1.8	0.7
CESM1-WACCM	261	219	223	286	0.8	1.9	1.8	0.7
CMAM	269	228	230	293	0.8	1.6	1.6	0.6
EMAC-L47MA	298	232	232	299	0.6	1.5	1.5	0.6
EMAC-L90MA	291	233	233	293	0.7	1.5	1.5	0.6
GEOSCCM	249	216	219	286	0.6	1.2	1.1	0.4
HadGEM3-ES	282	245	248	297	/	/	/	/
MOCAGE	212	224	245	280	/	/	/	/
MRI-ESM1r1	280	238	238	301	0.6	1.3	1.3	0.5
SOCOL3	277	238	238	297	0.6	1.1	1.1	0.5
UMUKCA-UCAM	241	236	236	256	/	/	/	/
Mean ± stand. dev.	267 ± 25	230 ± 9	233 ± 9	289 ± 13	0.7 ± 0.1	1.5 ± 0.3	1.5 ± 0.3	0.6 ± 0.1

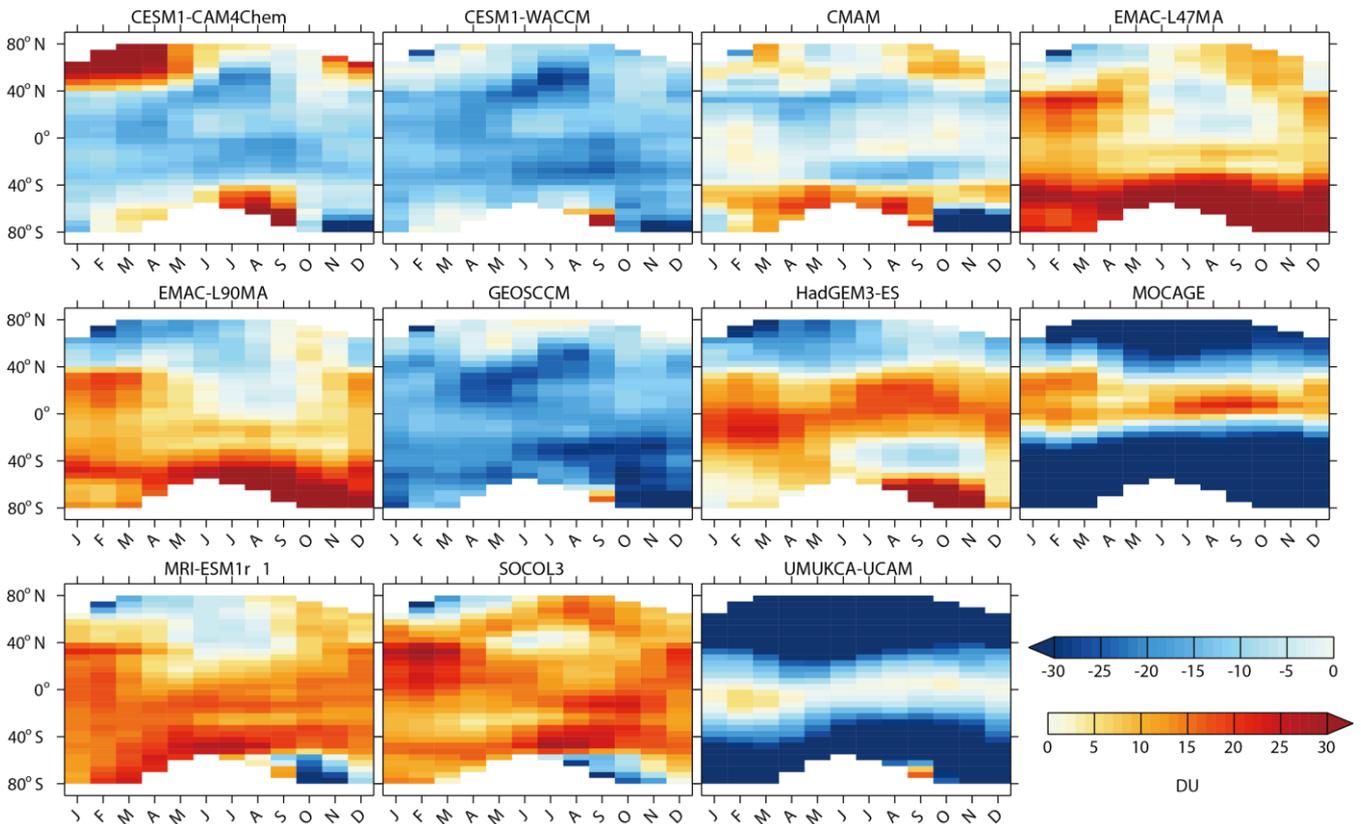


Figure S4. Monthly total column ozone bias from CCM1 simulations averaged over 2000-2010 compared to satellite measurements from Total Ozone Mapping Spectrometer/solar backscatter ultraviolet (TOMS/SBUV) (model minus measurement).

