Reply to SC1: 'Short comment on Zhao et al. (2019)'

We thank the reviewer for the helpful comments provided. All of them have been addressed in the revised manuscript. Please see our itemized responses below.

Comment 1: For certain experiments in this study, the authors use a compilation of methane emissions that is based on bottom-up estimates (inventories and process-models) and not constrained by atmospheric observations (lines 263-266). The resulting increase in emissions between 2000 and 2016 is 70 Tg/yr. This is a large difference, compared to emission scenarios constrained by methane mole fractions [CH4], which place the increase in the order of 20-40 Tg/yr (depending on start and end period). For examples, see Saunois et al. (2017, doi.org/10.5194/acp-17-11135-2017) with best estimates of around 24 Tg/yr between the two periods 2002-2006 and 2008-2012 or Nisbet et al. (2019, doi.org/10.1029/2018GB006009), who estimate a ~44 Tg/yr difference between 2000-2005 and 2015-2018. Bottom up emissions have been repeatedly shown to overestimate the increase in methane after 2007, as reviewed by Saunois et al. (2017). Consequently, the increase of [CH4] in the atmosphere is strongly overestimated in the present study as seen in Fig. 8, where the modelled difference in [CH4] between 2000 and 2010 is >70 ppb, while the observation is ~25 ppb. The modelling presented here is outside my area of expertise, yet it would be interesting how the overestimate in methane emissions will influence the simulated CH4-OH dynamics.

Comment 3: Would a lower rate of emissions increase produce a significantly different result? The unrealistic CH4 evolution makes it difficult to assess the importance of the findings for the recent methane budget. For example, the authors state that varying OH from 2000 to 2010 suppressed [CH4] by 5-15 ppb (line 538). Would that value hold for a slower [CH4] increase? Does the stated OH effect as equivalent to 7-20% of the emissions change (line 540) represent a fixed percentage of any emissions increase or would it scale with the emissions scenario (in which case the OH effect could be equivalent to 16-45% of the emissions change of Saunois et al., 2017)? In my opinion, the relevance of the presented findings for the wider community could be strongly enhanced by a more realistic emission scenario.

Response for these two related comments: We performed two additional experiments with emissions fixed to 2000 to test the influence of emission scenarios on the results presented.

We have added in the text:

Section 2.2.2 (method)

"In addition, we conducted two simulations during 2000-2010 driven by emission inventories fixed to the year 2000 to test the influences of the emission bias on our results. The two simulations use OH fields simulated by CESM-WACCM, one with inter-annual variations of OH (Run_fix_emis) and the other one with OH field fixed to 2000 (Run_fix_emis_OH)."

Section 4.2.2 (results)

"To test whether the impacts of [OH] year-to-year variations on CH_4 mixing ratios depends on the chosen emission scenarios, we compare the above results with that calculated by an extreme scenario where model simulations are driven by fixed emissions (year 2000, Run_fix_emis and Run_fix_emis_OH, table 2). With emissions fixed to 2000, the CH_4 mixing ratio increased by 2ppbv from 2000 to 2010, and increasing OH (CESM-WACCM OH fields) can reduce CH_4 mixing ratio by 13.5ppb in 2010, comparable to 13.9 ppb calculated by Run_std and Run_fix_OH with CESM_WACCM OH fields. The results indicate only small effect of emission scenario choice on the absolute changes in CH_4 mixing ratios due to OH variations. However, our choices have a large effect on the relative changes to the total modeled CH_4 increase. Indeed, if we use the emission scenarios that match observations (~+25ppbv of CH_4 mixing ratio increase from 2000-2010 instead of ~70 ppb here, Ed Dlugokencky, NOAA/ESRL, 2019), the CH_4 mixing ratio changes due to OH can contribute to more than half (13.5-13.9ppbv versus 25ppbv) of the changes driven by emissions."

We also included two model experiments in Table 2.

Table 2. List of LMDz experiments and model setups.

	Simulation period	Inter annual variability in [OH]	Inter annual variability in CH4 emissions
Run_standard	2000-2010	2000-2010	2000-2010
Run_REF-C2	2011-2016	2010 apply inter-annual variability from REF-C2	2011-2016
Run_OH_inc	2011-2016	2010 apply annual growth rate of 1‰	2011-2016
Run_OH_dec	2011-2016	2010 apply annual decrease rate of 1%	2011-2016
Run_fix_OH	2000-2016	Constant OH (year 2000)	2010-2016
Run_fix_emis	2000-2010	2000-2010(CESM-WACCM only)	Constant (2000)
Run_fix_emis_oh	2000-2010	Constant OH (year 2000 CESM-WACCM only)	Constant (2000)

Comment 2: E.g., is the offline LMDz model subject to CH4-feedback on OH?

Response: We clarify in the text:" Chemical sinks of CH_4 are calculated using prescribed three-dimensional OH and $O(^1D)$ fields, and variation in CH_4 cannot feedback on OH."

Reply to RC1: 'Review of Zhao et al. (2019)'

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 NOx and stratospheric ozone could be investigated. The authors also do not evaluate the models against proxy observational constraints (such as ozone, water vapor, CO, NOx 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 NOx 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 stretghten a bit model evaluation, we now compare ozone simulated by the CCMI models with TOMS/SBUV observations (Fig. S4) and we have calculated tropospheric chemical lifetime in table 4. In addition, the CO column and tropospheric O_3 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 CH_4 lifetime of the models that provided CH_4 chemical loss data are 8.7 ±1.1 yr. Both the multi-model mean and the (large) range of [OH] as well as tropospheric CH_4 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 O_3 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 CCMI 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 CCMI REF-C1 experiments.

We compare spatial and vertical distributions of OH fields from REF-C1 (main text) with that from REF-C1SD to access influences from dynamic biases. Of CCMI 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 molec.cm^{-3}) averaged over the years 2000 to 2010 from CCMI 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})	(10^5 molec.cm^{-3})	(10^5 molec.cm^{-3})	(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 10^5 molec cm^{-3}) over the years 2000 to 2010 from CCMI 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.

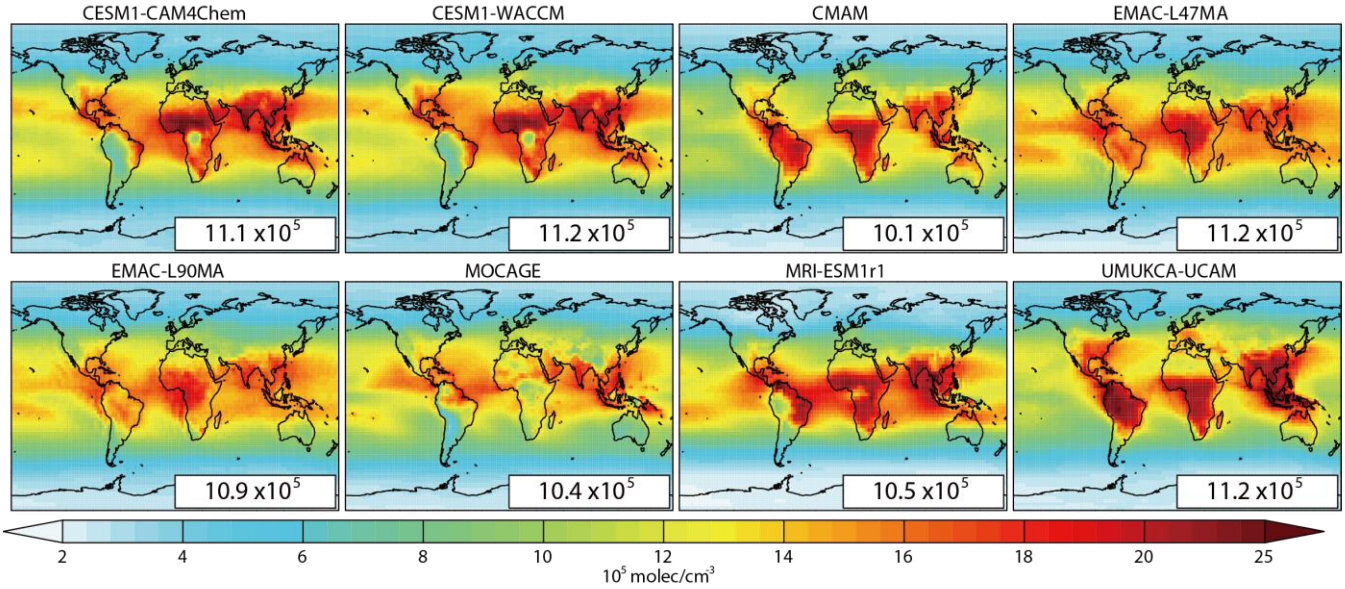

Figure S1. The spatial distributions of volume-weighted tropospheric mean OH fields CCMI REF-C1SD experiments averaged for 2000-2010 . Global mean values (10^5 molec cm^{-3}) are shown as insets.

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 MACCity emissions (therefore REF-C1 simulation) follow the RCP8.5 scenario. It would also be helpful to clarify whether biomass burning emissions are also from MACCity 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 MACCity 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 CH_4 variations since 2000, and to relate this contribution to characteristics of the different OH fields." to "We then conducted an ensemble of CH_4 simulations with different OH fields using the LMDz chemistry-tranport model to estimate a range for the contribution of changes in [OH] to the atmospheric [CH_4] 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(^{1}D) 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 NOx 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:

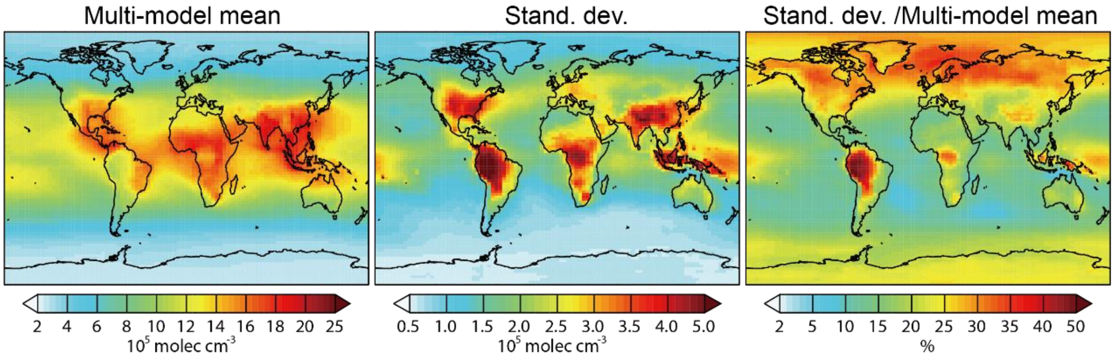

Figure S2. Multi-model mean (left), standard deviation(middle). and standard deviation relative to multi-model mean of tropospheric mean OH fields shown in figure 2.

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 ×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 NOx 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 NOx 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 NOx 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(^{1}D) 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

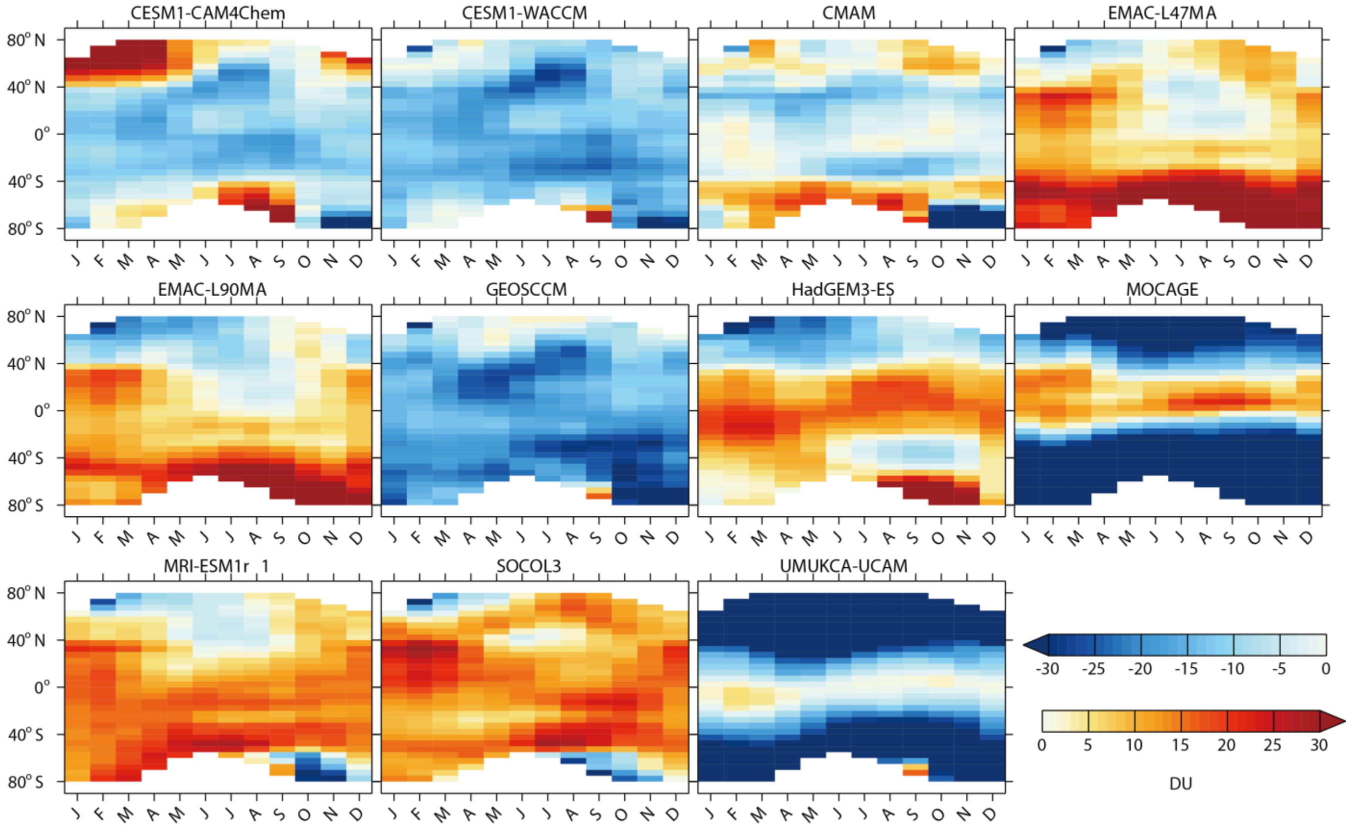

Figure S4. Monthly total column ozone bias from CCMI simulations averaged over 2000-2010 compared to satellite measurements from Total Ozone Mapping Spectrometer/solar backscatter ultraviolet (TOMS/SBUV) (model minus measurement).

We also add in the main text:

”The stratospheric ozone can contribute to inter-model OH discrepancies through influencing O(^{1}D) photolysis rates. However, we find that models that simulated lower stratosphere and total ozone column are not corresponding to higher O(^{1}D) 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(^{1}D) 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 NOx 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 NOx 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.

