Comments and responses on "Trends in global tropospheric hydroxyl radical and methane lifetime since 1850 from AerChemMIP" by David Stevenson et al.

We would like to thank the two anonymous referees for their useful and supportive comments. Their comments are repeated below with our responses in red.

Anonymous Referee #1

This paper analyses the OH trend and methane budget in the period 1850-2014. An important conclusion is that global OH was stable in 1850-1980, after which all three models show an increase of roughly 10%. The analysis convincingly shows that emission changes in Near-Term Climate Forcers (NOx & CO) are responsible for this behaviour.

The manuscript is relatively well prepared, but some improvements are needed, e.g. to the figures, referencing, and discussion.

Throughout the manuscript authors use "concentration", while I think in practical calculations, tables, and plots mole fractions are shown. Better to replace concentration by mole fraction.

We have replaced concentration with mole fraction in all instances except for references to OH, which are concentrations (molecules cm^{-3}).

Concerning the sensitivity simulations: they are sometimes difficult to understand, but I like the calculated impact on the methane mixing ratios.

All in all, the paper is concise and to the point, and clearly demonstrates that from a modelling point of view, OH should be increasing. I miss, however, a thorough discussion on the role of climate change on OH (temperature, natural emissions, lighting NOx.) This is certainly something that needs some more attention, also in light of earlier studies.

We have now added some more information about climate change. We don't have experiments that specifically isolate climate change impacts on OH. However, the difference between the histSST simulations and the sum of (histSST-piCH4, histSST-piNTCF and histSST-1950HC) leaves a residual signal that represents the effects of climate change, together with any non-linear interactions between these drivers (see the new Figure 5 below). Assuming these non-linear interactions are negligible, we find that climate change has increased OH, with a similar magnitude to the emissions drivers, and mainly attribute this to increases in water vapour, although other climate change effects may also be important.