We also add in the main text:

”The stratospheric ozone can contribute to inter-model OH discrepancies through influencing O(¹D) photolysis rates. However, we find that models that simulated lower stratosphere and total ozone column are not corresponding to higher O(¹D) photolysis rates and [OH] (table S5 and Fig. S4), since differences in the photolysis schemes coupled to CCMI models can also influence the calculation of O(¹D) photolysis rates (Sukhodolov et al., 2016).”

Reference: Sukhodolov, T., Rozanov, E., Ball, W.T., Bais, A., Tourpali, K., Shapiro, A.I., Telford, P., Smyshlyaev, S., Fomin, B., Sander, R., Bossay, S., Bekki, S., Marchand, M., Chipperfield, M.P., Dhomse, S., Haigh, J.D., Peter, T., Schmutz, W., 2016. Evaluation of simulated photolysis rates and their response to solar irradiance variability. *Journal of Geophysical Research: Atmospheres* 121, 6066-6084.

Comments: L356: Is it GEOSGCM or GEOSCCM?

Response: We change “GEOSGCM” to “GEOSCCM”, thanks for point out the typo.

Comments: L390-410: Murray et al. (2014) show that lightning NO_x plays a key role in controlling OH as also mentioned on lines 414 to 417. How different/similar are the models in their representation of the vertical distribution of lightning NO_x emissions?

Response: We answer this comment in the response to comments on L336-347.

Comments: Figure 3 would also benefit from plot of standard deviation across models for each region.

Response: We calculated the standard deviation in both figure 3 and figure S3 as suggested.

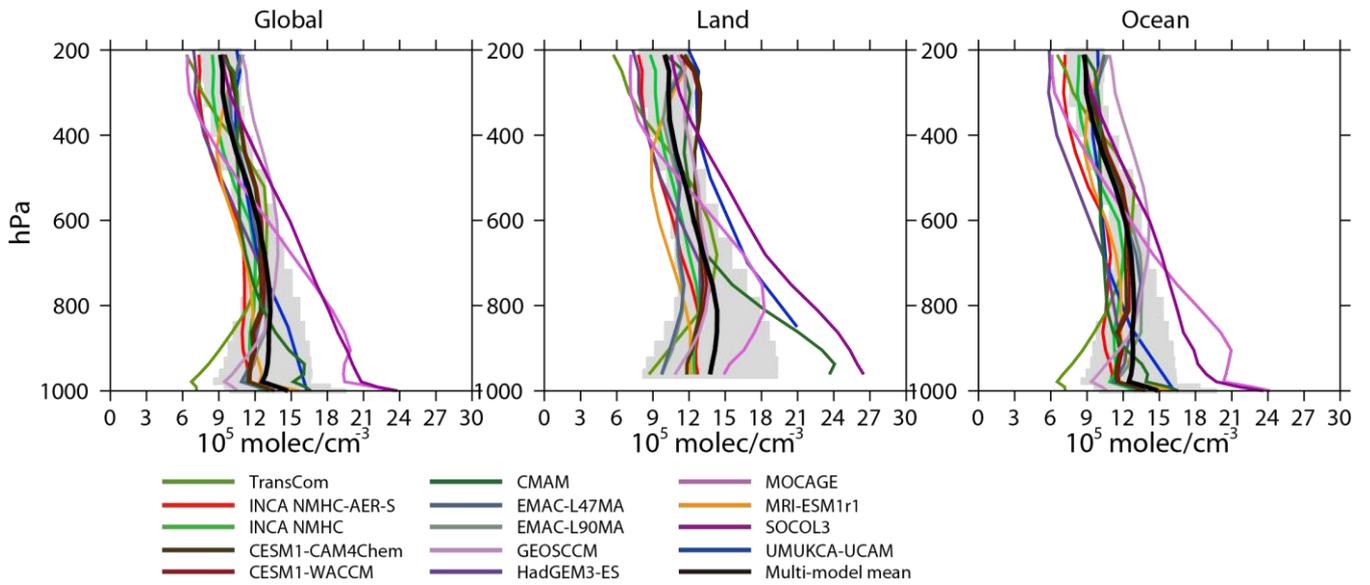


Figure 3. Vertical distributions of [OH] averaged over the globe (left), land (middle) and ocean (right) for 2000-2010. Color lines represent [OH] from individual model simulations, black lines represent multi-model mean values and grey shades represent the standard deviations.

