

**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 CH<sub>4</sub> using the LMDz chemical transport model. The manuscript contains novel investigation to reveal inter-annual variations in OH and its impact on CH<sub>4</sub> 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 account 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<sup>-1</sup> (Run\_OH\_inc) according to the slightly changing of OH calculated by ACCMIP models and one with [OH] decrease of -1% yr<sup>-1</sup> (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 O<sub>3</sub> and CO biases and unaccounted processes in some of the CCMI models, as reported by previous studies. Why is not the model performance on O<sub>3</sub> 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<sub>3</sub> 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 O<sub>3</sub> burden? The tropospheric O<sub>3</sub> burden should also affect primary production of OH.

Response: We have calculated global mean O<sub>3</sub> mixing ratios averaged over the tropospheric and their pressure altitude levels in table 5 and move the values of O<sup>(1)D</sup> and reactive humidity, which contribute less to the inter-model difference of [OH] to the supplement (Table S4).

	CO ppbv				NO pptv				O <sub>3</sub> 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<sub>3</sub> mixing ratios in table 5 as well as O<sup>(1)D</sup> photolysis rates and specific humidity in Table S4.

“Tropospheric O<sub>3</sub> can also influence primary production of OH, and tropospheric

**O<sub>3</sub> burden reflects combined effects of NO<sub>x</sub>, CO, and VOCs. The high O<sub>3</sub> over the lower troposphere simulated by SOCOL3 and the low O<sub>3</sub> 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 lightning NO production affect OH vertical distributions?

**Response:**

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

**Table S3. Lightning NO<sub>x</sub> emission (Tg N yr<sup>-1</sup>) 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:”**

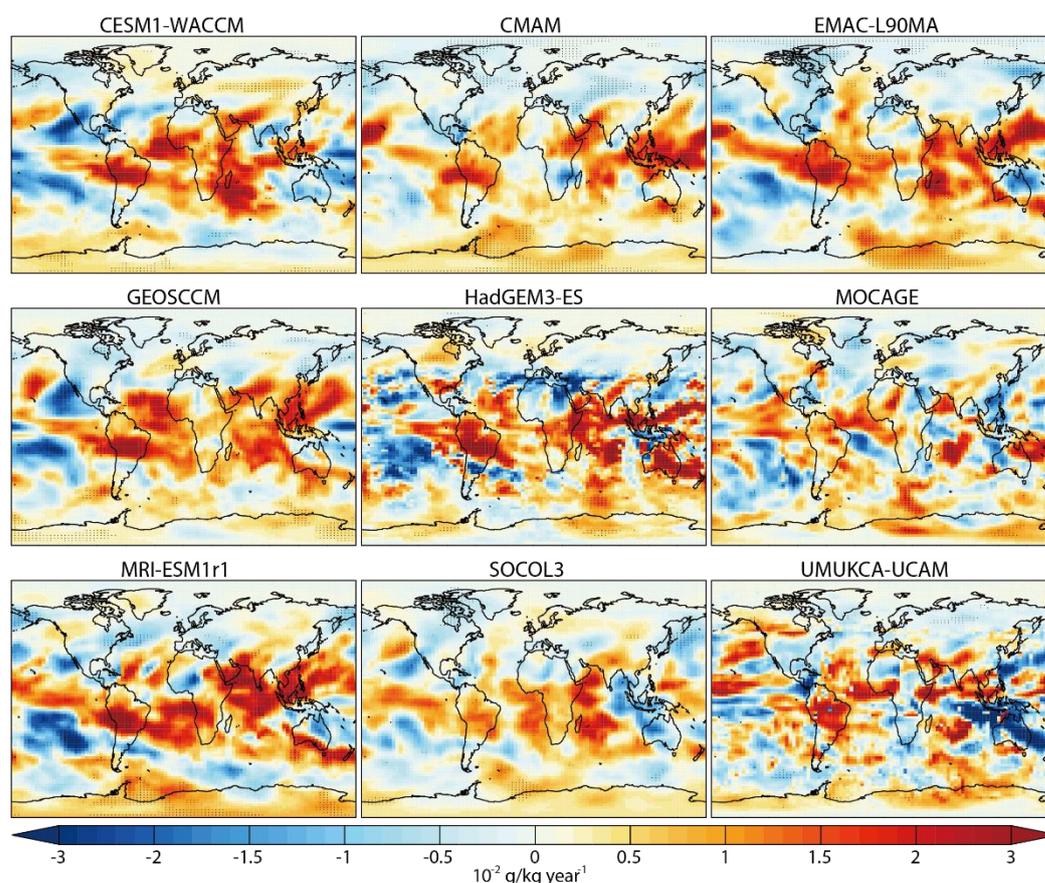
**L431-L435: “Lightning NO<sub>x</sub> emissions, which are mainly emitted in the middle and upper troposphere, can contribute to inter-model differences in NO and OH distributions (Murray et al., 2013; 2014). We compare lightning NO<sub>x</sub> emissions calculated by CCMi models in Table S3.. High lightning NO<sub>x</sub> 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 lightning NO<sub>x</sub> emissions in these models.”**

**L451: Lightning NO<sub>x</sub> emissions range from 3.7-10.2 Tg yr<sup>-1</sup>(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<sub>x</sub> and CO emissions (Fig. S6b), and stratospheric O<sub>3</sub> (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<sup>-1</sup>). Black dots denote model grid-cells with statistically significant trends (p-value < 0.05).

**Comments:** 4.2.1. Spatial distributions of tropospheric CH<sub>4</sub> mixing ratio Could you explain how inter-model differences in spatial and temporal OH variations affect the simulated global CH<sub>4</sub> mixing ratio more in depth?

**Response: In our paper, we attribute differences in LMDz simulated global mean CH<sub>4</sub> mixing ratio to different global OH mean value and trend, and the spatial**

distribution of CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> mixing ratios (Table 7 and Fig. 6). OH fields with increasing trend will lead to lower LMDz simulated CH<sub>4</sub> mixing ratios. The LMDz simulation using the TransCom OH fields (without inter-annual variability) shows the highest CH<sub>4</sub> mixing ratios (1735 ppbv), while the one using the CMAM OH (with slightly increasing OH trend during the decade) shows the lowest CH<sub>4</sub> mixing ratios (1727 ppbv). ”

And we add in the second paragraph:” The differences in spatial distribution of OH fields can influence LMDz simulated CH<sub>4</sub> 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.