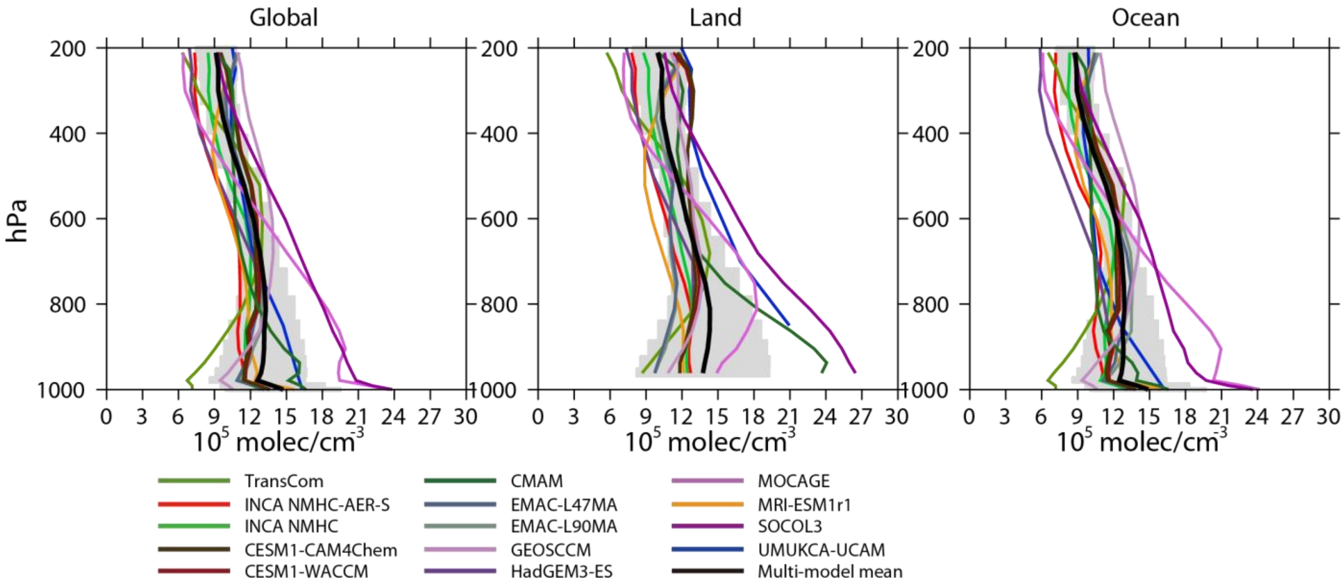

Figure 3. Vertical distributions of [OH] averaged over the globe (left), land (middle) and ocean (right) for 2000-2010. Color lines represent [OH] from individual model simulations, black lines represent multi-model mean values and grey shades represent the standard deviations.

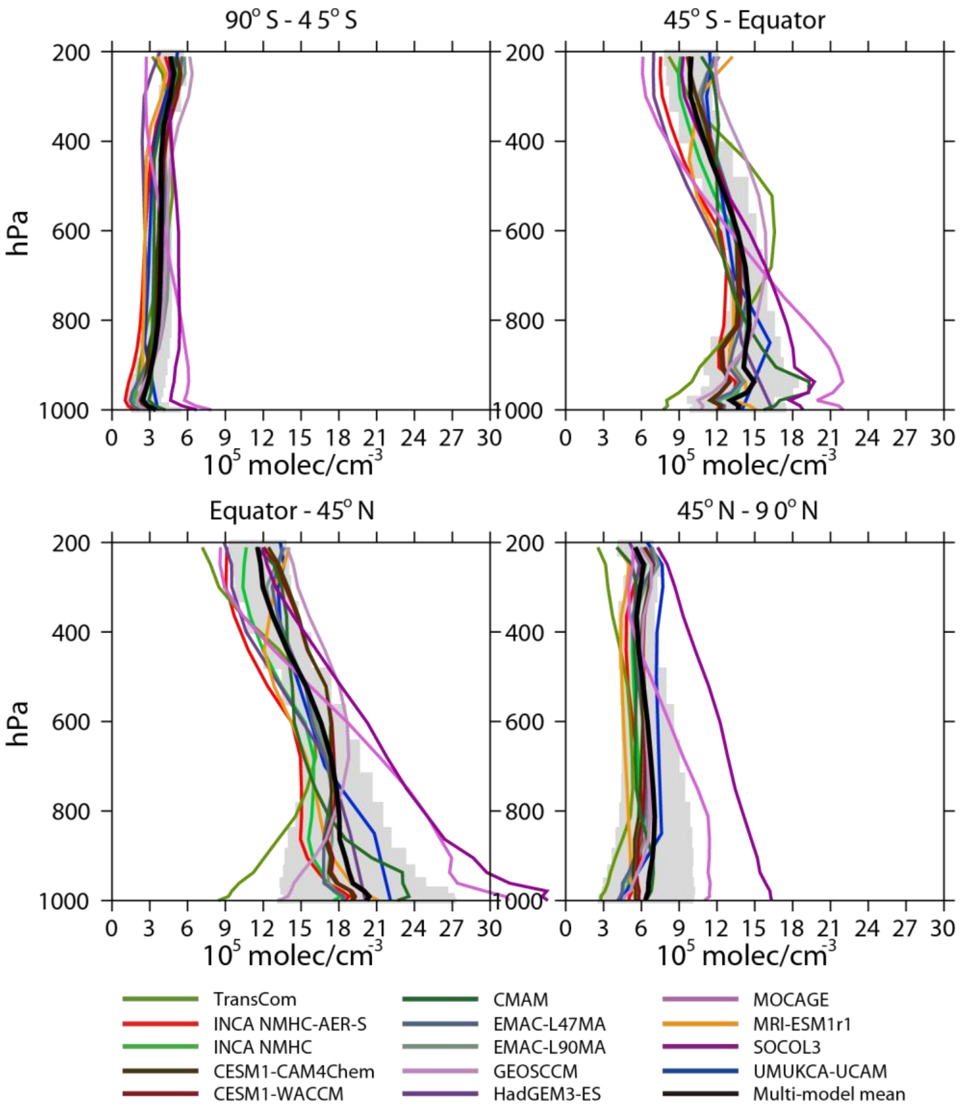

Figure S3. Vertical distribution of [OH] averaged over four latitude bands and over the years 2000 to 2010. Color lines represent [OH] from individual model simulations, black lines represent multi-model mean values and grey shades represent the standard deviations.

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 ±1.2%, remaining within ±0.5 ×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 (~-0.3 ×10^5 molec cm^{-3}; -3.4%); and increase in SOCOL3 (~+0.6 ×10^5 molec cm^{-3}; +4.5%), UMUKCA-UCAM (~+0.5 ×10^5 molec cm^{-3}; +4.8%), and MOCAGE (~+0.5 ×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 CH_4 (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 ±1.0% from 1980 to 2000 (Bousquet et al., 2005), and 2.3 ±1.5% from 1998 to

2007(Montzka et al., 2011), as compared to 2.1 ±0.8% and 1.0 ±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 ±2.2% from 1980 to 2000 (Naik et al., 2013). Here, for the same period the increase of CCMI [OH] is 4.6 ±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 CH_4 growth during the 2000s, as opposed to the decreasing [OH] from mid-2000s calculated by Rigby et al. (2017) and Turner et al (2107) 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 NOx emissions in each grid cell for the CCMI models to access 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 O_3 (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% yr^{-1}) than CO (<2% 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 O_3 in these models (Fig. S6c)."

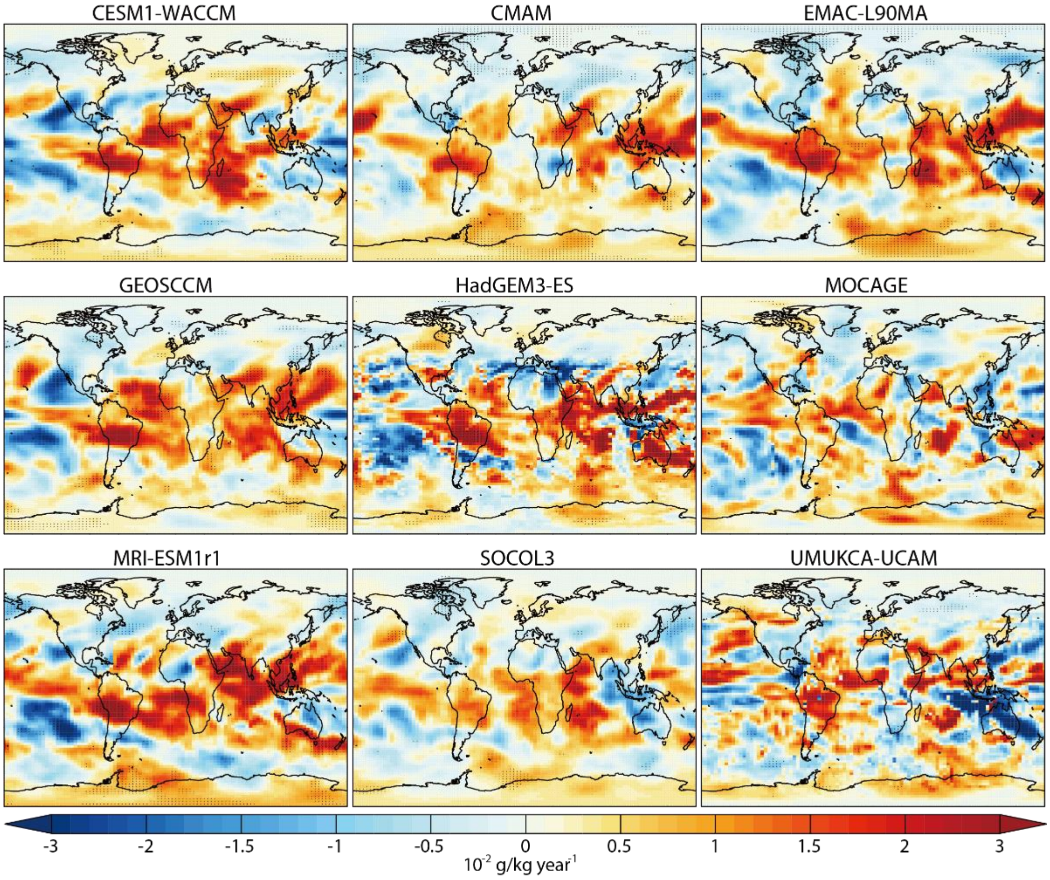

Figure S6a. Spatial distribution of tropospheric specific humidity trends from 2000 to 2010 (in 10^{-2} g/kg year^{-1}). Black dots denote model grid-cells with statistically significant trends (p-value < 0.05).

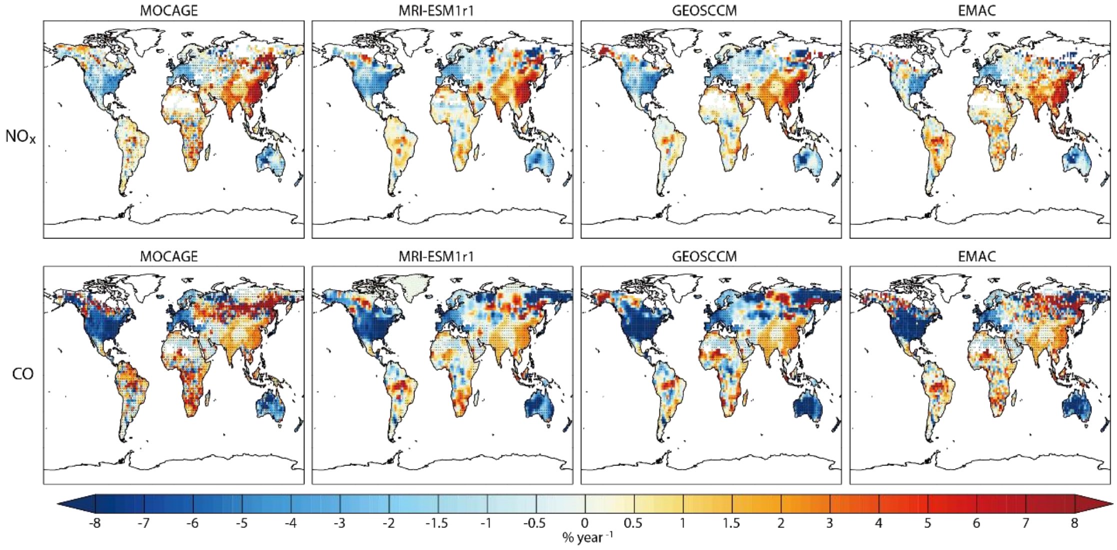

Figure S6b. Spatial distribution of NO_x (top panels) and CO (bottom panels) trend from 2000 to 2010 (in %). Black dots denote model grid-cells with statistically significant trends (p-value < 0.05).

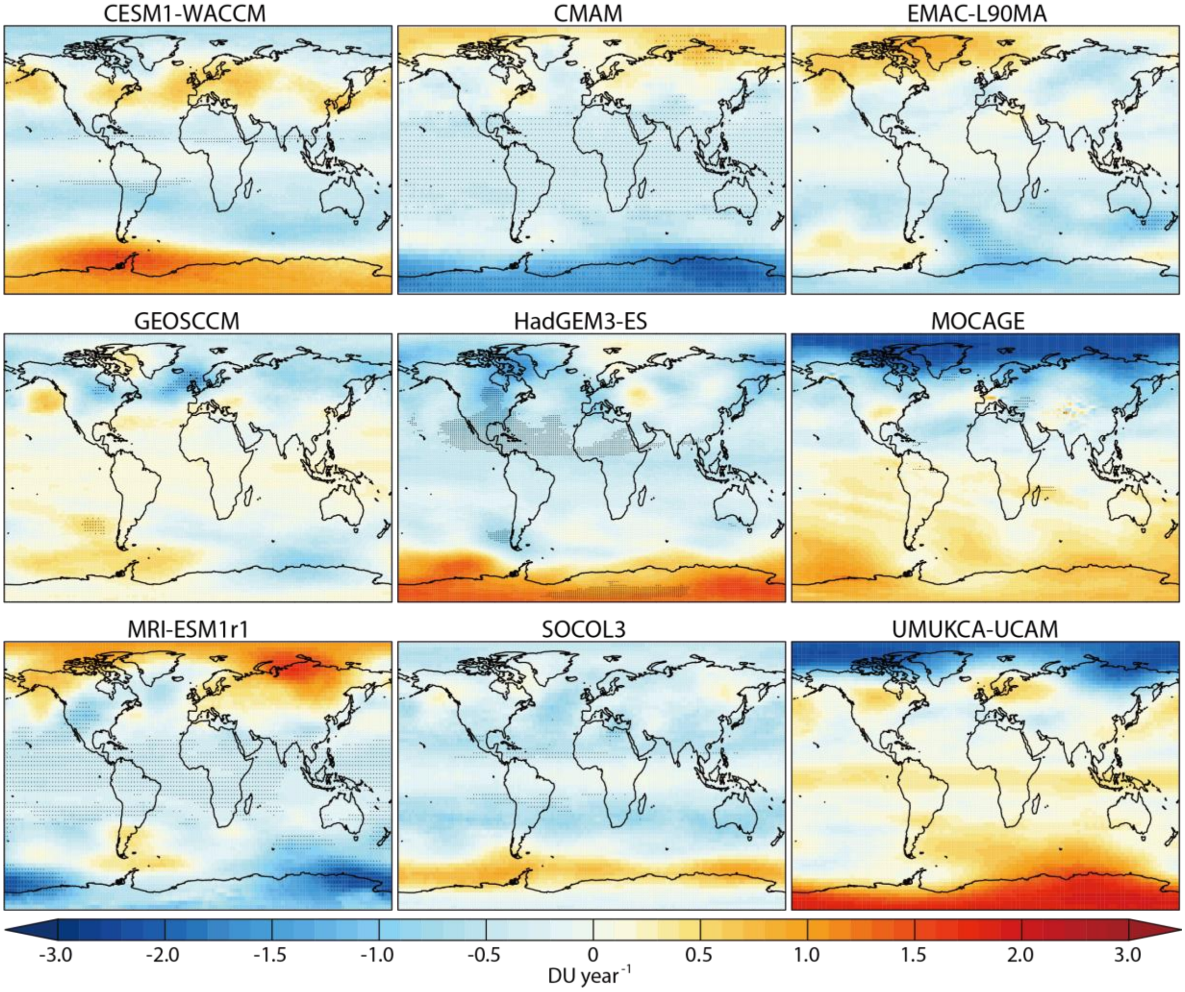

Figure S6c. Spatial distribution of stratosphere O_3 column trends from 2000 to 2010 (in DU year^{-1}). Black dots denote model grid-cells with statistically significant trends (p-value < 0.05).

