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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Analysis of present day and future OH and methane lifetime in the ACCMIP simulations" *by* A. Voulgarakis et al.

Anonymous Referee #1

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This study presents a multi-model analysis of OH and methane lifetime in the current atmosphere and also analyses the calculated changes in the future, using RCP scenarios. The paper is lengthy and although some interesting results can be extracted from the paper, these tend to be blurred in a wide range of analyses that are presented in the paper. The wide spread in the lifetimes calculated by the participating models is worrisome and warrant a detailed analysis before future OH and methane lifetime can be studied. Some of the analysis is presented in the paper (section 6), and this analysis shows that even on the most fundamental level (e.g. ozone photolysis) model differences are much larger than can be expected from physical reasoning. A first step should therefore be a thorough analysis of the model-to-model differences. Of course, this also includes scrutinizing the rates of OH formation and OH destruction, because these terms ultimately determinate the OH abundance in models. Instead the paper

continues with an analysis of the OH changes towards the future. Here another main problem arises: stratospheric ozone recovery is expected to feed back on tropospheric chemistry through reduced photolysis rates. Not all models properly account for this feedback. Although this fact is regularly repeated in the article, these models are still an integral part of the analysis, which makes the paper unbalanced. The same holds for the way NMVOC emissions are treated in the models. Some models emit CO as a proxy, other models take into account the effect of climate change in the emissions. In the end the study compares apples with oranges.

Still there is interesting information in the paper. But then the paper should be stripped to the main issues, and models that exhibit strange behavior should be excluded from the analysis. I acknowledge that this is not in line with the strategy taken for most inter comparisons. But we need some convergence towards better understanding of OH and the methane lifetime.

1 Main Comments

The analysis is difficult because the sensitivities are not uniform across the models. This makes it awkward to read at some places. On page 22956, line 26 I read: " The evolution of the methane lifetime between different time slices shows some agreement between different models, in terms of sign". Further in the manuscript: "There is generally a tendency for methane lifetime decreases". I understand that it is difficult to write a paper with multiple models. But at some point a more in depth analysis is required. For instance, some models are outliers. What is the underlying reason for this? What emission changes were responsible? Without such an in-depth analysis it seems we did not converge since the TAR report, but added complexity in terms of future climate change. Now there is a lot of speculation over possible causes of the 2000-2010 changes in OH. I mention a few (i) stratospheric ozone recovery (page 22959, page 22965) (ii) NOx

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emission changes (various places) (iii) NMVOC emissions (page 22961, line 9). However, a real in depth analysis is often not possible, because not all models submitted all the relevant information. For instance, it would be very interesting to see changes in the photolysis rates due to ozone recovery. But some models did not even include the feedback between stratospheric ozone and tropospheric photolysis! Models treat natural NMVOC in different ways, and only two models take climate-sensitive isoprene emissions into account. To unravel the possible effects, often sensitivity simulations are done with one model. Here, the GISS-E2-R model is used. The results are in itself interesting, but the paper becomes a mess, not being a model inter comparison anymore (section 5.2, figure 8 and 9).

My recommendation is therefore to revise the manuscript (major revision), discussing only some main points. One of the points is surely that climate warming leads to a methane lifetime reduction according to most models.

Also, it would be good to reduce the amount of models for some analysis. For instance, it does not make sense to discuss the HadGEM2 and UM-CAM results in detail, since they miss the coupling between stratospheric ozone and tropospheric photolysis. It is very annoying to read every time that these models behave differently, because they did not take this coupling into account (e.g. page 22972, bottom).

The interpretation of Table 5 is difficult. What I understand is that the emissions, temperatures, and other physical parameters of the models are correlated against their OH abundance. This is really strange for an inter comparison, where you hope that the driver datasets such as emissions are more or less the same for all models. So, what does a significant correlation mean here? That the spread in the driver is large (NMVOC emissions show a large diversity in the models)? Nevertheless, it addresses an important question I have after reading the paper: Why are the models so different? But this analysis should be moved to the front (at least for the present day). Ideally, it should also consider other differences, like OH production and loss fluxes, but these are probably not available. What is really striking in figure 10b us the large

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range in J(O^1D) values, from 0.5 to 2 s^{-1}. Since the author uses GISS-E2-R, this calls for a more in-depth analysis of that model. An explanation as given in the paper: "which means that the photolysis effect is masked by the consumption of OH radicals by CO" is by far insufficient. Also the discussion of the OH NMVOC emission relation is confusing. There is a weak positive relation between the two, with no mechanistic explanation. Still the authors claim: "The model with the highest abundance in OH is MOCAGE, which is likely explained by the fact that its NMVOC emissions are the highest of all models". More analysis is clearly needed here also. On page 22972 it is stated, for instance, that NMVOC and CO emissions can not be separated easily, because some models emit CO as proxy for NMVOC. As stated before, some analysis of the sources and sinks of OH is clearly needed.

I find it weird that the GISS-E2-R simulations use free running methane. This sounds like a failure in the inter comparison set-up (page 22963, top). How large are the differences? From the manuscript it is not totally clear how methane is constrained in the models. If models are forced at the surface, similar burdens should appear? Since the main author can control GISS-E2-R, it would be logical to do a simulation with a similar methane boundary condition as other models.

2 Minor Comments

- Page 22948, line 9: "methane cycling" should be "methane oxidation"
- Page 22948, line 20: "combines" should be replaced by "may combine" since most of the oxygen radicals are quenched
- Page 22948, line 24: water vapor abundances are determined by temperature, and not by temperature changes. "related underlying processes" is awkward. Please be specific what you mean here

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- Page 22949, line 1: "reflections" should be "reflection"
- Page 22950, line 19: please check referencing: Zeng and Pyle should be Zeng et al.. This was the first I checked!
- Page 22955, line 5: Table 2, S1, S2, S3: changes are presented in %. is this based on the temperature in K or in C? Or is the temperature change itself given? Later in the manuscript it becomes clear, but please mention this in a footnote
- Page 22955, line 26: According to Table 1 the tropopause is defined as 150 ppb ozone, or following a cos(lat) dependence. Why this 200 hPa?
- Page 22956, line 19: Reference in preparation is not acceptable I guess?
- Page 22958, line 9:I do not understand. I see that the NH/SH ratio increases in the future (fig. 3b). This is in contradiction with what is said: "reducing the fraction of global OH that exists in the NH"
- Page 22958, line 21:Caption should also mention that a multi-model mean is presented in figure 4
- Page 22960, line 4: "reducing OH through higher water vapour" sounds wrong to me
- Page 22964, line 7: "which implies ...loss". This is not true in general. A non-linear relation does not automatically lead to a large interdependency
- Page 22965, line 4: I think the ozone recovery being faster for higher CO2 only holds for the tropical stratosphere. Be less general here.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 22945, 2012.