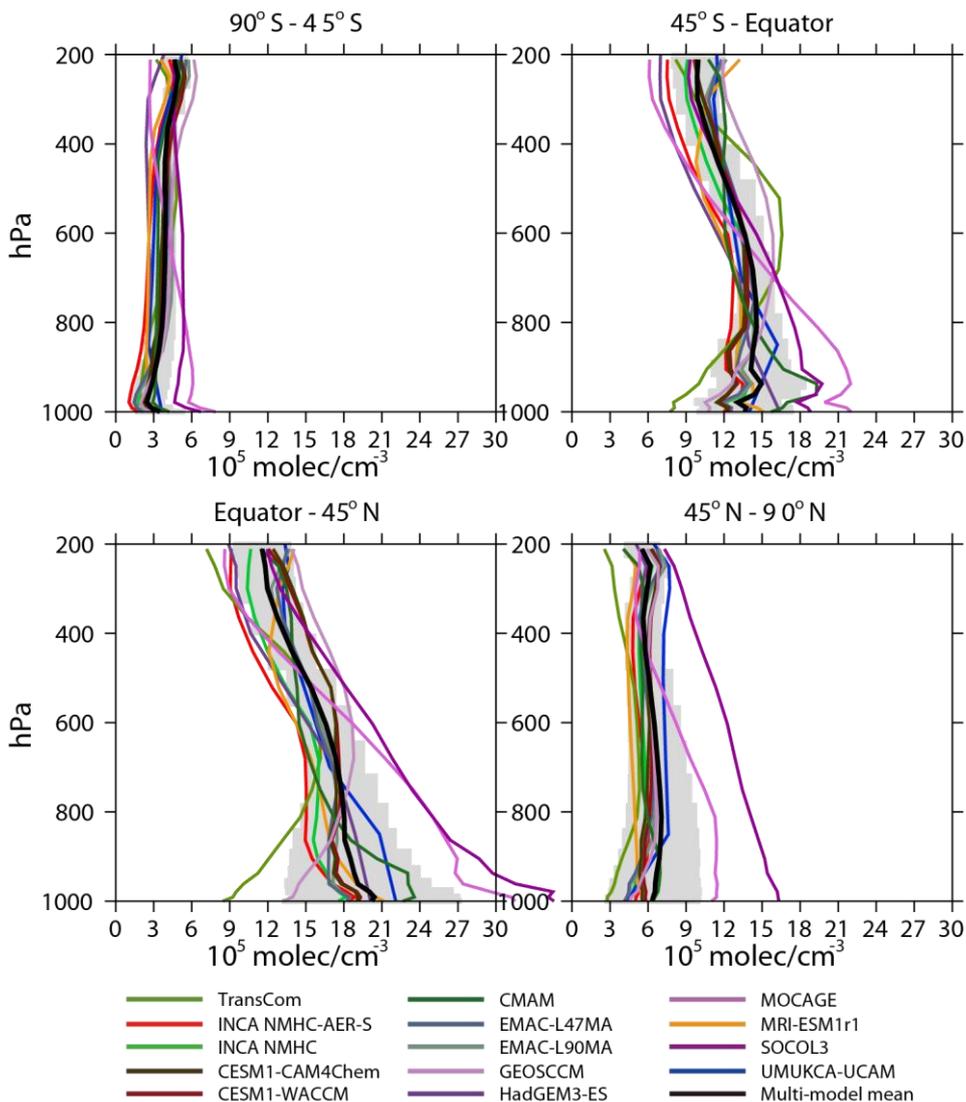


Figure S3. Vertical distribution of [OH] averaged over four latitude bands and over the years 2000 to 2010. Color lines represent [OH] from individual model simulations, black lines represent multi-model mean values and grey shades represent the standard deviations.

Comments: L430-431: What is the mean year to year variation over this period in units of percent?

Response: We add then value as suggested:

“During this period, all OH fields show small year-to-year variations of $1.9 \pm 1.2\%$, remaining within $\pm 0.5 \times 10^5$ molec cm^{-3} .”

Comments: L432-434: Are the numbers in parentheses the changes in OH concentrations from 1960 to 1980? If so, it would be helpful to provide percent changes as well.