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 O_3 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. CH_4 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 CH_4 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 CH_4 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 CH_4 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.

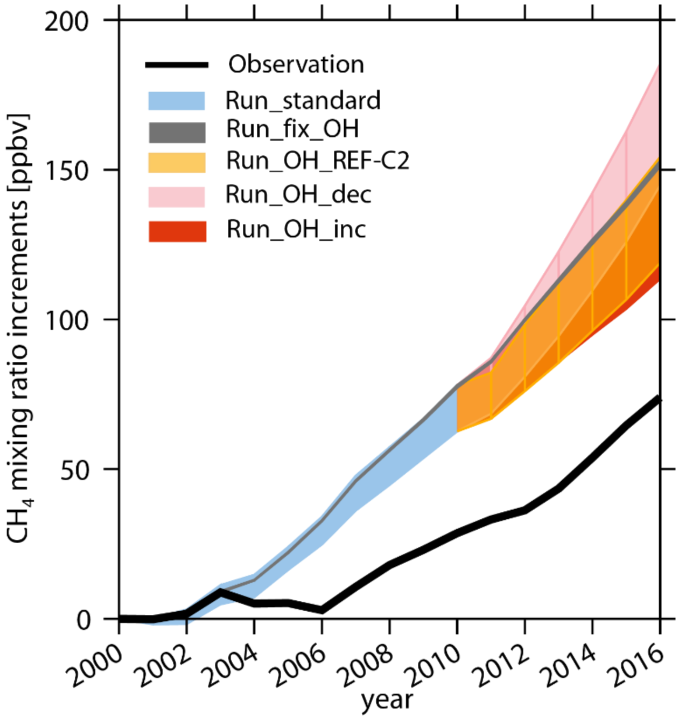

Figure 8. Time series of surface CH_4 mixing ratio increments compared to 2000 for NOAA observations (black line) and model ranges from all the LMDz experiments collected at observation sites (shades) and described in the text and in Table 2.

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 CH_4 mixing ratios simulated by different OH as $CH_{4\text{-H}}$, lowest CH4 mixing ratios as $CH_{4\text{-L}}$, and CH_4 simulated by Run_fix_OH as $CH_{4\text{-fix\_OH}}$. 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 ($CH_{4\text{-H}}$ — $CH_{4\text{-fix\_OH}}$)/($CH_{4\text{-fix\_OH}}$- observed CH_4) during 2000-2016), or limit the model-observation mismatch by up to 54% (mean values of ($CH_{4\text{-fix\_OH}}$ — $CH_{4\text{-fix\_L}}$)/($CH_{4\text{-fix\_OH}}$- observed $CH_{4\text{-}}$) 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 (>0.1×10^5molec cm^{-3} 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. "

Reply to RC2: 'Review comments on acp-2019-281'

General comments: The manuscript "Inter-model comparison of global hydroxyl radical (OH) distribution and their impact on atmospheric methane over the 2000-2016 period" written by Yuanhong Zhao describes the inter-model differences in spatial distribution and temporal evolution of OH concentrations, and elucidates the impacts of simulated OH concentration fields on CH4 using the LMDz chemical transport model. The manuscript contains novel investigation to reveal inter-annual variations in OH and its impact on CH4 over recent decades using multi-model approach. The topic of the manuscript is certainly within the scope of ACP. Overall, the manuscript is well written and easy to follow. I would like to consider the publication of the manuscript from ACP, while I have several comments below which should be addressed before publication.

Response:

We thank the reviewer for the helpful comments. All of them have been addressed in the revised manuscript. Please see our itemized responses below.

Specific comments:

Comments: OH field Is the prescribed biogenic NMVOC emissions (p. 8, l. 187) climatology? Please clarify.

We mean here that some models just prescribed a fix scenario for NMVOC emissions and do not accoiunt for time variability.

Text has been clarified : "Biogenic NMVOC emissions in CESM and GEOSCCM are calculated based on the distribution of plant functional types and meteorology conditions with MEGAN, whereas the other models prescribe climatological biogenic NMVOC emissions."

Comments: How did the authors prescribe the ECLIPSE and RCP85 emission inventories in the INCA simulations during the periods before 2004, between 2006-

2009, and after 2011?

Response: We clarify by changing this sentence to" Anthropogenic emissions from Short-Lived Pollutants (ECLIPSE) inventory (Stohl et al., 2015) for 2005 and RCP 85 emission inventory (Riahi et al., 2011)) for 2010 are applied to every year of INCA NMHC-AER-S and INCA NMHC simulations, respectively."

Comments: 2.2.2. Model simulations Please clarify how the OH increasing and decreasing rates are determined in the Run_OH_inc and Run_OH_dec simulations. Why are the rates +1 and −1.

Response: We clarify by add in the text : " In order to assess the recent change in [OH], we tested two additional scenarios between 2010 and 2016: one with [OH] increase of +0.1% yr^{-1} (Run_OH_inc) according to the slightly changing of OH calculated by ACCMIP models and one with [OH] decrease of -1% yr^{-1} (Run_OH_dec) according to obviously decreasing of OH calculated by top-down approaches constrained by observations."

Comments: 3.1. Spatial distributions of tropospheric OH The authors attributed possible causes of too large interhemispheric differences in OH in the CCMI models to model O3 and CO biases and unaccounted processes in some of the CCMI models, as reported by previous studies. Why is not the model performance on O3 and CO in the CCMI ensembles evaluated or referred? It might be better to cite Strode et al. (2016), Revell et al. (2018), and other papers.

Response: 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. We have added in the text:

"Previous studies have attributed the inconsistency between the simulated and the observed OH N/S ratios to a model overestimation of O_3 and underestimation of

CO over the Northern Hemisphere (Naik et al., 2013; Young et al., 2013; Strode et al., 2015), which have also been reported for CCMI models (Strode et al., 2016; Revell et al., 2018), …”

Add references:” Strode SA et al. (2016) Interpreting space-based trends in carbon monoxide with multiple models Atmos Chem Phys 16:7285-7294 doi:10.5194/acp-16-7285-2016”

We have increased the depth of the analysis of the root causes possibly explaining what we find in the paper, all along with the text (see answers to reviewer 1).

Comments: 3.3. Factors contributing to inter-model differences Why the authors did not assess inter-model differences in tropospheric O3 burden? The tropospheric O3 burden should also affect primary production of OH.

Response: We have calculated global mean O3 mixing ratios averaged over the tropospheric and their pressure altitude levels in table 5 and move the values of O1(D) and reactive humidity, which contribute less to the inter-model difference of [OH] to the supplement (Table S4).

	CO ppbv				**NO pptv**				**O3 ppbv**			
	750	500	250	Tp	750	500	250	Tp	750	500	250	Tp
CESM1-CAM4Chem	76	71	70	71	9	4	12	13	32	42	57	48
CESM1-WACCM	75	70	69	70	9	5	12	12	31	41	55	47
CMAM	77	68	64	69	17	4	17	26	34	43	60	52
EMAC-L47MA	85	77	70	75	8	4	11	14	38	48	63	56
EMAC-L90MA	84	76	69	74	8	5	11	17	38	48	61	54
GEOSCCM	78	74	73	74	9	5	13	13	33	43	61	49
MOCAGE	67	68	67	67	26	14	17	20	37	42	46	43
MRI-ESM1r1	93	86	83	86	10	5	20	32	36	48	67	56
SOCOL3	79	73	74	74	48	10	14	25	43	54	67	61
Mean ± stand. dev.	79±7	74±6	71±5	73±5	16±13	6±3	14±3	19±7	36±4	45±5	60±7	52±6

And we have added in the text:

” To analyze inter-model differences in OH vertical distributions, we compared CO, NO, and O$_3$ mixing ratios in table 5 as well as O(^{1}D) photolysis rates and

specific humidity in Table S4.

"Tropospheric O_3 can also influence primary production of OH, and tropospheric O_3 burden reflects combined effects of NO_x, CO, and VOCs. The high O_3 over the lower troposphere simulated by SOCOL3 and the low O_3 over the upper troposphere simulated by MOCAGE can contribute to explain the high and low [OH] simulated the two models over the corresponding altitudes, respectively. "

Comments: Do inter-model differences in vertical distribution of lighting NO production affect OH vertical distributions?

Response:

Yes indeed. We have added table S3 in the supplement:

Table S3. Lighting NOx 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 have also added in the text:"

L431-L435: "Lighting NO_x emissions, 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 NOx 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 NOx emissions range from 3.7-10.2 Tg yr^{-1}(table S3)

Comments:" 3.4. Inter-annual variations of OH What is possible cause of significant positive [OH] trends over the tropics (p. 19, l. 454)?**"**

Response: We add in the text:" By comparing spatial distribution of OH trend with specific humidity (Fig.S6a), NO$_x$ and CO emissions (Fig. S6b), and stratospheric O$_3$ (Fig.S6c), we find that positive OH trend over tropical regions are mainly corresponding to increases in water vapor (Fig. S6a)"

And we add figure S6a in the supplement:

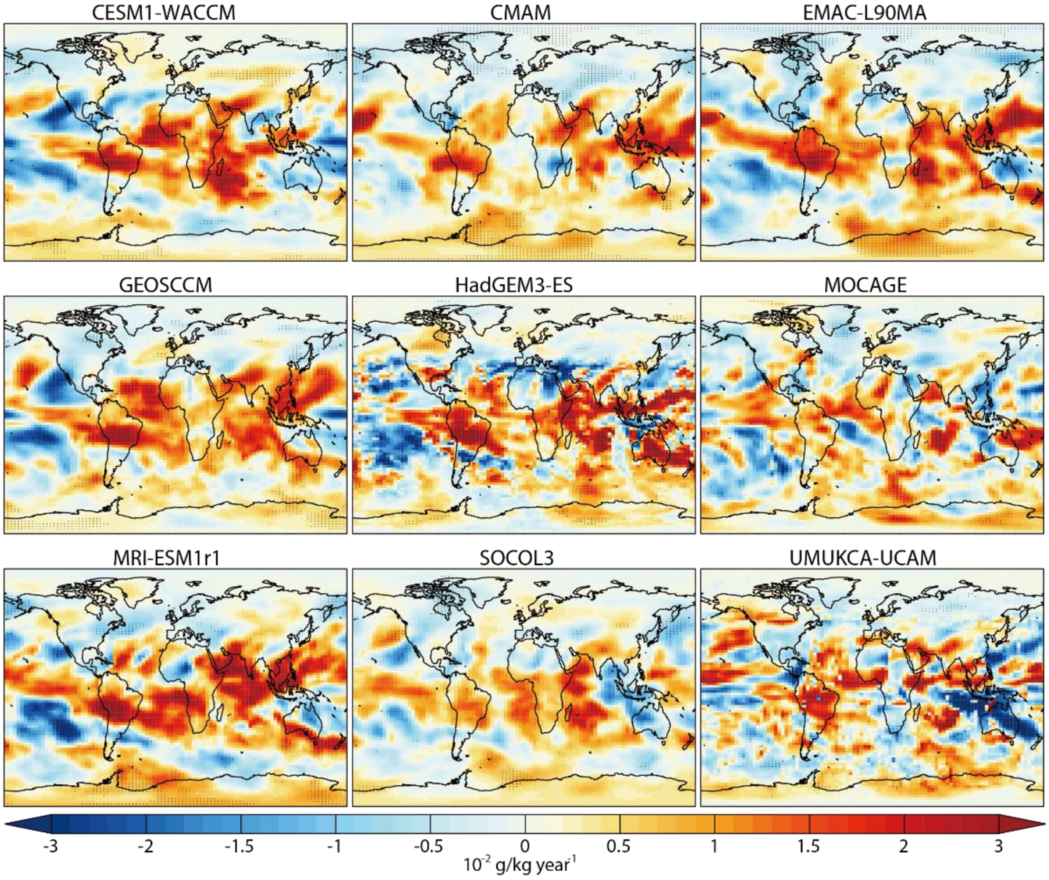

Figure S6a. Spatial distribution of tropospheric specific humidity trends from 2000 to 2010 (in 10^{-2} g/kg year^{-1}). Black dots denote model grid-cells with statistically significant trends (p-value < 0.05).

Comments: 4.2.1. Spatial distributions of tropospheric CH$_4$ mixing ratio Could you explain how inter-model differences in spatial and temporal OH variations affect the simulated global CH4 mixing ratio more in depth?

Response: In our paper, we attribute differences in LMDz simulated global mean CH4 mixing ratio to different global OH mean value and trend, and the spatial distribution of CH4 to multi-model spread in OH spatial and temporal distributions. To clarify our point, we have re-organized the first paragraph of section 4.2.1:

” We used the scaled OH fields to perform simulations between 2000 and 2010. Figure 6 shows the spatial distribution of tropospheric CH_4 mixing ratios for the simulation Run_standard (Table. 2, driven by OH with inter-annual variations) averaged over 2000-2010. Although all simulations started from the same initial conditions and OH fields were scaled to give the same global CH_4 loss as INCA NMHC in 2000, LMDz simulations using the different scaled OH fields still generated a spread of tropospheric mean (8 ppbv) and spatial distribution in CH_4 mixing ratios averaged during 2000-2010. Differences between the global tropospheric mean [OH] cannot explain these differences (see Table 4). Clearly, the different spatial (horizontal and vertical) and temporal variations of the OH fields (as described in Sect. 3), which were kept in this experiment by only scaling [OH] globally , significantly modify the simulated CH_4 mixing ratios (Table 7 and Fig. 6). OH fields with increasing trend will lead to lower LMDz simulated CH_4 mixing ratios. The LMDz simulation using the TransCom OH fields (without inter-annual variability) shows the highest CH_4 mixing ratios (1735 ppbv), while the one using the CMAM OH (with slightly increasing OH trend during the decade) shows the lowest CH_4 mixing ratios (1727 ppbv). ”

And we add in the second paragraph:” The differences in spatial distribution of OH fields can influence LMDz simulated CH_4 spatial distributions.”

Comments: Technical corrections: p. 13, l. 317: publication year is missing.

Response: We add the publication year, thank you very much for pointing out.

Inter-model comparison of global hydroxyl radical (OH) distributions and their impact on atmospheric methane over the 2000-2016 period

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Abstract

The modeling study presented here aims to estimate how uncertainties in global hydroxyl radical (OH) distributions, variability, and trends may contribute to resolve discrepancies between simulated and observed methane (CH_4) changes since 2000. A multi-model ensemble of 14 OH fields were analysed and were aggregated into 64 scenarios to force the offline atmospheric chemistry transport model LMDz with a standard CH_4 emission scenario over the period 2000-2016. The multi-model simulated global volume-weighted tropospheric mean OH concentration([OH]) averaged over 2000-2010 ranges between 8.7×10^5 and 12.8×10^5 molec cm^{-3}. The inter-model differences in tropospheric OH burden and vertical distributions are mainly determined by the differences in the nitrogen oxide (NO) distributions, while the spatial discrepancies between OH fields are mostly due to differences in natural emissions and VOC chemistry. From 2000 to 2010, most simulated OH fields show an increase of $0.1\text{-}0.3\times10^5$ molec cm^{-3} in the tropospheric mean [OH], with year-to-year variations much smaller than during the historical period 1960-2000. Once ingested into the LMDz model, these OH changes translated into a 5 to 15 ppbv reduction in CH_4 mixing ratio in 2010, which represent 7%-20% of the model simulated CH_4 increase due to surface emissions. Between 2010 and 2016, the ensemble of simulations showed that OH changes could lead to a CH_4 mixing ratio uncertainty of $>\pm30$ ppbv. Over the full 2000-2016 time period, using a common state-of-the-art but non-optimized emission scenario, the impact of [OH] changes tested here can explain up to 54% of the gap between model simulations and observations. This result emphasizes the importance of better representing OH abundance and variations in CH_4 forward simulations and emission optimizations performed by atmospheric inversions.

