

Interactive comment on “Preindustrial to present day changes in tropospheric hydroxyl radical and methane lifetime from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP)” by V. Naik et al.

Anonymous Referee #2

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The authors report a comparison between a number of global models used to investigate the changes in OH radical concentrations and methane lifetimes between 1850 and 2000, and their causes. The topic is certainly within the scope of ACP and the manuscript is well written. However, I have several comments below which should be addressed before publication in ACP.

Major comment

One of the major findings of the work is that there are considerable differences be-

C11916

tween models in both the sign and the magnitude of changes in OH concentration and methane lifetime in the period 1850-2000. In several instances it is stated that these differences arise from the ‘unique ways in which the chemical and physical drivers of OH interact within each model’. Given the aims of the paper, there needs to be a more in-depth discussion of these drivers and an attempt to establish which processes are the dominant factors in determining the reported changes.

Minor comments

Pg 30759, line 21: Please consider changing ‘O(1D) radicals’ to ‘O(1D) atoms’.

Pg 30759, line 22: Please state that the reaction is with water vapor.

Pg 30759, line 23: The production rates of OH are highest in the tropical lower to middle troposphere for the reasons stated, it does not necessarily follow that the concentrations will be highest in this region.

Pg 30759, line 26: Please amend to ‘OH has a tropospheric lifetime on the order of seconds’.

Pg 30760, lines 7-9: It should be stated that OH rapidly cycles with the HO₂ radical, and that H₂O₂ is formed from HO₂ and not OH.

Pg 30760, line 10: It should be made clear that the sources of OH in unpolluted forested low NO_x environments are still uncertain. There are several papers now published which demonstrate this uncertainty and indicate that the mechanisms proposed by Lelieveld et al. (2008) and Peeters et al. (2009) cannot fully explain the observations.

Pg 30761: It should be noted that relatively long-term observations of OH do exist (Rohrer & Berresheim, 2006). There is also little mention in the manuscript of comparisons between observations of OH and model calculations which provide insight to the key drivers of OH concentrations.

Pg 30763: A table summarising some of the key characteristics of the model would be

C11917

helpful here in addition to the references provided.

Pg 39764, lines 3-5: What is the value for k_{OH+CH_4} for those models not using the recommendation by Sander et al. (2011)? What is the range in the different values used between models and how much is this likely to influence the modelled OH and CH₄ concentrations?

Pg 30765: The authors state that the methane lifetime calculated using different definitions of the tropopause height vary by less than 3 %, and thus that the definition of the tropopause height has minimal effect on the methane lifetime. However, the paper concludes that in the period 1980-2000 the methane lifetime decreases by ~ 4 %. Are the 3 % changes in methane lifetime using the different definitions of tropopause height truly insignificant to the conclusions?

Pg 30766, line 10: Increased NO_x concentrations increase the rate of cycling between OH and HO₂. It is not strictly a source of OH since it is most likely that the HO₂ will have been produced via OH in the first instance.

Pg 30766: The classification of models into two groups requires further discussion. Why do the models behave differently?

Pg 30767, line 18: There should be some further discussion here of OH observations in the atmosphere. Aircraft measurements in particular offer information regarding OH concentrations over large spatial and temporal domains.

Pg 30771, line 28: Why does the HadGEM2 model show no change in OH?

Pg 30772, line 18: There are a number of other papers which have attempted to assess the validity of tropospheric chemistry schemes used in models which ought to be referenced.

Pg 30776, line 14 and pg 30778 line 6: There are a number of other important papers which address the response of OH to biogenic NMVOCs which ought to be discussed/referenced.

C11918

References

Rohrer, F. and Berresheim, H.: Strong correlation between levels of tropospheric hydroxyl radicals and solar ultraviolet radiation, *Nature*, 442, 7099, 184-187, 2006

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 12, 30755, 2012.

C11919