Response: We add percent changes as suggested.

“For example, [OH] continuously decrease in the CMAM and HadGEM3-ES simulations ($\sim -0.3 \times 10^5$ molec cm^{-3} ; -3.4%); and increase in SOCOL3 ($\sim +0.6 \times 10^5$ molec cm^{-3} ; $+4.5\%$), UMUKCA-UCAM ($\sim +0.5 \times 10^5$ molec cm^{-3} ; $+4.8\%$), and MOCAGE ($\sim +0.5 \times 10^5$ molec cm^{-3} ; $+4.8\%$) during this 1960-1980,…”

Comments: L447-450: How do the year-to-year variations in OH from CCMI and INCA models compare with the results of Turner et al. (2017), Rigby et al. (2017) and Nicely et al. (2018) using different approaches?

Response: We including the comparisons by re-organizing this paragraph (see also answer to comment L127-130)

“Previous atmospheric chemistry model studies have concluded that anthropogenic activities lead to only a small perturbation of the OH burden, as the increased OH production tend to be compensated by an increased loss through reactions with CO and CH₄ (Lelieveld et al., 2000; Naik et al., 2013). By combining factors that influencing OH, Nicely et al. (2018) modeled a small inter-annual variability of 1.6% during 1980-2015. The year-to-year variations of most CCMI and INCA OH fields are consistent with Nicely et al. (2018), but much smaller than the OH inter-annual variability based on MCF observations (e.g. Bousquet et al., 2005; Montzka et al., 2011), which can reach $8.5 \pm 1.0\%$ from 1980 to 2000 (Bousquet et al., 2005), and $2.3 \pm 1.5\%$ from 1998 to

2007(Montzka et al., 2011), as compared to $2.1 \pm 0.8\%$ and $1.0 \pm 0.5\%$ here for these two periods. As for OH trend, the ensemble of ACCMIP models simulated large divergent OH changes (even in their signs) from 1850 to 2000, but revealed a consistent and significant increase of $3.5 \pm 2.2\%$ from 1980 to 2000 (Naik et al., 2013). Here, for the same period the increase of CCM1 [OH] is $4.6 \pm 2.4\%$, consistent with the ACCMIP project (Naik et al., 2013) and with other atmospheric chemistry model studies (Dentener et al., 2003; John et al., 2012; Holmes et al., 2013; Dalsøren et al., 2016). The slightly increasing [OH] after 2000 inferred here as well as previous model simulations (e.g. Nicely et al., 2018) cannot help to explain stalled and renewed CH₄ growth during the 2000s, as opposed to the decreasing [OH] from mid-2000s calculated by Rigby et al. (2017) and Turner et al. (2017) based on MCF observations. ”

Comments: L455-458: During this time period, there have been increases in water vapor as well (e.g., Dessler and Davis 2010) that could potentially influence OH trends in the tropical regions. Admittedly, the increase in NO_x emissions has been significant, but I am not sure if the analysis presented here can be used to say, with confidence, that the strong positive trend in OH is being solely driven by NO_x emissions. It would be helpful to perform some regression analysis to build confidence in the conclusions here.

Response: We have calculated the trend of stratospheric O₃, specific humidity, CO and NO_x emissions in each grid cell for the CCM1 models to assess the contribution of each factor to OH trends. We cannot do the regression analysis here since only part of the model provide these data and we focus on the spatial distribution of the trends.

We have added figure S6a, figure S6b, and figure S6c in the supplement, and in the main text:

” By comparing spatial distribution of OH trend with specific humidity (Fig.S6a), NO_x and CO emissions (Fig. S6b), and stratospheric O₃ (Fig.S6c), we find that positive OH trend over tropical regions are mainly corresponding to increasing water vapor (Fig. S6a) while faster NO_x emission increases ($>5\% \text{ yr}^{-1}$) than CO ($<2\% \text{ yr}^{-1}$) are consistent with positive OH trend over East and Southeast Asia (Fig. S6b).

And

“CMAM and HadGEM3-ES show significant increasing and decreasing OH trend over the Antarctic region, respectively, consistent with the significant changes found for stratospheric O₃ in these models (Fig. S6c).”