1 Introduction

The hydroxyl radical (OH) is the main oxidizing agent in the troposphere (Levy, 1971). OH is produced by the reaction of water vapor with excited oxygen atoms ($O(^1D)$), produced by ozone (O_3) photolysis (λ <340nm). In the troposphere, OH is rapidly removed by reactions with carbon monoxide (CO), methane (CH_4) and non-methane volatile organic compounds (NMVOCs) to generate hydroperoxyl radical (HO_2) or organic peroxy radicals (RO_2), resulting in a short lifetime of a few seconds (Logan et al., 1981; Lelieveld et al., 2004). HO_2 and RO_2 can further react with nitrogen oxide (NO) to regenerate OH (Crutzen, 1973; Zimmerman et al., 1987). At high latitudes, such a secondary production plays an important role, because the OH primary production is limited by the supply of $O(^1D)$ and water vapor (Spivakovsky et al., 2000). The abundance of OH reflects the combined effects of atmospheric composition (tropospheric O_3, and NO, CO, CH_4, and NMVOCs) and of meteorological factors such as humidity, UV radiation, and temperature.

Due to its short lifetime, global [OH] is difficult to estimate from direct measurements. Current understanding on global [OH] has been obtained either from inversion of 1-1-1trichloroethane (methyl chloroform, MCF) (Prinn et al., 2005; Bousquet et al., 2005; Montzka et al., 2011; Rigby et al., 2017; Turner et al., 2017), or using atmospheric chemistry models (Naik et al., 2013; Voulgarakis et al., 2013, Lelieveld et al., 2016). The former approach relies on the fact that OH is the main sink of MCF and on the hypotheses that emissions and concentrations of MCF are well known and well measured, respectively. The latter approach relies on chemistry-transport modeling with chemistry schemes of varying complexity. The global mass-weighted tropospheric mean [OH] in the 2000s calculated by atmospheric chemistry models was found to be about 11.5×10^5 molec cm^{-3}, with an inter-model dispersion of ±15% (Naik et al., 2013; Voulgarakis et al., 2013). Atmospheric chemistry models usually calculate higher [OH] over the Northern hemisphere than the Southern hemisphere (N/S ratio>1) (Naik et al., 2013) whereas MCF and ^{14}CO observations indicate a N/S ratio slightly smaller than 1 (Brenninkmeijer et al., 1992; Bousquet et al., 2005; Patra et al., 2014).

OH determines the lifetime of most pollutants and non-CO_2 greenhouse gases including CH_4, the second most important anthropogenic greenhouse gas after carbon dioxide (CO_2) (Ciais et al., 2013). About 90% of tropospheric CH_4 is removed by reacting with OH (Ehhalt et al., 1974; Kirschke et al., 2013; Saunois et al., 2016). The tropospheric CH_4 chemical lifetime against OH oxidation (global annual mean atmospheric CH_4 burden divided by annual CH_4 tropospheric loss by OH) calculated by the models that participated in the Atmospheric Chemistry and Climate Model Inter-comparison Project (ACCMIP) is 9.3 ±1.6 years, and the CH_4 total lifetime including all sink processes is 8.3 ±0.8 years (Naik et al., 2013; Voulgarakis et al., 2013), smaller than that of 9.1 ±0.9 years lifetime constrained by observations (Prather et al., 2012).

The tropospheric CH_4 burden has more than doubled compared to the pre-industrial era due to anthropogenic activities and climates change, resulting in about 0.62 W m^{-2} additional radiative forcing (Etminan et al., 2016). The global mean CH_4 growth rate decreased to near zero in the early 2000s but resumed increasing at ~5ppbv yr^{-1} since 2006 and reached more than 10 ppbv yr^{-1} in 2014 and in 2018 (Ed Dlugokencky, NOAA/ESRL, 2019). The growth rate of CH_4 is determined by the imbalance of its sources, primarily from anthropogenic activities (agriculture, waste, fossil fuel production and usage, and biomass burning) but also from natural emissions (mainly wetland and other inland waters), and sinks (OH oxidation, other chemical reactions with chlorine and oxygen radicals, and soil uptake). The precise reasons for the stagnation and renewed CH_4 growths still remains unclear (e.g. Rigby et al 2017; Saunois et al., 2017; Nisbet et al., 2019).

Several studies have linked such CH_4 variations to inter-annual variations and trend of OH. Based on MCF inversions, McNorton et al. (2016) concluded that an increase in [OH] significantly contributed to the stable atmospheric CH_4 before 2007; Rigby et al. (2008) found that a decrease of 4 ±14% in [OH] could partly explain the CH_4 growth between 2006 and 2007; Bousquet et al. (2011) found a smaller decrease in [OH] (<1% over the two years) and attributed the increase in CH_4 mostly to enhanced emissions over tropical regions; Montzka et al. (2011) also calculated a small inter-annual variation of 2.3 ±1.5% in [OH] during 1998 to 2007. More recently, based on multi-species box model inversions, Rigby et al. (2017) and Turner et al. (2017) inferred a decrease of 8 ±11% and 7% in [OH] during 2004-2014 and 2003-2016 respectively. Both of these studies suggested that such a decrease in [OH] is equivalent to an increase of more than 20 Tg yr^{-1} in CH_4 emissions, and therefore could significantly contribute to explain the post-2007 CH_4 atmospheric growth, although a solution with constant OH cannot be discarded. 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. The discrepancy between individual process-based models and MCF-proxy approaches, and the uncertainties, limit our ability to be conclusive on the role of [OH] changes to explain the CH_4 changes over the past decades.

To better understand OH distributions, trends, and influences on CH_4 since 2000, we have performed an inter-model comparison of 14 OH fields, including 11 derived from chemistry transport and chemistry-climate models that took part in the phase 1 of the Chemistry-Climate Model Initiative (CCMI) (Hegglin and Lamarque, 2015; Morgenstern et al., 2017), 2 from different configurations of the LSCE atmospheric chemistry transport model LMDz-INCA (Hauglustaine et al., 2004;Szopa et al., 2013), and 1 from the TRANSCOM 2011 inter-comparison exercise (Patra et al., 2011). 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 CH_4 mixing ratio changes since 2000, and to relate this contribution to spatio-temporal characteristics of the different OH fields. 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. In the following, our analysis first provides a brief description of the OH fields used in this study and the LMDz offline model (section 2). Section 3 compares the OH fields, analyses the factors contributing to inter-model differences and presents their inter-annual variability. Section 4 presents and discusses the impact of the different OH fields on the global CH_4 burden and growth rates simulated by LMDz. Section 5 summarizes the results and conclusion.

2 Method

2.1 OH fields

The CCMI project aims to conduct a detailed evaluation of atmospheric chemistry models in order to assess uncertainties in the models' projections of various climate-related topics such as tropospheric composition (Hegglin and Lamarque, 2015; Morgenstern et al., 2017). The CCMI OH fields used in our study are obtained from 10 different models and 3 CCMI reference experiments: REF-C1 (covering the time period 1960-2010), REF-C2 (covering 1960-2100), and REF-C1SD (1980-2010). The REF-C1 experiment is driven by state-of-the-art historical forcings and sea surface temperatures (sst) and sea ice concentrations (sic) based on observations, while the REF-C2 experiment is using either coupled ocean and sea ice modules or prescribes sst and sic obtained from another climate model. Since the REF-C1 experiment is supposed to be more realistic regarding sea surface conditions, our analysis focused on OH fields from the REF-C1 experiment before 2010 and only tested the influences of OH on CH_4 simulations after 2010 by applying the inter-annual variability from the REF-C2 experiment. 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). Detailed descriptions of CCMI simulations can be found in Morgenstern et al. (2017).

In this study, we used only the CCMI models that include detailed tropospheric ozone chemistry as listed in Table 1. Note that EMAC offers fields at two different model resolutions. The level of detail in chemical mechanism, in particular with respect to included NMVOCs, varies among the models. For example, CMAM does not include any NMVOC species, but added 250 Tg CO emissions to account for CO production from isoprene oxidation. UMUKCA-UCAM only include HCHO (formaldehyde) and SOCOL3 only include HCHO and C_5H_8 (isoprene). Other models include multiple primary NMVOC species and more complex VOC chemistry.

The anthropogenic emissions recommended for the two CCMI reference simulations are the MACCity inventory (Granier et al., 2011) for 1960-2000. After 2000, the REF-C1 experiment continued to use the MACCity inventory (which follow the RCP8.5 inventory after 2000), while the REF-C2 used the RCP6.0 inventory (Masui et al., 2011). The CMAM model did not follow this procedure and used the ACCMIP historical database of emissions (Lamarque et al., 2010) until 2000 followed by RCP8.5 emissions (Riahi et al., 2011) instead. 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. CCMI model simulations also include natural emissions from lightning, soil and biogenic sources. Lightning NO_x emissions are calculated based on meteorological data such as cloud top height (Price and Rind, 1994; Grewe et al., 2001) and updraft mass flux (Allen and Pickering, 2002). Soil NO_x emissions are calculated interactively in EMAC and GEOSCCM using the scheme described by Yienger and Levy (1995), but are prescribed in other models. Biogenic NMVOC emissions in CESM and GEOSCCM are calculated based on the distribution of plant functional types and meteorology conditions with MEGAN, whereas the other models apply prescribed biogenic NMVOC emissions.

The CCMI models do not represent CH_4 emissions explicitly but prescribe CH_4 surface mixing ratios vary in time according to the RCP6.0 scenario (global mean of ~1750ppbv averaged over 2000-2010) with different spatial distributions: GEOSCCM, CESM and EMAC models consider the full latitudinal gradient and prescribe CH_4 surface mixing ratios about 50 ppbv higher over the Northern hemisphere than over the Southern hemisphere; while CMAM, MRI-ESM1r1, and SOCOL3 use global uniform values. Photolysis rates are calculated either following online schemes such as FAST-JX (Neu et al., 2007; Telford et al., 2013) by GEOSCCM, HadGEM3-EA, UMUKA-UCAM, JVAL (Sander et al., 2014) by EMAC, or based on look-up tables with online cloud corrections by the rest of the models used in this study. Kinetics and photolysis data are mainly from Sander et al. (2011) with a few exceptions. More information on model characteristics can be found in Morgenstern et al. (2017) and references listed in Table 1.

Additionally to CCMI OH fields, we also included 2 OH fields simulated by the Interaction with Chemistry and Aerosols (INCA) coupled to the general circulation model of the Laboratoire de Meteorologie Dynamique (LMD), LMDz (Sadourny and Laval, 1984; Hourdin and Armengaud, 1999; Hourdin et al., 2006; Hauglustaine et al., 2004). The two INCA simulations are driven by different versions of the LMDz GCM (INCA NMHC-AER-S covering time period 2000-2010 (Terrenoire et al., 2019), and INCA NMHC covering time period 2000-2009 (Szopa et al., 2013)), which provide different water vapor fields, and include different chemistry and emissions. The INCA NMHC-AER-S used the latest version of INCA model including both gas phase (NMHC) and aerosol (AER) chemistry in the troposphere and the stratosphere (S) (Terrenoire et al., 2019), while INCA NMHC used a former version that only includes tropospheric gas-phase chemistry (Szopa et al., 2013). Anthropogenic emissions from Short-Lived Pollutants (ECLIPSE) inventory (Stohl et al., 2015) for 2005 and RCP 85 emission inventory (Riahi et al., 2011)) for 2010 are applied to every year of INCA NMHC-AER-S and INCA NMHC simulations, respectively.

Finally, we included in this study the OH field used in TransCom simulations, which results from a combination of the semi-empirical tropospheric 3-dimensional OH field from Spivakovsky et al. (2000) and a 2-dimensional simulated stratospheric OH for year 2000. The tropospheric OH was calculated using prescribed chemical species (O_3, nitrogen oxides, and CO) as well as meteorological fields (temperature, humidity, and cloud optical depth) to fit the observations. The original tropospheric [OH] has been reduced by 8% to match CH_3CCl_3 observations (Patra et al., 2011). The TransCom OH field is only climatological (one year of monthly fields).

In total, we compared 14 OH fields: 11 from CCMI, 2 from the online LMDz-INCA model and 1 from TransCom. We analyzed spatial distributions and annual variations of OH fields by calculating volume-weighted tropospheric mean [OH] with tropopause pressure using WMO tropopause definition on 3D temperature for each model (World Meteorological Organization, 1957). Since employing different weightings can results in large differences in mean [OH] (Lawrence et al., 2001), we also calculated dry air mass-weighted tropospheric mean [OH] to better compare with previous studies.

2.2 LMDz model simulations

2.2.1 Model description and setup

We have run the offline version LMDz5B of the LMDz model (Locatelli et al., 2015) at a horizontal resolution of 3.75 °×1.85 ° with 39 vertical layers up to 3 hPa to assess the impact of OH on tropospheric CH_4. All monthly mean OH fields have been interpolated to the LMDz model grid. The transport of atmospheric tracers is driven by prescribed air mass fluxes provided by the general circulation model LMDz with horizontal wind fields nudged to ERA-Interim re-analysis meteorology data produced by the European Center for Medium-Range Weather Forecasts (Dee et al, 2011). The vertical transport is parameterized according to updates of the Emanuel (1991) scheme for convection and of the Louis (1979) scheme for boundary layer mixing (Hourdin et al., 2016; Locatelli et al., 2015). Chemistry module applied here is the simplified chemistry module SACS (Pison et al., 2009). Chemical sinks of CH_4 are calculated using prescribed three-dimensional OH and $O(^1D)$ fields, and variation in CH_4 cannot feedback on OH. No chlorine-related sink is simulated in this version of the model. To assess the influences of OH only, all LMDz simulations used the same $O(^1D)$ fields generated by INCA model simulations. The reaction rate co-efficient (k) for CH_4 destruction by OH in the model is computed depending on temperature following Sander et al. (2011):

$$\mathrm{k} = 2.45 \times 10^{-12} e^{-1775\times(\frac{1}{T})} \qquad (1)$$

The LMDz model has been applied in various studies focusing on long-lived gases such as CH_4, CO_2 and MCF (Bousquet et al., 2005; Pison et al., 2009; Lin et al., 2018). It has also been used in model inter-comparison projects such as the TransCom experiment (Patra et al., 2011) with the simplified chemistry module SACS (Pison et al., 2009) and CCMI (Morgenstern et al., 2017) but only with the stratospheric chemistry model PEPROBUS (Jourdain et al., 2008).

The CH_4 emissions input to LMDz simulations are provided by the Global Carbon Project (GCP) methane and include anthropogenic and biofuel emissions from EDGARv4.3.2 (Janssens-Maenhout et al., 2017), the mean wetland emissions from Poulter et al., (2017), fire emissions from the Global Fire Emissions Database Version 4.1 (GFED4) (Randerson et al., 2018), termite emissions as described by Saunois et al. (2016), geological emissions based on the spatial distribution of Etiope (2015), ocean emissions from Lambert and Schmidt (1993) and soil uptake from Ridgwell et al. (1999). EDGARv4.3.2 data, available until 2012, were extrapolated from 2013 to 2016 using economical statistics according to the methodology described by Saunois et al., (2016). Anthropogenic and fire emissions vary from 2000 to 2016 while natural emissions are applied as a climatology.