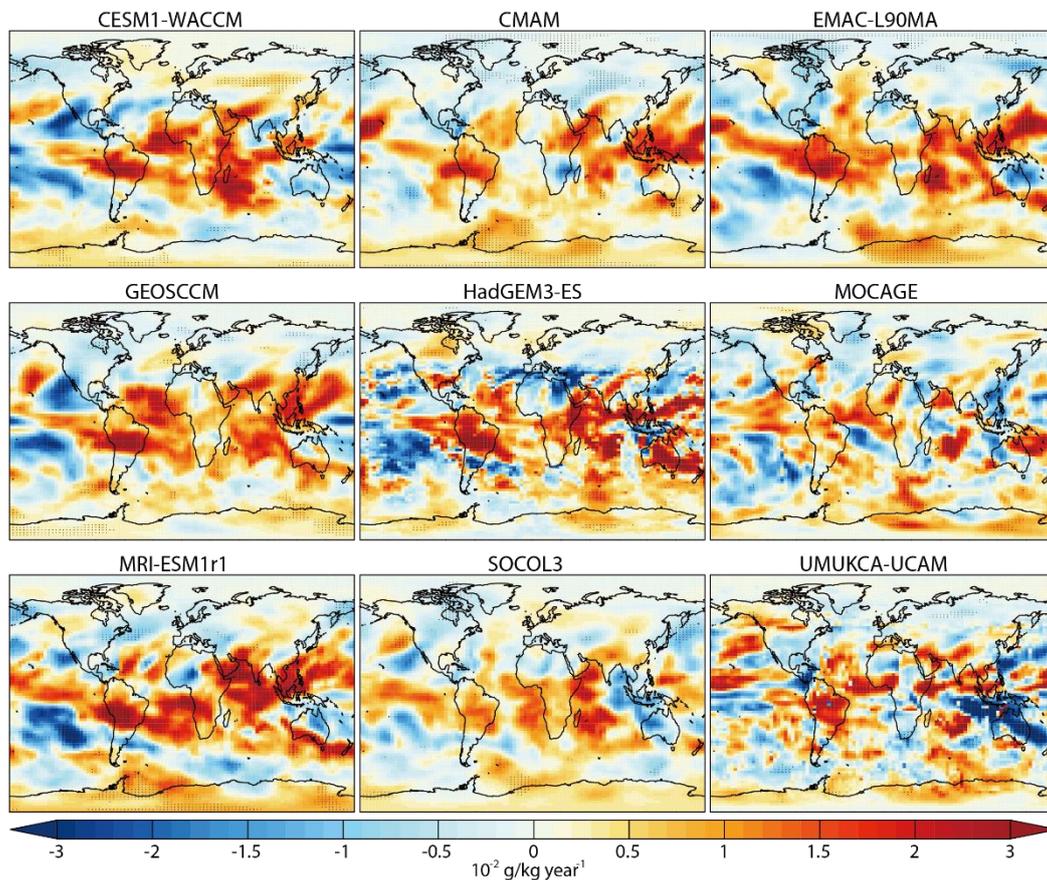


Figure S6a. Spatial distribution of tropospheric specific humidity trends from 2000 to 2010 (in 10^{-2} g/kg year $^{-1}$). Black dots denote model grid-cells with statistically significant trends (p -value < 0.05).

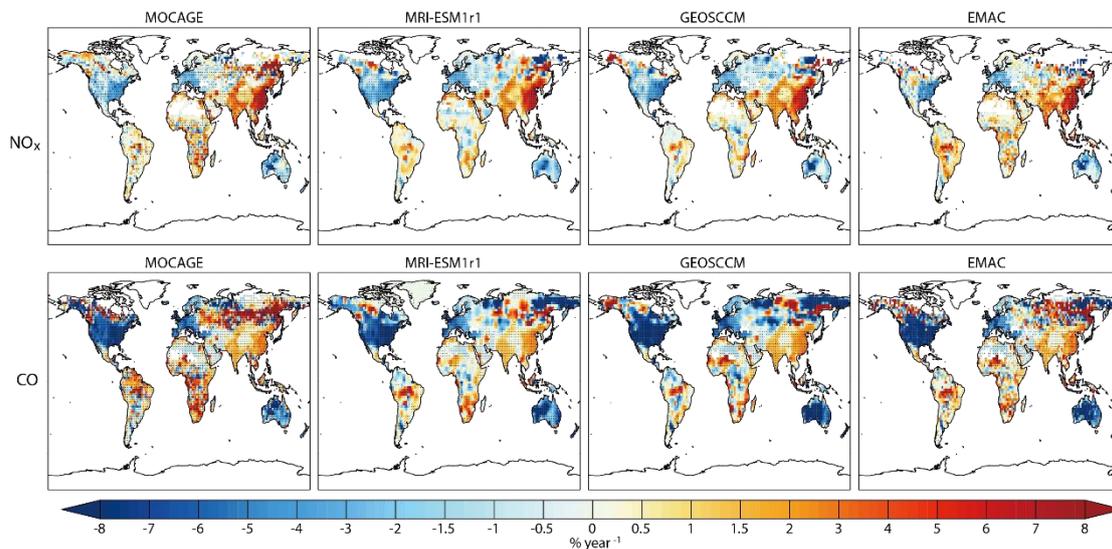


Figure S6b. Spatial distribution of NO $_x$ (top panels) and CO (bottom panels) trend from 2000 to 2010 (in %). Black dots denote model grid-cells with statistically significant trends (p -value < 0.05).

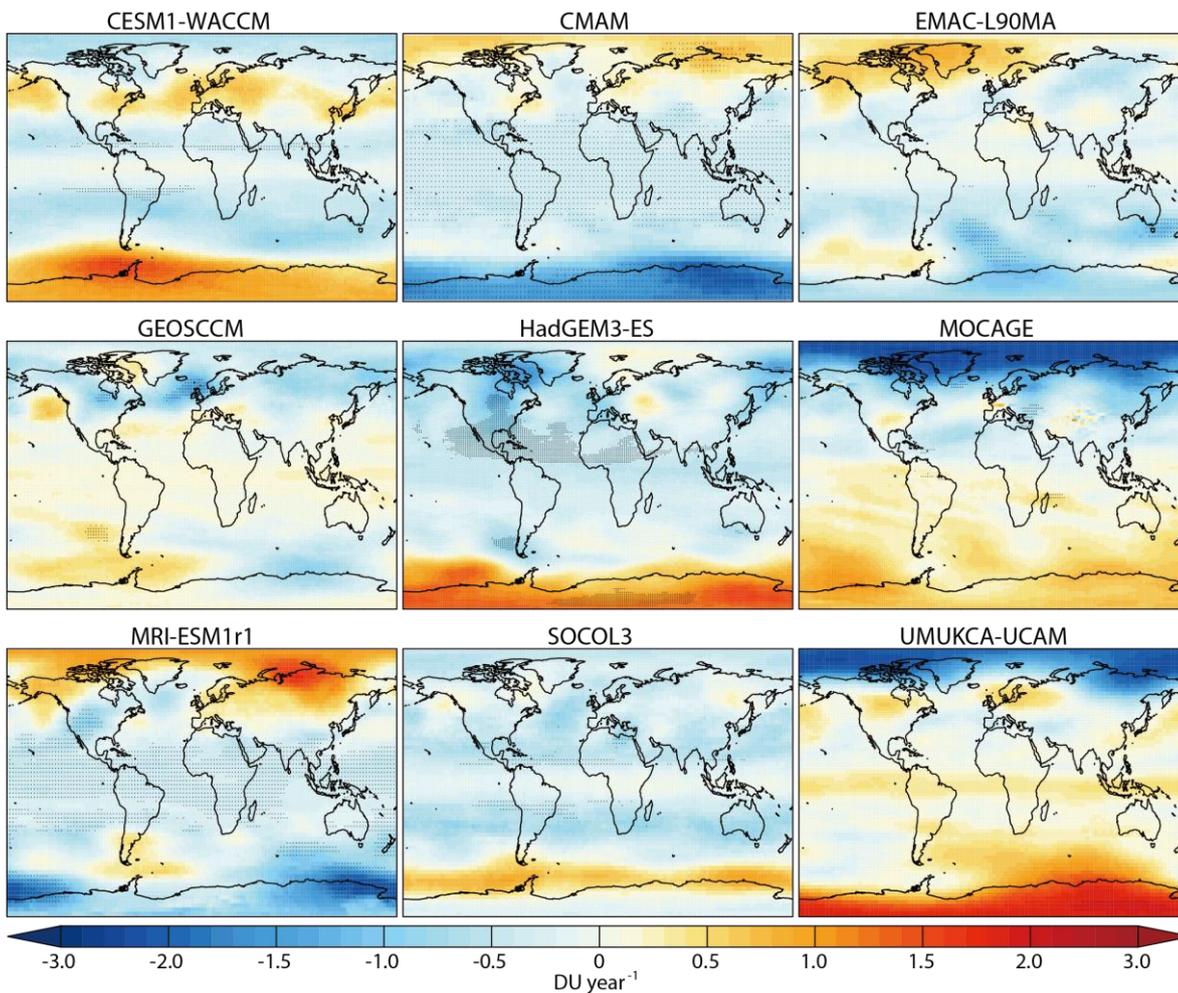


Figure S6c. Spatial distribution of stratosphere O₃ column trends from 2000 to 2010 (in DU year⁻¹). Black dots denote model grid-cells with statistically significant trends (p-value < 0.05).