The spatial distributions and annual variations of the CH_4 emissions during the study period are shown in Fig.1. CH_4 emissions range from 10 to 40 kg ha^{-1} yr^{1} over most natural ecosystems and can exceed 100 kg ha^{-1} yr^{-1} over wetlands in Canada, South America, and Central Africa, as well as over densely populated regions such as South and East Asia. Global net CH_4 emissions (soil uptake included) increased by 15% from 482 Tg yr^{-1} in 2000 to 552 Tg yr^{-1} in 2016. Of this 70 Tg yr^{-1} increase, 60 Tg yr^{-1} (85%) are emissions from the Northern hemisphere, mainly contributed by livestock (18 Tg yr^{-1}, 25%), oil and gas (16 Tg yr^{-1}, 23%), coal burning (17 Tg yr^{-1}, 24%) and waste (13Tg yr^{-1}, 18%). The three emission peaks in 2002, 2006 and 2015 are driven by biomass burning. This CH_4 emission scenario is state-of-the-art but has not been optimized for the simulated CH_4 mixing ratios to fit the observations.

2.2.2 Model simulations

Two sets of experiments (steady-state and transient simulations) have been performed to examine the impacts of the input OH fields on the global CH_4 burden as well as the CH_4 spatial distribution and annual variation. These tests excluded the OH fields from CESM1-CAM4chem and EMAC-L47MA, since they are similar to those of CESM1-WACCM and EMAC-L90MA, respectively. We also discarded the OH fields from HadGEM3-ES and UMUKCA-UCAM because output from these two models has been supplied on too coarse vertical pressure levels. Finally, 10 different OH fields (seven from CCMI, two from LMDz-INCA and one from Transcom) were used in the two sets of simulations.

Initially, for each OH field described in Sect. 3, we ran 30 consecutive years of LMDz simulations (with recycled same emissions, sinks, and meteorology of the year 2000) to allow the simulation to reach a steady-state (CH_4 has an approximate lifetime of 9 years in the atmosphere). This step aims to examine the impact of the magnitude and distribution of OH on the global CH_4 burden.

Secondly, we performed transient simulations starting from the year 2000, which are forced by time-varying OH fields as well as time-varying emissions and meteorology fields. In order to compare the impacts of the different OH fields on realistic CH_4 mixing ratios, for each simulation (except the one using the OH fields from INCA NMHC), the OH field has been scaled to get the same LMDz simulated CH_4 loss as the one calculated by INCA NMHC in 2000, as INCA is the OH field consistently obtained using the LMDz transport. Then a series of LMDz model simulations is conducted to investigate the impact of the various OH fields on CH_4 growth rates between 2000 and 2016 as summarized in Table 2.

The standard simulations (Run_standard in Table 2) using the 10 different OH fields (7 are from CCMI REF-C1), included annual variations and were performed from 2000 to 2010. Since REF-C1 experiments are only available up to 2010, the influence of OH on CH_4 mixing ratios after 2010 have been tested based on alternative scenarios. First, for CCMI simulations, we tested a scenario that takes into account the annual variability from the REF-C2 experiments (Run_REF-C2 in Table 2). Previous ACCMIP model experiments showed slightly decreasing or increasing [OH] from 2000 to 2030 according to the largest or lowest radiative forcing pathways (RCP8.5 or RCP2.6), respectively (Voulgarakis et al., 2013). Top-down approaches suggested that global OH decreased by 0.5-1% annually from 2003 to 2016 (Rigby et al., 2017; Turner et al., 2017). In order to assess the recent change in [OH], we tested two additional scenarios between 2010 and 2016: one with [OH] increase of +0.1% yr^{-1} (Run_OH_inc) according to the slightly changing of OH calculated by ACCMIP models and one with [OH] decrease of -1% yr^{-1} (Run_OH_dec) according to obviously decreasing of OH calculated by top-down approaches constrained by observations. To assess influences from OH alone, we also conducted additional simulations of the period 2000 to 2016 with annually repeated prescribed [OH] equal to the year 2000 (Run_fix_OH) for each OH field. The differences between these constant OH simulations and the corresponding time-varying OH simulations indicate the impact of OH inter-annual variations and trends on atmospheric CH_4 changes. In addition, we conducted two simulations during 2000-2010 driven by emission inventories fixed to the year 2000 to test the influences of the emission bias on our results. The two simulations use OH fields simulated by CESM-WACCM, one with inter-annual variations of OH (Run_fix_emis) and the other one with OH field fixed to 2000 (Run_fix_emis_OH).

3 Analysis of OH fields

3.1 Spatial distributions of tropospheric OH

Fig. 2 shows the spatial distributions of volume-weighted tropospheric mean [OH] averaged from 2000 to 2010. Based on the 14 OH fields we have assembled, the global mean volume-weighted tropospheric [OH] vary from 8.7×10^5 to 12.8×10^5 molec cm^{-3}. SOCOL, which overestimation of [OH] have been reported by Staehelin et al. (2017), simulated the highest [OH]. To better compare with previous studies, we also calculated dry air mass-weighted tropospheric mean [OH] in table 4, which vary from 9.4×10^5 to 14.4×10^5 molec cm^{-3} and multi-model mean value of $11.3\pm1.3\times10^5$ molec cm^{-3} . The tropospheric chemical CH_4 lifetime of the models that provided CH_4 chemical loss data are 8.7 ± 1.1 yr. Both the multi-model mean and the (large) range of [OH] as well as tropospheric CH_4 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). The model spread remains large as ~50% of the minimum value, as noted in previous studies (e.g. Naik et al., 2013).

Table 3 summarizes their inter-hemispheric ratios of tropospheric OH and mean values over four latitudinal bands. The inter-hemispheric ratios (N/S ratios) of CCMI and INCA OH fields are within the range of 1.2-1.5, similar to those from the ACCMIP project (Naik et al., 2013). In contrast, the TransCom

OH field has a ratio of 1.0, which is more consistent with that of MCF and ^{14}C constrained OH (Brenninkmeijer et al., 1992; Krol and Lelieveld, 2003; Bousquet et al., 2005). However, as discussed by Spivakovsky et al. (2000), the TransCom OH field may overestimate Southern Extra-tropics OH by ~25%. The lower N/S ratios inferred from MCF observations are mainly due to high [OH] over the Southern Tropics (35% higher than Northern Tropics) (Bousquet et al., 2005). In contrast, process-based simulated OH is 10-26% more abundant over the Northern Tropics than over the Southern Tropics, and 35% to > 90% higher over 30 °N-90 °N than 30 °S-90 °S in the CCMI models. Previous studies have attributed the inconsistency between the simulated and the observed OH N/S ratios to a model overestimation of O_3 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 CCMI models (Strode et al., 2016; Revell et al., 2018), as well as to a lack of OH recycling due to the presence of VOCs over rainforest (mainly located in the Southern Tropics) (Lelieveld et al., 2008; Archibald et al., 2011).

We further assessed the simulated OH spread by comparing the detailed spatial distributions of OH fields in Fig. 2 and Fig.S2. Nearly all CCMI models and two versions of the INCA model simulated high [OH] over eastern North American and South and East Asia, which is related to higher tropospheric O_3 concentrations (Cooper et al., 2014; Lu et al., 2018) and NO_x emissions from human activities (Lamarque et al., 2010; Miyazaki et al., 2012). High [OH] over these emission hotspots dominate the aforementioned simulated large N/S ratio. Some models also simulated high OH values over the African savanna plains (MOCAGE and INCA excluded), regions with intense biomass burning (van der Werf et al., 2006) and soil NO_x emissions (Yienger and Levy 1995; Vinken et al., 2014). The O_3 concentrations used to generate the TransCom OH field were larger in the Southern Tropics than in the Northern latitudes (Spivakovsky et al., 2000), in contrast to recent observations (Cooper et al., 2014). Therefore, TransCom shows the highest [OH] over the Southern Tropics during biomass burning seasons (Spivakovsky et al., 2000) and thus a lower N/S ratio.

Despite consistency on high OH values over regions influenced by human activities and biomass burning, models show the largest discrepancies over some natural ecosystem such as tropical rainforests (Fig.S2). For example, INCA, CESM, HadGEM3-Es, MRI-ESM1r1, MOCAGE and GEOSCCM simulated overall low [OH] (4×10^5-14×10^5 molec cm^{-3}) over tropical rainforests, despite differences in details, while EMAC, CMAM, SOCOL3 and UMUKCA-UCAM simulated overall high [OH] (16×10^5- more than 25×10^5 molec cm^{-3}). Tropospheric mean [OH] over the Amazon forest show the largest variations of >5.0 $\times10^5$ molec cm^{-3}, count for more than 50% of 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). Most simulated OH fields show higher concentrations over continents or coastal areas due to higher precursor emissions, while MRI-ESM1r1, EMAC, and GEOSCCM also simulated high values (>15×10^5 molec cm^{-3}) over the open ocean. Factors contributing to these inter-model differences are further discussed in Sect. 3.3

3.2 Vertical distributions

Figure 3 shows the vertical distribution of OH fields and Table 4 provides the volume-weighted mean [OH] averaged over the troposphere and over three pressure latitudinal intervals representing the planetary boundary layer, the mid-troposphere, and the upper troposphere (surface-750, 750-500, and 500-250 hPa, respectively). At the global scale, the mean tropospheric concentration of TransCom OH increases by a factor of nearly two from the surface (7×10^5 molec cm^{-3}) to 600hPa (13×10^5 molec cm^{-3}) and then decreases rapidly with altitude (7×10^5 molec cm^{-3} at 250hPa). UMUKA-UCAM, HadGEM3-ES, CMAM, MOCAGE, and SOCLO3 on the other hand all show a continuous decrease of [OH] with altitude from the surface to the upper troposphere (e.g. the global mean concentrations of MOCAGE OH decreases from 23.6 $\times10^5$ molec cm^{-3} at the surface to 6.4×10^5 molec cm^{-3} at 250hPa). Other OH profiles show much smaller vertical variations in the troposphere (standard deviations of mean value below 200hPa < 2×10^5 molec cm^{-3}).

Model simulated OH vertical distributions can also be different over land versus ocean (Fig. 3) and between the different latitudinal bands (Fig. S3). For example, SOCOL3 [OH] continuously decreases with altitude over both, land and ocean; MOCAGE OH increases from the surface (14.9×10^5 molec cm^{-3}) to 800hPa (18.2×10^5 molec cm^{-3}) and then decreases over land but almost continuously decreases over the ocean; CMAM and UMUKCA-UCAM only show significant vertical variations in [OH] over land. Vertical variations of most OH fields can be attributed to mid and low latitude regions, except for those of SOCOL3 and MOCAGE, that also decrease with altitude over mid and high northern latitudes (45 °N -90 °N, see Fig. S3).

3.3 Factors contributing to inter-model differences

Tropospheric OH is produced primarily through the reaction of $O(^1D)$ with H_2O and secondarily through the reaction of NO with HO_2 and RO_2, and is removed primarily by reacting with CO and CH_4 (Logan et al., 1981). Hence, factors controlling inter-model OH discrepancies can be complex as differences in model emissions, chemistry, and dynamics can together impact [OH]. Here we propose a qualitative analysis focusing on both, emissions and chemical mechanisms. A more quantitative analysis would require a detailed model output of production and loss pathways and is beyond the scope of this work.

To analyze inter-model differences in OH vertical distributions, we compared CO, NO, and O_3 mixing ratios in table 5 as well as $O(^1D)$ photolysis rates and specific humidity in Table S4. The inter-model variations (calculated as standard deviation/multi-model mean) in tropospheric $O(^1D)$ photolysis rates, specific humidity, and CO mixing ratios are usually <10%-20%, while NO mixing ratios show a larger variation of 38% (12-32pptv). MRI-ESM1r1 simulated the highest NO tropospheric mixing ratio, mainly attributable to high values above 250hPa, where OH formation is limited by H_2O. In addition, MRI-ESM1r1 has ~20% more CO emissions than MOCAGE and GEOSCCM (Fig. S5), leading to about 10 ppbv higher CO mixing ratios, offsetting (for [OH]) its higher NO_x emissions and NO mixing ratios. The high NO mixing ratios near the surface and mid-troposphere simulated by SOCOL3 (48 pptv below 750hPa and 10 pptv from 750 to 500hPa), MOCAGE (26 pptv below 750hPa and 14 pptv from 750 to 500hPa) and CMAM (17 ppbv below 750hPa) are consistent with their high tropospheric and near-surface [OH]. Tropospheric O_3 can also influence primary production of OH, and tropospheric O_3 burden reflects combined effects of NO_x, CO, and VOCs. The high O_3 over the lower troposphere simulated by SOCOL3 and low the O_3 over upper troposphere simulated by MOCAGE can contribute to explain the high and low [OH] simulated the two models over the corresponding altitudes, respectively.

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 mixing ratios in the lower troposphere simulated by MOCAGE and SOCOL3 are not corresponding to high lighting NO_x emissions for these models. Besides emissions, previous studies have reported additional factors leading to high surface NO and NO_2. The overestimation of NO by MOCAGE could be due to the lack of N_2O_5 heterogeneous hydrolysis on tropospheric aerosol, which is an efficient sink for NO_x (Teyssèdre et al., 2007). SOCOL3 does not include N_2O_5 heterogeneous hydrolysis and also overestimates tropospheric NO production by NO_2 photolysis compared to other models, due to issues with the look-up tables used in the calculation of photolysis rates (Revell et al. 2018). We conclude here that physical and chemical processes related to NO production and loss can have a large impact on OH burden and its vertical distribution. In this context, an improved representation of the partitioning between NO and other nitrogen species in the models seems of great importance to correctly simulate tropospheric [OH].

Concerning the spatial distributions, as aforementioned in Sect. 3.1, the largest model discrepancies are found over tropical rain forests. The [OH] over tropical rainforest regions are mostly sensitive to natural emissions including NO_x and NMVOCs, which vary among the models. Previous studies showed that [OH] is more sensitive to soil and lightning emissions than to wildfires, because the former sources only emit NO_x (OH source), whereas the latter emits NO_x, CO and VOCs together (OH sources and OH sinks, see Murray et al., 2014). Soil NO_x emissions in CCMI models range from around 4 Tg N yr^{-1} in MOCAGE to more than 7 Tg N yr^{-1} in GEOSCCM and 9Tg N yr^{-1} in CMAM (Naik et al., 2013; Yienger and Levy, 1995); Lighting NO_x emissions range from 3.7-10.2 Tg yr^{-1}(table S3). In particular, lower NO_x emissions over South America and Africa in MOCAGE might be linked to lower [OH] over this region (Fig. S5). Isoprene and other NMVOCs remove about 3% and 7% of tropospheric OH on a global scale, respectively (Spivakovsky et al., 2000; Murray et al., 2014) and can be more important over tropical regions with higher emission rates (Sindelarova et al., 2014). The higher [OH] over tropical rainforests simulated by CMAM and UMUKCA-UCAM may be due to a lack of or the lower OH destruction by VOCs in these models. Therefore, the inter-model differences in OH spatial distributions over tropical rainforests may result from differences in natural emissions of VOC species and different related chemical reactions. 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 photolysis schemes coupled to CCMI models can also influence the calculation of $O(^1D)$ photolysis rates(Sukhodolov et al., 2016).