Comments: L459-460: Changes in stratospheric ozone also dominate in the southern hemisphere and could potentially add to inter-model differences.

Response: see previous answer : "CMAM and HadGEM3-ES show significant increasing and decreasing OH trend over the Antarctic region, respectively, consistent with the significant changes found for stratospheric O₃ in these models (Fig. S6c)."

Comments: L478-485: The sensitivity of CH₄ oxidation due to OH to lower tropical tropospheric temperature has been established by prior studies (see John et al., 2012 and references therein), some of which should be cited here. Additionally, the discussion here will benefit from a table of CH₄ loss flux simulated by LMDz using the different OH fields for tropics (30S-30N), northern mid to high lat (30N-90N), and southern mid to high (30S-90S) and for three vertical levels. Or these numbers could be plotted in the form of Lawrence plots as in Lawrence et al. (2001) for each OH field. This would clearly show

the diversity in the spatial distribution of methane loss resulting from the different OH fields.

Response: We have added table S6 in the supplement:

Table S6. CH₄ loss by OH oxidation (unit: Tg yr⁻¹) as simulated by LMDz using different OH fields and repeating year 2000 over 30 times.

Run name		TransCom	INVSAT	INCA	CESM1-WACCM	CMAM	EMAC-L90MA	GEOSCCM	MOCAGE	MRI-ESM1r1	SOCOL3
Surface-750hPa	30-90 N	42.9	58	53.1	56	56	51	50.2	70.2	54.2	79.7
	0-30 N	90.5	106.5	105.1	101.9	115	106.2	93.8	123.7	111.6	112.9
	0-30 S	77.9	85.1	83.5	74.6	89.9	79.6	75.2	91.7	85.4	77.4
	30-90 S	16.7	16.3	18.6	18.5	18.2	18.1	17.7	24.3	16.5	20.7
750-500hPa	30-90 N	25.8	25.9	25.7	28	22.9	26.8	26.4	26.1	22.6	31.1
	0-30 N	66.8	56.4	57.5	59.5	49.5	57.9	63.5	51.1	54.4	49.1
	0-30 S	61	45.6	46.3	45	38.9	44.4	49.9	34.6	42.8	35.5
	30-90 S	15.1	10.6	12.2	11.2	9.6	13	11.7	9.2	9	10.5
500-250hPa	30-90 N	9.9	11.5	11.9	13.8	11.9	12.3	12.6	8.8	11.2	12.8
	0-30 N	26.1	21.9	23.3	27.4	26.4	25.7	31	16.7	26.9	19.7
	0-30 S	24.7	17.7	19.2	20.7	21.5	20.2	23.6	11.5	20.8	14.4
	30-90 S	7	5.6	6.2	5.8	5.4	6.1	6	3	4.8	4.5

We also add in the text:” Previous studies have demonstrated that the sensitivity of CH₄ oxidation to lower tropical temperature (Spivakovsky et al., 2000; John et al., 2012), and our simulations show that 36%-46% of CH₄ is oxidized over lower tropical region (surface-750hPa, 30 S-30 N) (Table S6).”

Comments: L497-499: Is the scaling applied to every year over the 2000-2010 period or just for year 2000?

Response: we clarify by adding in the text:” The single global scaling factor (per OH field) for the year 2000 is applied to every year between 2000 and 2010.”

Comments: L510-513: I am not sure if it is surprising that there was a spread in the simulated methane distributions across models, particularly, because the scaling was performed on a global scale (matching the global methane loss flux) rather than at the grid-cell level. While the global OH may match with INCA NMHC OH field after scaling but the spatial distribution may still be different producing differences in the simulated global mean methane distributions.

Response: The global scaling approach is what methane inverse modelers usually do. We have applied the same approach here, indeed aiming at assessing how the difference in OH spatial distribution can influence CH₄ spatial distributions.

L545: It should be EMAC-L90MA.

Response: Thank you for pointing out the typo, we change as suggested.

Comments: L544-547: Add “relative to the Run_fix_OH” after “....further reducing CH₄ mixing ratios by up to 20-30 ppbv in 2016...”