3.4 Inter-annual variations of OH

Figure 4 shows the time series of volume-weighted tropospheric mean [OH] from 1960 to 2010 (from

REF-C1 CCMI comparison). During this period, all OH fields show small year-to-year variations of 1.9 ±1.2%, remaining within ±0.5 ×10^5 molec cm^{-3}. CCMI models simulated significantly different OH long-term evolutions from 1960 to 1980. For example, [OH] continuously decrease in the CMAM and HadGEM3-ES simulations (~-0.3 ×10^5 molec cm^{-3}; -3.4%); and increase in SOCOL3 (~+0.6 ×10^5 molec cm^{-3}; +4.5%), UMUKCA-UCAM (~+0.5 ×10^5 molec cm^{-3}; +4.8%), and MOCAGE (~+0.5 ×10^5 molec cm^{-3}; +4.8%) during 1960-1980, while other models show no obvious long-term trend. After 1980 (1990 for CMAM), all models show stabilized or slightly increasing [OH]. For our period of interest (after 2000) and focusing on the anomaly in [OH] compared to the 2000-2010 mean (Fig. 4b), OH year-to-year variations are found to be smaller than in previous decades and [OH] only increases by about 0.1-0.3 ×10^5 molec cm^{-3} from 2000 to 2010.

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 CH_4 (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 ±1.0% from 1980 to 2000, and 2.3 ±1.5% from 1998 to 2007, as compared to 2.1 ± 0.8% and 1.0 ±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 ±2.2% from 1980 to 2000 (Naik et al., 2013). Here, for the same period the increase of CCMI [OH] is 4.6 ±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 CH_4 growth during the 2000s, as opposed to the decreasing in [OH] from mid-2000s calculated by Rigby et al. (2017) and Turner et al (2107) based on MCF observations.

We further analyzed regional [OH] trends from 2000 to 2010 in Fig. 5. Instead of dividing subdomains as Naik et al. (2013) did, we calculated the trend for each model grid-cell to identify and distinguish regions with different trends. Most models show significant positive [OH] trends over tropical regions (0.05-0.1×10^5molec cm^{-3} yr^{-1}) and over East and Southeast Asia ($>0.1\times10^5$ molec cm^{-3} yr^{-1}). By comparing spatial distribution of OH trend with specific humidity (Fig.S6a), NO_x and CO emissions (Fig. S6b), and stratosphere O_3 column trend, we find that positive OH trend over tropical regions are mainly corresponding to increases in water vapor (Fig. S6a) while faster NO_x emission increases (>5% yr^{-1}) than CO (<2% yr^{-1}) are consistent with positive OH trend over East and Southeast Asia (Fig. S6b). From 2000 to 2010, NO_x emissions in the MACCity (RCP85) inventory increased by 83% over East Asia, which is much larger than the CO increase (8%) (Riahi et al., 2011). Over the rest of the extra-tropical regions such as North America and Western Europe, the models disagree on the sign of OH change. In the Southern hemisphere, where biogenic and fire emissions dominate, most OH fields do not show clear trends and the inter-model differences are even larger. For example, MOCAGE simulated and OH decrease of $>0.1\times10^5$ molec cm^{-3} yr^{-1} over the Amazon, South Africa, and Indonesia, whereas MRI-ESM and EMAC-L90MA simulated positive OH trends over these regions. CMAM and HadGEM3-ES show significant increasing and decreasing OH trend over the Antarctic region, respectively, consistent with the significant changes found for stratospheric O_3 (Fig. S6c).

In the following, we investigate how the differences in mean [OH] and variations presented in this section affect CH_4 burden and its variations for the period 2000-2016.

4 Influences of OH fields on CH_4 simulations

4.1 Global total CH_4 burden

We now present the results based on the first set of LMDz experiments, where the LMDz model was run for 30 years recycling the year 2000 until the steady-state is reached. The simulations using the OH fields as given by CCMI and INCA models provide a wide range of values for the tropospheric global mean CH_4 mixing ratios (Table 6), from 1204 ppbv (SOCOL3, with a global volume-weighted tropospheric mean [OH] of 12.8×10^5 molec cm^{-3}) to 1822 ppbv (INCA NMHC-AER-S, with a global volume-weighted tropospheric mean [OH] of 8.7×10^5 molec cm^{-3}). It appears that the global CH_4 burden is not only sensitive to the global mean [OH], but also to its vertical distribution. Indeed, the OH radicals in the lower troposphere are more efficient to oxidize CH_4 molecules, because the CH_4+OH reaction rate increases with temperature (Eq. 1). When considering the standard atmosphere, the reaction rate corresponding to the surface temperature of 288K (5.2×10^{-15} s^{-1}) is more than twice that for the 500hPa temperature of 253K (2.2×10^{-15} s^{-1}). Despite similar volume-weighted tropospheric mean [OH] of ~10.4 $\times 10^5$ molec cm^{-3}, MOCAGE simulated much lower CH_4 mixing ratios (1275 ppbv) than CMAM (1540 ppbv) and MRI-ESM1r1 (1639 ppbv) because of its higher near surface [OH] (19×10^5 molec m^{-3}) (Table 4). Previous studies have demonstrated that the sensitivity of CH_4 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).The spatial distribution of the OH radicals also slightly influences CH_4 oxidation. Indeed, the [OH] of EMAC-L90MA are higher than those of CESM-WACCM for both, tropospheric (11.1×10^5 versus 10.7×10^5 molec cm^{-3}) and near-surface (12.5×10^5 versus 12.4×10^5 molec cm^{-3}) means, but a slightly higher CH_4 burden is found for the former (1579 versus 1575ppbv, Table 6). This is because EMAC-L90MA simulated higher [OH] over the ocean, while CESM-WACCM OH is more concentrated over land closer to CH_4 source regions. The model experiments also emphasize that volume-weighted tropospheric concentrations cannot fully indicate the atmospheric oxidizing efficiency for CH_4, as has been discussed by Lawrence et al. (2001). Tropospheric mean [OH] weighted by reaction rates with CH_4, which consider both temperature and CH_4 distributions, can be a better indicator for CH_4 oxidation (Lawrence et al., 2001).

4.2 Impacts on CH_4 spatial distribution and growth rate

In order to address the question of inter-annual variability of atmospheric CH_4, we scaled each OH field globally to get the same CH_4 loss (for the year 2000) as the one obtained with INCA NMHC OH field (see Sect. 2.2.2). The single global scaling factor (per OH field) for the year 2000 is applied to every year between 2000 and 2010. As listed in Table 4, after scaling most OH fields have volume-weighted tropospheric mean concentrations closer to INCA NMHC (9.7×10^5 molec cm^{-3}), within the range of 9.0-10.4×10^5 molec cm^{-3}. One exception is MOCAGE, with tropospheric mean [OH] scaled to 7.7×10^5 molec cm^{-3}, due to its distinct vertical distribution (Sect.3.2). This scaling of OH makes it possible to start model experiments at the same initial CH_4 burden. Although slightly modifying the magnitude of the global mean [OH], this scaling maintained the spatial and temporal differences and trend over the 2000-2010 period.

4.2.1 Spatial distributions of tropospheric CH_4 mixing ratios

We used the scaled OH fields to perform simulations between 2000 and 2010. Figure 6 shows the spatial distribution of tropospheric CH_4 mixing ratios for the simulation Run_standard (Table. 2, driven by OH with inter-annual variations) averaged over 2000-2010. Although all simulations started from the same initial conditions and OH fields were scaled to give the same global CH_4 loss as INCA NMHC in 2000, LMDz simulations using the different scaled OH fields still generated a spread of tropospheric mean (8ppbv) and spatial distribution of CH_4 mixing ratios averaged during 2000-2010. Differences between the global tropospheric mean [OH] cannot explain these differences (see Table 4).Clearly the different spatial (horizontal and vertical) and temporal variations of the OH fields (as described in Sect. 3), which were kept in this experiment by only scaling [OH] globally, significantly modify the simulated CH_4 mixing ratios (Table 7 and Fig. 6). OH fields with increasing trend will lead to lower LMDz simulated CH_4 mixing ratios. The LMDz simulations using the TransCom OH fields (without inter-annual variability) shows the highest CH_4 mixing ratios (1735 ppbv), while the one using the CMAM OH (with slightly increasing OH trend during the decade) shows the lowest mixing ratios (1727 ppbv).

The differences in spatial distribution of OH fields can influence LMDz simulated CH_4 spatial distributions. Looking at latitudinal CH_4 mixing ratios, the inter-model differences appear larger than in the global mean (Fig. 6 and Table 7). The model spreads of the mean CH_4 mixing ratios over 60 °S-90 °S and 60 °N-90 °N range from 1771 to 1794ppbv and 1784 to 1812ppbv, respectively. Here, we define the N/S gradient of CH_4 as the difference in mean CH_4 mixing ratio between the latitudinal bands 60 °N - 90 °N and from 60 °S - 90 °S. With the TransCom OH field (N/S ratio =1.0), the model simulated 12-43% larger N/S gradients of CH_4 (129 ppbv) than other simulations (90-115 ppbv) driven by OH fields with higher N/S OH ratios of 1.2-1.5. Previous model studies have attributed the overestimation of the CH_4 N/S ratio to an underestimation of model inter-hemispheric exchange time (e.g. Zimmermann et al., 2018). Our results show that uncertainties in OH distributions can also contribute to such model biases.

4.2.2 Changes in CH_4 mixing ratios

To assess the influence of OH inter-annual variations on CH_4 mixing ratios, we calculated the difference in the simulated CH_4 between the standard run (Run_standard) and the simulations with fixed [OH] (Run_fix_OH, Table 2). The Run_fix_OH simulations show that global tropospheric mean CH_4 mixing ratios increased by 75 ppbv from 2000 to 2010 (Fig. 7, black dashed lines), due to the enhanced emissions (Fig.1). The increase in [OH] can obviously reduce CH_4 growth. An increase in [OH] by $0.1\text{-}0.3\times10^5$ molec cm^{-3} (1%-3%) (Fig. 7, orange lines) during this period leads to a reduction of the CH_4 mixing ratios by 5-15 ppbv by 2010 (Fig. 7, blue lines).The largest reductions are found when using CESM1-WACCM and CMAM OH fields, given the continuous OH growth in these models. Compared to Run_fix_OH, we estimated that such reductions in CH_4 mixing ratios offset 7- 20% of the CH_4 increase driven by the rising CH_4 emissions of our scenario over the period 2000 to 2010.

To test whether the impacts of [OH] year-to-year variations on CH_4 mixing ratios the chosen emission scenarios, we compare the above results with that calculated by an extreme scenario where model simulations are driven by fixed emission(year 2000, Run_fix_emis and Run_fix_emis_OH, table 2). With emissions fixed to 2000, the CH_4 mixing ratio increased by 2ppbv from 2000 to 2010, and increasing OH (CESM-WACCM OH fields) can reduce CH_4 mixing ratio by 13.5ppb in 2010, comparable to 13.9 ppb calculated by Run_std and Run_fix_OH with CESM-WACCM OH fields. The results indicate only a small effect of emission scenario choices on the absolute changes of CH_4 mixing ratios due to OH variations. However, our choices have a large effect on relative change to the total modeled CH_4 increase. Indeed, if we use the emission scenarios that match observations (~+25ppbv of CH4 mixing ratio increase from 2000-2010, Ed Dlugokencky, NOAA/ESRL, 2019) instead of ~70ppb here , the CH_4 mixing ratio changes due to OH can contribute to more than half (13.5-13.9ppbv versus 25ppbv) of the changes driven by emissions.

After 2010, CCMI REF-C2 experiments simulated increasing, relatively stable, or decreasing OH variations, thus having a variable influence on CH_4 variations. Over the period 2011-2016, [OH] simulated by EMAC-L90MA, CESM-WACCM and CMAM stabilizes at a level $0.2\text{-}0.4\times10^5$ molec cm^{-3} higher than the concentrations in 2000, further reducing CH_4 mixing ratios by up to 20-30 ppbv in 2016 (Fig. 7, blue lines). Other OH fields have similar concentrations over 2010-2016 as in the early 2000s (Fig. 7, orange lines), thus simulating CH_4 mixing ratios that remain close to Run_fix_OH with differences less than a few +/-ppbv.

As large uncertainties remain regarding the inter-annual variations and trend of OH after 2010, we have tested two additional OH scenarios: Run_OH_inc (with an annual increase of 0.1% yr^{-1}) and Run_OH_dec (with an annual decrease of 1% yr^{-1}), to assess the uncertainty range of the impact of OH changes (the orange areas in Fig. 7) on CH_4 mixing ratios (the blue areas in Fig. 7). In these two scenarios, the mean [OH] of run_OH_dec is ~ 7×10^5 molec cm^{-3} (7%) lower than run_OH_inc in 2016 relative to the Run_fix_OH . If OH decreases at 1% yr^{-1} after 2010, by 2016, the differences in CH_4 mixing ratios between Run_OH_dec and Run_fix_OH range -7–30ppbv, with the lower end (-7ppbv) simulated by OH from CESM1-WACCM given its highest [OH] in 2010. On the contrary, Run_OH_inc simulated 3-39 ppbv lower CH_4 mixing ratios compared to Run_fix_OH (the blue areas in Fig. 7). As such, uncertainties in the OH trend can clearly lead to >±30 ppbv changes in CH_4 mixing ratios (the gray areas in Fig. 7) after only 6 years of simulations, as compared to the fix-OH case.

It is now interesting to compare the range of simulated [CH_4] changes induced by OH scenarios to changes in surface CH_4 observations, in order to quantify how much of the model-observation mismatch could potentially be attributed to uncertainties in [OH] and its variability (Fig. 8). To do so, we used surface CH_4 observations from the National Oceanic and Atmospheric Administration (NOAA) networks and selected stations with 17 years continuous records over 2000-2016. The modeled surface CH_4 mixing ratios are sampled according to station locations. Since the simulated absolute CH_4 mixing ratios largely depend on the initial conditions and OH fields, we compared changes in the simulated and observed global CH_4 mixing ratios starting at the same point in 2000. The observed CH_4 shows zero growth between 2000 and 2006 and then increases by 5.6 ppbv yr^{-1} between 2006 and 2012 (6.4 ppb yr^{-1} for 2006-2010) and by 9.4 ppbv yr^{-1} after 2012(Fig.8). In this study we do not expect to fit these CH_4 trends as this inter-comparison was not conducted with a set of optimized emissions. It has already been noticed that standard CH_4 emission inventories lead to overestimated CH_4 mixing ratios (e.g. Saunois et al., 2016). Indeed, neither Run_standard nor Run_fix_OH simulations do capture the stagnation during 2003-2006, and overestimated surface CH_4 increments by 2.5-5.2 ppbv yr^{-1} during the period 2006-2010. We define highest CH_4 mixing ratios simulated by different OH as $CH_{4\text{-H}}$, lowest CH_4 mixing ratios as $CH_{4\text{-L}}$, and CH_4 simulated by Run_fix_OH as $CH_{4\text{-fix\_OH}}$. 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 ($CH_{4\text{-H}}$－$CH_{4\text{-fix\_OH}}$)/($CH_{4\text{-fix\_OH}}$- observed CH_4)) during 2000-2016), or limit the model-observation mismatch by up to 54% (mean values of ($CH_{4\text{-fix\_OH}}$－$CH_{4\text{-fix\_L}}$)/($CH_{4\text{-fix\_OH}}$- observed CH_4) during 2000-2016) (figure 8). Such comparisons strongly suggest that a better understanding of OH inter-annual variations and trends is required in order to simulate more reliable CH_4 trends in atmospheric chemistry models. Atmospheric chemistry transport model (Dalsøren et al., 2016) and box model studies (Rigby et al., 2017; Turner et al., 2017) have pointed out that variations in OH can partly explain the recent CH_4 trends. However, current top-down estimates of CH_4 emissions usually assume constant [OH] (Saunois et al., 2017) and attribute the model-observation discrepancies only to surface emissions rather than changes in [OH]. Our results confirm the potentially significant role played by the still uncertain OH changes in the actual changes of methane emissions since 2000.

5 Conclusions

We have analyzed 14 OH fields (11 from CCMI experiments, 2 from INCA model simulations, 1 from TransCom) to investigate the inter-model differences in the spatial distributions and trends of tropospheric OH, and estimated the influences of OH spatio-temporal distributions on tropospheric CH_4 by feeding them in different simulations with LMDz offline chemistry transport model.

Simulated global volume-weighted tropospheric mean [OH] are within the range of 8.7×10^5-12.8×10^5 molec cm^{-3}, which is consistent with the (large) multi-model range of previous estimates. CCMI and INCA models simulated larger [OH] in the Northern hemisphere than in the Southern hemisphere (N/S ratio of 1.2-1.5), with consistently high OH values over anthropogenic emission hotspots in North America, East and South East Asia, while TransCom OH shows a N/S ratio close to 1.0. In the vertical, TransCom OH reaches its maximum value at about 600 hPa, while CCMI and INCA OH fields either continuously decrease with altitude or show very small vertical variations in the troposphere. The factors most likely responsible for these inter-model differences include i) large NO mixing ratios leading to high surface and mid-tropospheric [OH] (Teyssèdre et al., 2007; Pevell et al. 2018), and ii) different natural emissions as well as VOC species and chemical mechanisms driving spatial model discrepancies over natural ecosystems.

Simulated OH fields show small year-to-year variations, within ±0.5×10^5 molec cm^{-3} during 1960-2010. From 2000 to 2010, year-to-year variations in OH are smaller than in previous decades and all OH fields increase by about $0.01\text{-}0.03\times10^5$ molec cm^{-3} yr^{-1}. Such an increase in OH is mainly attributed to the significant positive OH trend over East and Southeast Asia (>0.1×10^5molec cm^{-3} 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.

The inter-model differences in tropospheric OH burden generate a wide range of CH_4 burdens (1204-1882ppbv) when used to simulate steady-state CH_4 mixing ratios in the atmospheric chemistry model LMDz. Our findings suggest that not only different global mean [OH], but also differences in the horizontal and vertical distributions between OH fields are responsible for this range (CH_4 destruction rates by OH increase with temperature).

The CH_4 simulations for 2000-2016 using OH with inter-annual variation show that inter-model differences of the OH N/S ratio lead to 12-43% differences in the CH_4 N/S gradient. For the time period 2000-2010, we found that a 1%-3% increase in [OH] leads to a 5-15 ppb reduction of the CH_4 mixing ratio until 2010, accounting for 7-20% of the simulated emission driven CH_4 increase over this period. After 2010, the ensemble of OH scenarios tested here leads to differences in CH_4 mixing ratio of up to 30 ppb by 2016. Comparing with surface observations, we found that [OH] changes can emphasize the model-observation mismatch by up to 19%, or fill the gap between model simulations and observations by up to 54% (Figure 8). Therefore, addressing the OH variability in CH_4 source inversions seems critical to avoid a wrong attribution of CH_4 changes to emission changes only. Future work is needed to quantify the impact of this ensemble of OH fields on CH_4 emissions obtained by inversion and to generate improved OH fields to be used in CH_4 inversion studies.

Author contributions

YZ, MS, and PB designed the study, analyzed data and wrote the manuscript. BZ and XL helped with data analysis and model simulations. JC, RJ, and AS discussed the results. ED provided the atmospheric in situ data. Other co-authors provided numerical model outputs. All co-authors commented on the manuscript.

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Tables

Table 1. List of CCMI models included in this study with model versions and references.[1]

Model	**Version**	**References**
CESM1-CAM-chem	CCMI_23	Tilmes et al.(2015, 2016)
CESM1-WACCM	CCMI_30	Solomon et al. (2015); Garcia et al. (2016); Marsh et al. (2013)
CMAM	v2.1	Jonsson et al. (2004); Scinocca et al. (2008)
EMAC(offers two resolutions: EMAC-L47MA and EMAC-L90MA)	v2.51	Jöckel et al. (2010, 2016)
GEOSCCM	v3	Molod et al. (2012, 2015); Oman et al. (2011, 2013); Nielsen et al. (2017)
HadGEM3-ES	HadGEM3 GA4.0, NEMO 3.4, CICE, UKCA, MetUM8.2	Walters et al.(2014); Madec(2008); Hunke and Lipscombe(2008); Morgenstern et al.(2009); O'Connor et al.(2004); Hardiman et al.(2017)
MOCAGE	v2.15.1	Josse et al. (2004); Guth et al. (2016)
MRI-ESM1r1	v1.1	Yukimoto et al. (2012, 2011); Deushi and Shibata (2011)
SOCOL3	v3	Revell et al. (2015); Stenke et al. (2013)
UMUKCA-UCAM	MetUM 7.3	Morgenstern et al. (2009); Bednarz et al. (2016)

[1] The table refers to Table 2 in Morgenstern et al. (2017)

Table 2. List of LMDz experiments and model setups.

	Simulation period	Inter annual variability in [OH]	Inter annual variability in CH4 emissions
Run_standard	2000-2010	2000-2010	2000-2010
Run_REF-C2	2011-2016	2010 apply inter-annual variability from REF-C2	2011-2016
Run_OH_inc	2011-2016	2010 apply annual growth rate of 1‰	2011-2016

Run_OH_dec	2011-2016	2010 apply annual decrease rate of 1%	2011-2016
Run_fix_OH	2000-2016	Constant OH (year 2000)	2010-2016
Run_fix_emis	2000-2010	2000-2010(CESM-WACCM only)	Constant (2000)
Run_fix_emis_oh	2000-2010	Constant OH (year 2000 CESM-WACCM only)	Constant (2000)

Table 3. Inter-hemispheric ratios (N/S) of hemispheric mean OH and volume-weighted tropospheric mean [OH] for four latitude bands (in 10^5 molec.cm^{-3}) averaged over the years 2000 to 2010. Multi-model means and standard deviations (Mean ± stand. dev.) are also shown.

OH fields	N/S ratio	90 °S-30 °S (10^5 molec.cm^{-3})	30 °S-0 ° (10^5 molec.cm^{-3})	0 °-30 °N (10^5 molec.cm^{-3})	30 °N-90 °N (10^5 molec.cm^{-3})
TransCom	1.0	5.8	12.7	11.8	6.2
INCA NMHC-AER-S	1.3	4.7	10.6	12	7.5
INCA NMHC	1.2	5.7	11.9	13.4	7.8
CESM1-CAM4Chem	1.4	5.7	12.4	15.3	9.2
CESM1-WACCM	1.3	5.9	12.3	15.1	9.3
CMAM	1.2	5.6	13.1	14.3	8.3
EMAC-L47MA	1.2	6	13.5	15.6	8.4
EMAC-L90MA	1.2	6.3	13.8	15.7	8.6
GEOSCCM	1.2	6.5	14.8	16.8	9.1
HadGEM3-ES	1.4	4.1	10.4	12.5	8.1
MOCAGE	1.5	5.5	11.4	14.3	10.2
MRI-ESM1r1	1.2	4.7	13.7	15.3	7.3
SOCOL3	1.5	6.8	13.5	17.0	14.0
UMUKCA-UCAM	1.3	5.6	13.7	14.9	9.9
Mean ± stand. dev.	1.3±0.1	5.6 ±0.7	12.7±1.3	14.6±1.6	8.9±1.8

Table 4. Global mean [OH] averaged over the troposphere and three vertical pressure levels (in 10^5 molec cm^{-3}) over the years 2000 to 2010. Multi-model means and standard deviations (Mean ± stand. dev.) are also shown.

	Tp-v[1]	Tp-m[2]	750[3]	500	250	Tp scaled[4]	CH_4 lifetime[5]
TransCom	9.1	10.0	9.9	12.8	9.2	9.5	/

INCA NMHC-AER-S	8.7	9.4	11.3	10.4	7.8	9.3	/
INCA NMHC	9.7	10.4	11.8	11.4	8.9	9.7	/
CESM1-CAM4Chem	10.7	11.3	12.2	12.3	10.7	/	9.4 yr
CESM1-WACCM	10.7	11.4	12.4	12.5	10.7	9.9	9.3 yr
CMAM	10.4	11.3	14.3	11	10.5	9.3	9.0 yr
EMAC-L47MA	10.9	11.3	12.1	12	10.3	/	/
EMAC-L90MA	11.1	11.5	12.5	12.2	10.2	10.3	/
GEOSCCM	11.8	12.3	12.3	13.7	12	10.4	8.9 yr
HadGEM3-ES	8.8	9.9	12.7	10.8	7.7	/	/
MOCAGE	10.4	12.5	19	13.5	7.7	7.7	7.5 yr
MRI-ESM1r1	10.3	10.6	12.2	10.4	9.4	10.2	10.0 yr
SOCOL3	12.8	14.4	19.4	15.1	10.9	9.0	7.1 yr
UMUKCA-UCAM	11.0	11.9	14.9	11.7	10.5	/	/
Mean ± stand. Dev.	10.5 ±1.1	11.3 ±1.3	13.4 ±2.7	12.1 ±1.3	9.8 ±1.3	9.5 ±0.8	8.7 ± 1.1 yr

[1] Tp-v refers to the volume-weighted tropospheric mean [OH].

[2] Tp-m refers to the mass-weighted tropospheric mean [OH]

[3] 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.

[4] Tp scaled refer to the volume-weighted global tropospheric mean [OH] after scaling to the same CH_4 loss as with INCA NMHC in 2000.

[5] CH_4 lifetime is calculated global atmospheric CH_4 burden divided by annual total CH_4 tropospheric chemical loss.

Table 5. Global volume-weighted mean CO, N, and O3 mixing ratios averaged over the whole troposphere and three pressure altitude levels for CCMI models over 2000 to 2010.[1] Multi-model means and standard deviations (Mean ± stand. dev.) are also shown.

	CO ppbv				**NO pptv**				**O_3 ppbv**			
	750	500	250	Tp	750	500	250	Tp	750	500	250	Tp
CESM1-CAM4Chem	76	71	70	71	9	4	12	13	32	42	57	48
CESM1-WACCM	75	70	69	70	9	5	12	12	31	41	55	47
CMAM	77	68	64	69	17	4	17	26	34	43	60	52
EMAC-L47MA	85	77	70	75	8	4	11	14	38	48	63	56
EMAC-L90MA	84	76	69	74	8	5	11	17	38	48	61	54
GEOSCCM	78	74	73	74	9	5	13	13	33	43	61	49
MOCAGE	67	68	67	67	26	14	17	20	37	42	46	43
MRI-ESM1r1	93	86	83	86	10	5	20	32	36	48	67	56

SOCOL3	79	73	74	74	48	10	14	25	43	54	67	61
Mean ± stand. dev.	79 ± 7	74 ± 6	71 ± 5	73 ± 5	16 ± 13	6 ± 3	14 ± 3	19 ± 7	36 ± 4	45 ± 5	60 ± 7	52 ± 6

[1] HadGEM3-ES and UMUKCA-UCAM are not analyzed since model output has been regridded to too coarse vertical pressure levels.

[2] Tp refers to the total tropospheric average, 750 refers to the average from the surface to 750hPa, 500 refers to the average from 750hPa to 500hPa, and 250 refers to the average from 500hPa to 250hPa.

Table 6. Global mean tropospheric CH_4 mixing ratios as simulated by LMDz using different OH fields and repeating year 2000 over 30 times.

CH_4 mixing ratio (ppbv)			
INCA NMHC-AER-S	1822	**CESM1-WACCM**	1575
TransCom	1776	**CMAM**	1540
INCA NMHC	1709	**GEOSCCM**	1503
MRI-ESM1r1	1693	**MOCAGE**	1275
EMAC-L90MA	1579	**SOCOL3**	1204

Table 7. LMDz simulated CH_4 mixing ratios (in ppbv) averaged over each latitudinal band and the years 2000 to 2010 simulated from the standard experiment (Run_standard) using different OH fields. Multi-model means and standard deviations (Mean ± stand. dev.) are also shown.

	90°S-60°S	60°S-0°	0°-60°N	60°N-90°N	N/S gradient[1]
TransCom	1683	1697	1769	1812	129
INCA NMHC-AER-S	1687	1698	1757	1795	108
INCA NMHC	1687	1700	1762	1802	115
CESM1-WACCM	1688	1701	1757	1794	106
CMAM	1682	1694	1756	1796	114
EMAC-L90MA	1685	1698	1759	1798	113
GEOSCCM	1688	1701	1764	1803	115
MOCAGE	1686	1699	1753	1788	102
MRI-ESM1r1	1691	1702	1762	1803	112
SOCOL3	1694	1707	1754	1784	90
Mean ± stand. dev.	1687 ± 4	1700 ± 3	1759 ± 5	1798 ± 8	110 ± 10

[1] N/S gradient is defined as the difference between 60°N to 90°N and 60°S to 90°S.

Figures

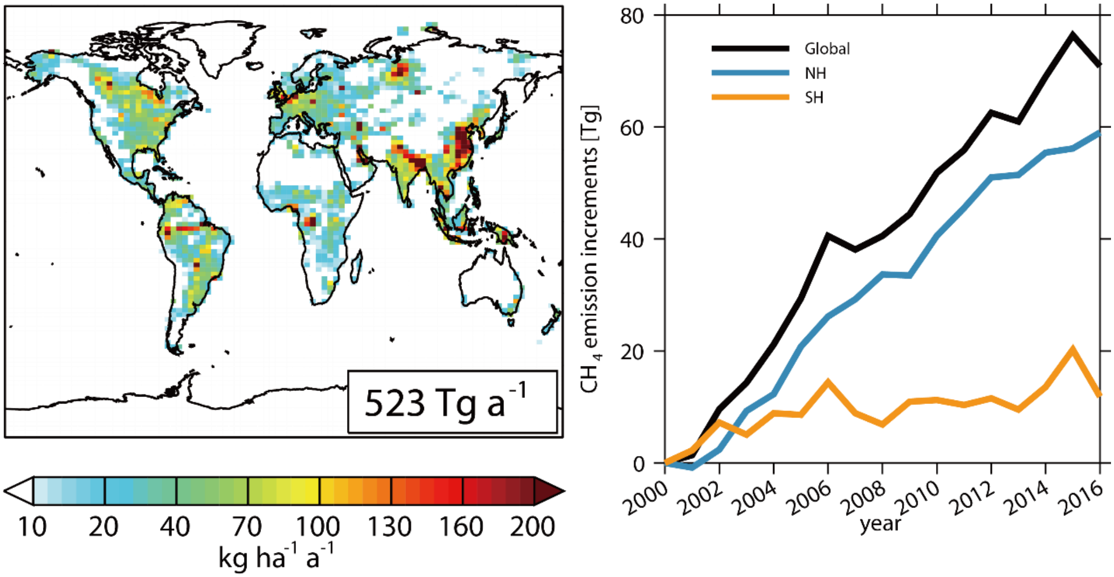

Figure 1. Spatial distribution of global CH_4 emissions averaged between 2000 and 2016 (left) and a time series of CH_4 emissions relative to year 2000 emissions (482 Tg CH_4 a^{-1}) (right) for the globe (black line), Northern hemisphere (NH, blue line) and Southern hemisphere (SH, yellow line), respectively.