Response: Changed as suggested.

Comments: L565: Have the measurements been combined in a specific way to create global mean? Are the model CH₄ values sampled at the location of these stations?

Response: We clarify by adding in the text “The modeled surface CH₄ mixing ratios are sampled according to station locations.”

Comments: L569-571: Reference Figure 8 here.

Response: Changed as suggested.

Comments: L574-576: From Figure 8, it looks like that the three lines (black obs, blue Run_standard and grey Run_fix_OH) are overlapping until about 2003. y-axis of Figure 8 also needs concentration units (ppb?)

Response: We change “Indeed, neither Run_standard nor Run_fix_OH simulations do capture the stagnation before 2006” to “Indeed, neither Run_standard nor Run_fix_OH simulations do capture the stagnation during 2004- 2006”. And we add the unit to figure 8 as suggested.

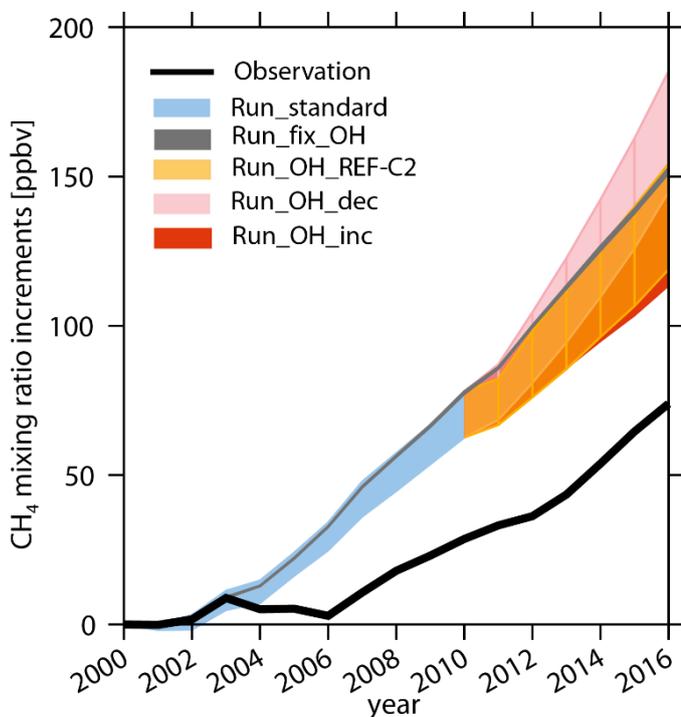


Figure 8. Time series of surface CH₄ mixing ratio increments compared to 2000 for NOAA observations (black line) and model ranges from all the LMDz experiments collected at observation sites (shades) and described in the text and in Table 2.

Comments: L576-578: I am not sure if I understand this sentence (especially “fill the gap between model simulations and observations by up to 50 %”). Could the authors please clarify and also how these percentages have been calculated.

Response: We clarify by adding in the text “We define highest CH₄ mixing ratios simulated by different OH as CH_{4-H}, lowest CH₄ mixing ratios as CH_{4-L}, and CH₄ simulated by Run_fix_OH as CH_{4-fix_OH}. Based on Run_fix_OH, on average over 2000-2016 and depending on the OH scenario, we found that [OH] changes can emphasize the model-observation mismatch by up to 19% (mean values of $(\text{CH}_{4-H} - \text{CH}_{4-\text{fix_OH}}) / (\text{CH}_{4-\text{fix_OH}} - \text{observed CH}_4)$ during 2000-2016), or limit the model-observation mismatch by up to 54% (mean values of $(\text{CH}_{4-\text{fix_OH}} - \text{CH}_{4-\text{fix_L}}) / (\text{CH}_{4-\text{fix_OH}} - \text{observed CH}_4)$ during 2000-2016) (figure 8).”

Comments: L609-610: This assertion needs to be substantiated or toned down in the absence of more detailed analysis (due to lack of diagnostics such as OH prod and loss). I believe the Riahi et al 2011 reference is not appropriate here as it documents RCP8.5 emissions but does not comment on chemistry-composition impacts from changes in these emissions.

Response: We have changed the text here based on the new analysis of emissions, water vapor, and ozone column trend now provided in the paper. “Such an increase in OH is mainly attributed to the significant positive OH trend over East and Southeast Asia ($>0.1 \times 10^5 \text{ molec cm}^{-3} \text{ yr}^{-1}$) in response to more OH production by NO_x than OH destruction by CO, and over tropical regions in response to increasing water vapor. ”