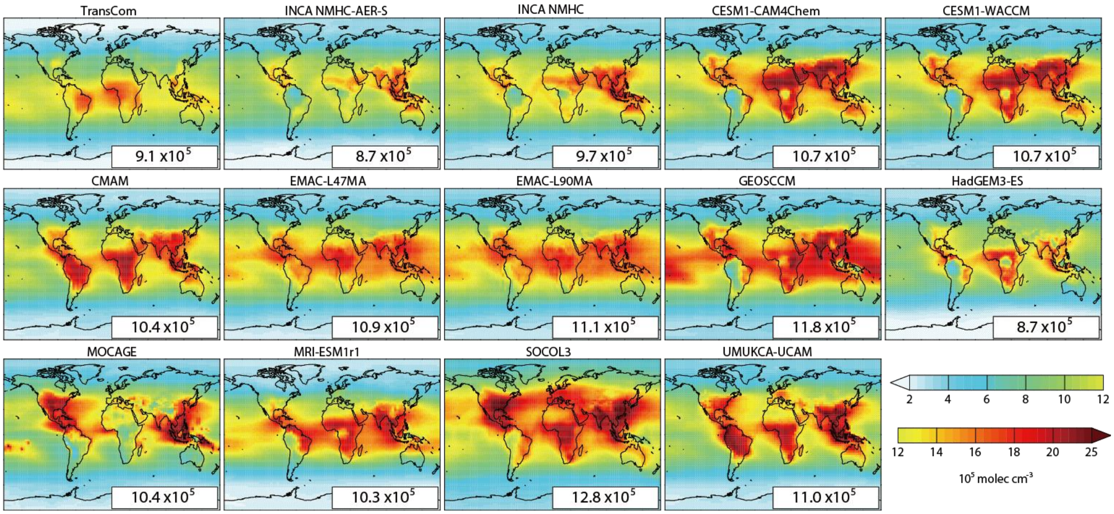

Figure 2. The spatial distributions of volume-weighted tropospheric mean OH fields of TransCom, INCA, and CCMI models averaged for 2000-2010. Global mean values (10^5 molec cm^{-3}) are shown as insets.

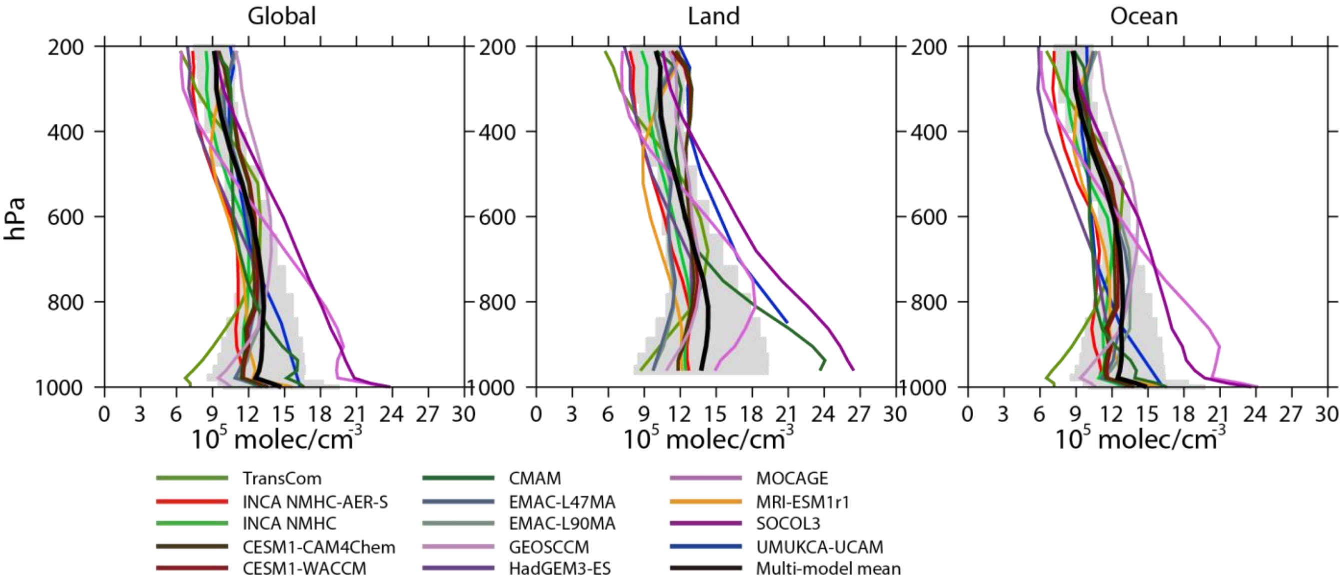

Figure 3. Vertical distributions of [OH] averaged over the globe (left), land (middle) and ocean (right) for 2000-2010. Color lines represent [OH] from individual model simulations, black lines represent multi-model mean values and grey shades represent the standard deviations.

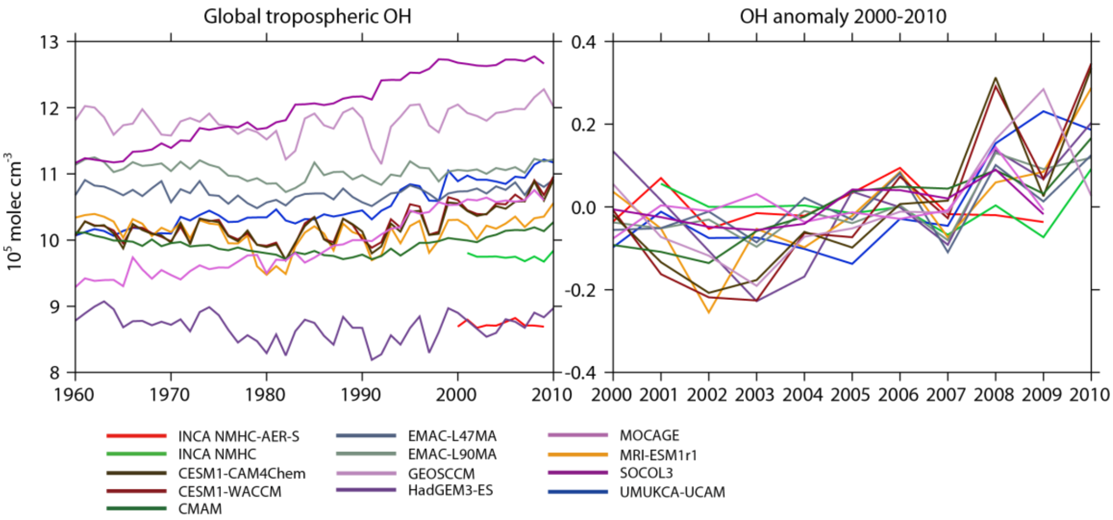

Figure 4. Left: Inter-annual variations of global volume-weighted tropospheric mean [OH] from CCMI and INCA model simulations from 1960 to 2010. Right: OH anomaly during 2000-2010, in reference to the mean concentration over the period 2000-2010 for each model.

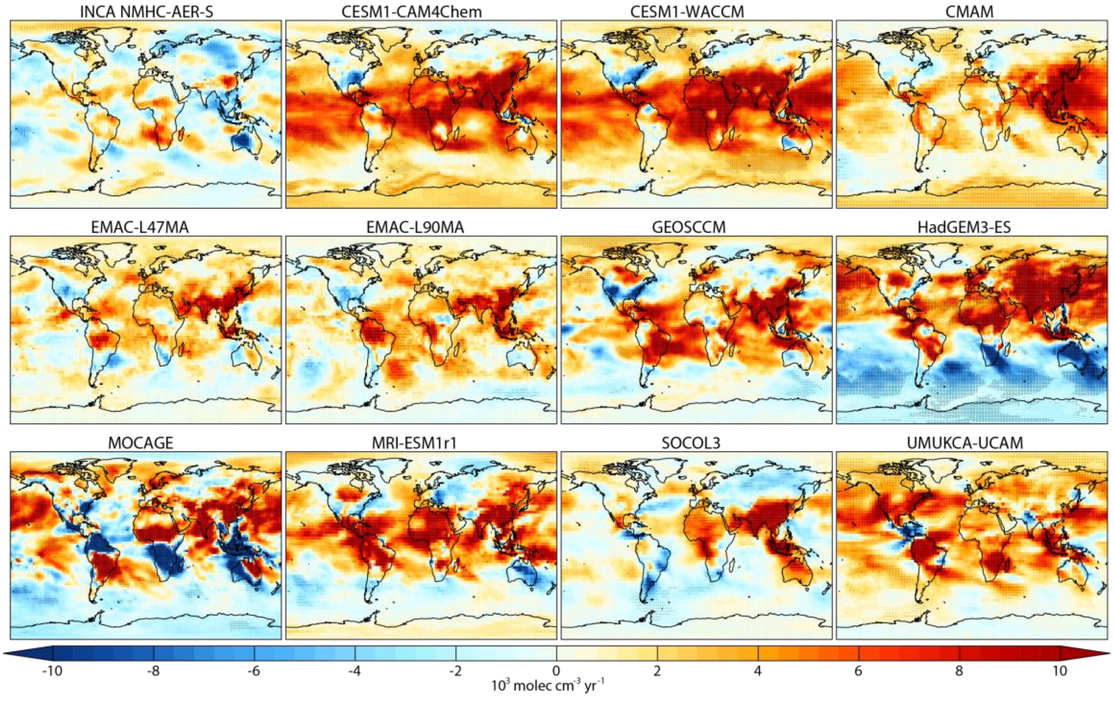

Figure 5. Spatial distribution of tropospheric OH trends from 2000 to 2010 (in 10^3 molec cm^{-3} yr^{-1}). Black dots denote model grid-cells with statistically significant trends (p-value < 0.05)

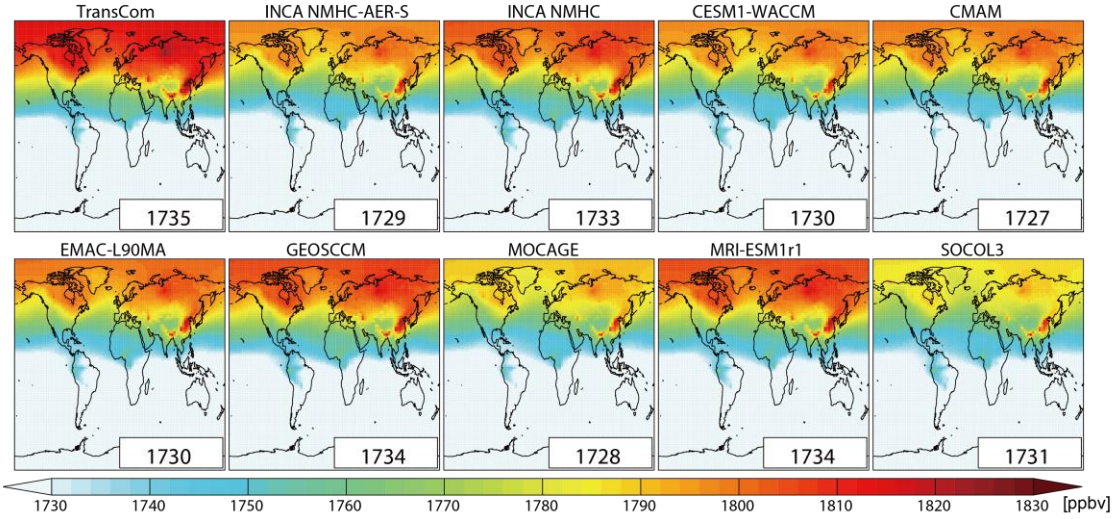

Figure 6. Spatial distribution of volume-weighted tropospheric mean CH_4 mixing ratios averaged from 2000 to 2010 as simulated by LMDz with different OH fields in the LMDz model. The global mean values in units of ppbv are shown as insets.

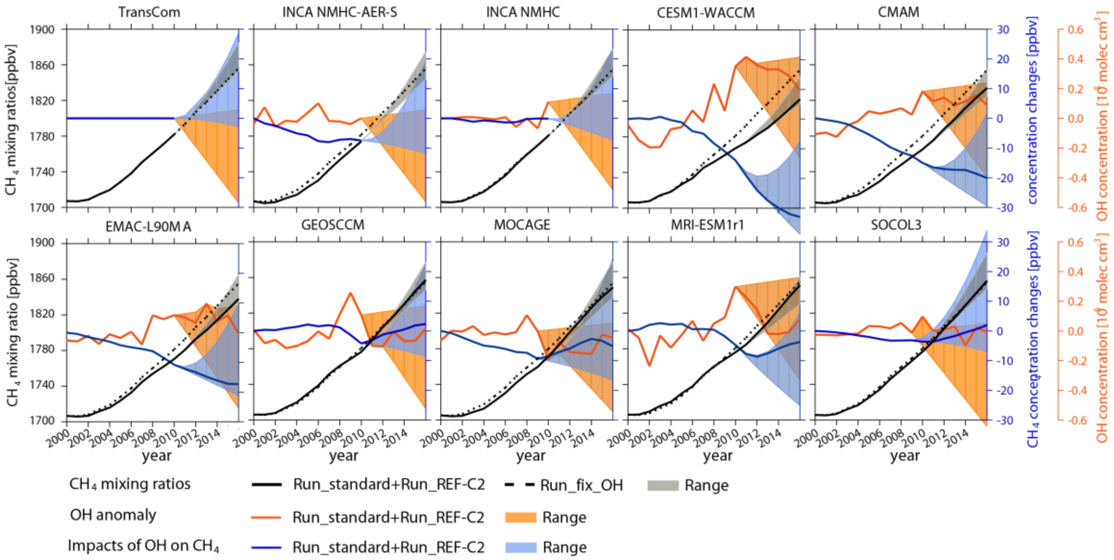

Figure 7. Time-series of global tropospheric CH_4 mixing ratios and [OH] associated with the model experiments listed in Table 2. The black lines represent the evolution of CH_4 mixing ratios with varying (solid lines) or with constant (dashed lines) OH. The varying OH case is obtained using OH inputs from Run_standard from 2000 to 2010 followed by Run_REF-C2 from 2011 to 2016 (see Table 2). The blue solid lines represent the corresponding differences between the simulations with varying OH and with constant OH. The orange solid line represents the corresponding anomalies in tropospheric [OH] (with

the average over 2000-2010 as reference). The shaded areas correspond to the range obtained from all simulations over 2010-2016 (Table 2) for tropospheric CH_4 mixing ratios (grey), for changes in tropospheric CH_4 mixing ratios (blue) and for changes in tropospheric [OH] (orange).

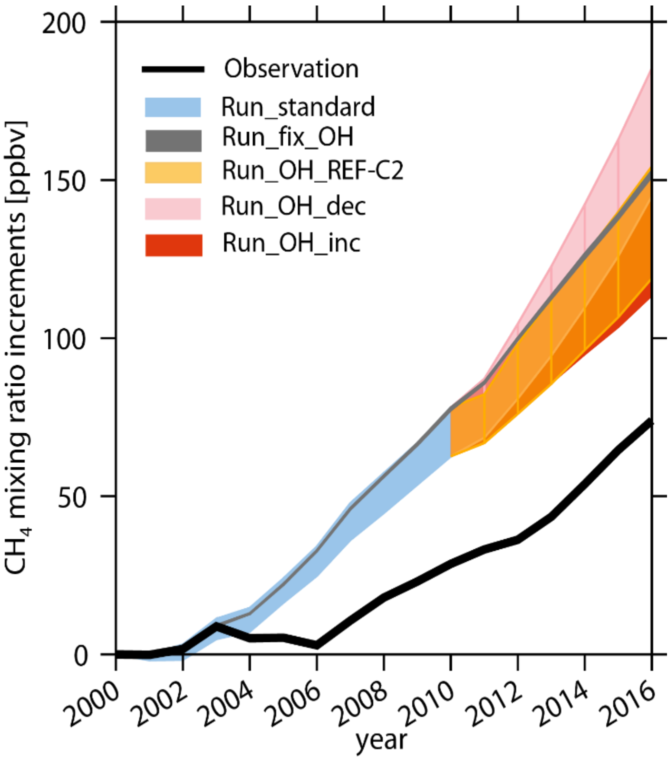

Figure 8. Time series of surface CH_4 mixing ratio increments compared to 2000 for NOAA observations (black line) and model ranges from all the LMDz experiments collected at observation sites (shades) and described in the text and in Table 2.