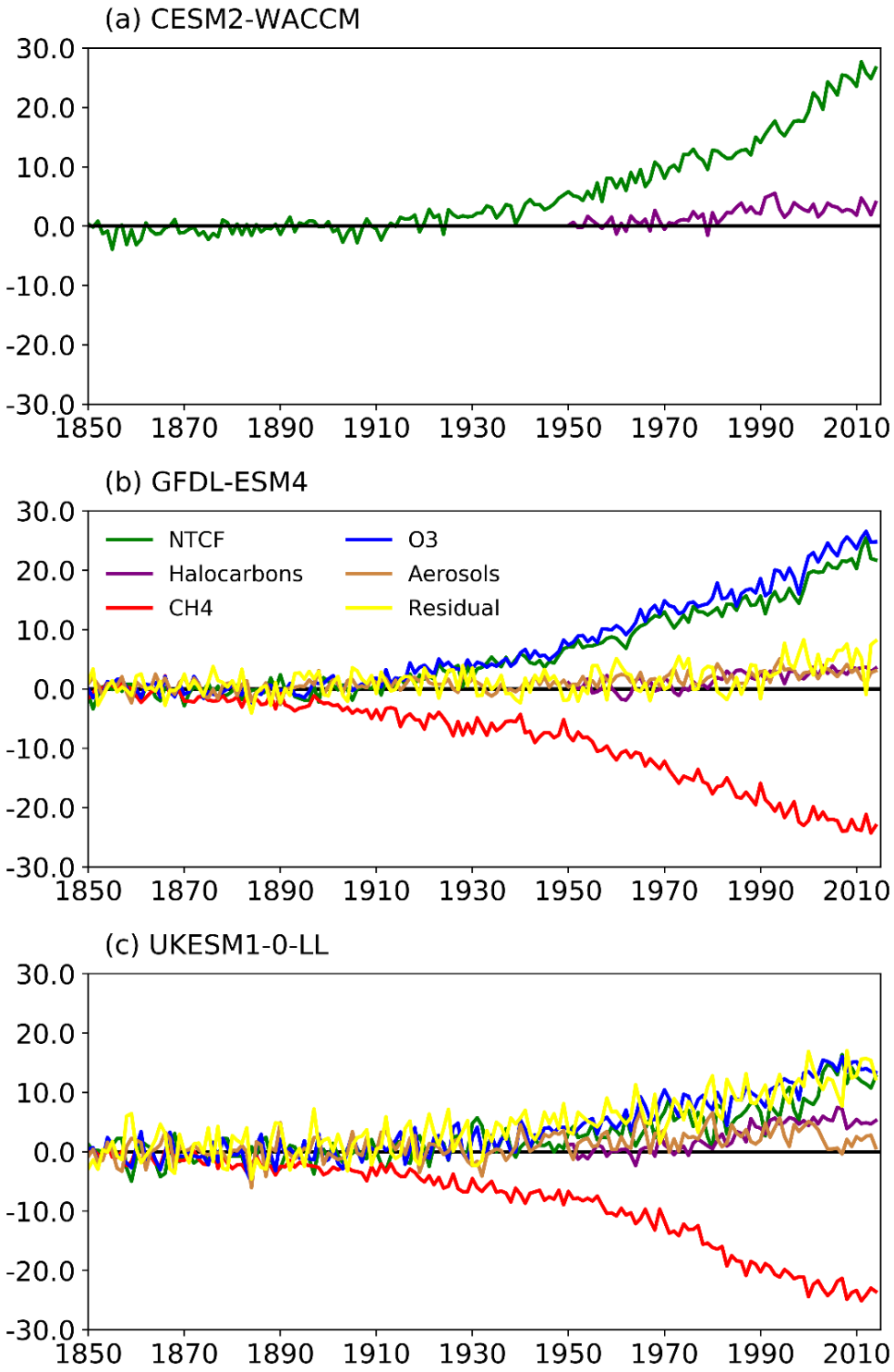

Figure 5 Tropospheric OH anomaly (%) for sensitivity experiments (X), expressed as: (X – histSST)/histSST(1850-1859), for (a) CESM2-WACCM, (b) GFDL-ESM4 and (c) UKESM1-0-LL. 'Aerosols' is the difference between histSST-piO3 and histSST-piNTCF. 'Residual' is the difference between the sum of histSST-piCH4, histSST-piNTCF and histSST-1950HC minus histSST – and represents the sum of climate change effects and non-linear interactions between forcings. NB for UKESM-0-LL, we used historical-piNTCF as histSST-piNTCF was not available.

What also clearly misses is some validation of the model results. I understand that the individual models are (or will be) published, but to gain some confidence in the results, it would be nice to see how e.g. trends in CO are reproduced.

We will refer to some of the other AerChemMIP studies and other relevant papers.

Minor comments:

R4: wrong. H2O instead of HO2.

Thank you for pointing out this error. We fixed it.

Line 62: a sink –> a dominant sink

Fixed.

Line 67: Wrongly suggests that ozone reacts directly with H2O

Adjusted text to clarify it is $O(^1D)$ that reacts with H_2O.

Line 94: GFDL-ESM4 is later called GFDL-AM4, please be consistent.

We apologise for inconsistencies in naming in the submitted paper. We have revised to consistently use the model names: CESM2-WACCM; GFDL-ESM4; and UKESM1-0-LL, based on the Earth System Grid Federation (ESGF) file names.

Line 187: Referencing: I miss references to some recent satellite assimilation work which is relevant, e.g. https://www.atmos-chem-phys.net/15/8315/2015/acp-15-8315-2015.pdf

Thank-you for this and the other Miyazaki et al. papers. We have included discussion of these papers (see below).

Figure 1: I It would be nice to show also the modelled natural NMVOC emissions and how they changed due to climate change and variability in the different models.

Griffiths et al. (2020) Figure 1 has BVOC emissions. We will update Figure 1 to include natural NMVOC emissions.

Figure 2: inset: why is the GFDL-ESM4 simulation not included?

This was a mistake. It is now included.

Figure 3: The use of the vertical coordinate "model level" is not acceptable.

We have converted Figures 3 and 4 (and the related figures in the Supplementary Material) to now use pressure as the vertical co-ordinate.

Line 214: It would be nice to compare and discuss these new estimates to existing estimates. Methane is forced to observations, so the lifetime may be biased due to model biases.

We will compare CH_4 lifetime estimates to existing estimates and discuss potential biases due to experimental set-up.

Line 251: I do not see why the values of f are unreliable due to changes in halocarbon mole fractions.

Our method for calculating the methane-OH feedback factor, f, differs from the 'standard' method, which would normally use dedicated sensitivity experiments, with a simple +20% perturbation to prescribed methane mole fractions (e.g., Prather et al., 1996, 2001). We use the histSST_piNTCF simulations, which hold NTCFs at pre-industrial levels, but allow methane to increase. These simulations are not ideal, as they also have climate (i.e., temperature, water vapour, clouds, etc.) changing. From the 1950s onwards, these simulations also allow halocarbons to increase. Elsewhere in the paper, we show that increasing halocarbon levels, and in particular the associated stratospheric ozone depletion, has an impact on OH. For the diagnosis of f we need runs that only perturb methane. Hence we think that when halocarbons also change, the values of f should be considered unreliable. Figure 7 suggests that the effect on f is probably quite small; nevertheless we think it is sensible to just use values of f for the time period 1930-1960 to exclude the later time period when halocarbons (and climate) show larger changes.

Line 300: Read papers of Miyazaki et al.

In the revised version we now refer to:

Miyazaki, K., Eskes, H. J., and Sudo, K.: A tropospheric chemistry reanalysis for the years 2005–2012 based on an assimilation of OMI, MLS, TES, and MOPITT satellite data, Atmos. Chem. Phys., 15, 8315–8348, https://doi.org/10.5194/acp-15-8315-2015, 2015

Miyazaki, K., Eskes, H., Sudo, K., Boersma, K. F., Bowman, K., and Kanaya, Y.: Decadal changes in global surface NOx emissions from multi-constituent satellite data assimilation, Atmos. Chem. Phys., 17, 807–837, https://doi.org/10.5194/acp-17-807-2017, 2017.

Miyazaki, K. and Bowman, K.: Evaluation of ACCMIP ozone simulations and ozonesonde sampling biases using a satellite-based multi-constituent chemical reanalysis, Atmos. Chem. Phys., 17, 8285–8312, https://doi.org/10.5194/acp-17-8285-2017, 2017.

Miyazaki et al. (2015, 2017) and Miyazaki and Bowman (2017) showed that assimilation of O_3, CO and NO_2 satellite data into a 3-D Chemistry-Transport Model (CTM) improved the simulated NH/SH ratio of OH from 1.26 to 1.18 (cf. an observed ratio of 0.97 ± 0.12, Patra et al., 2014). These studies clearly show that global OH is sensitive to assimilation of O_3, CO and NO_2 data, due to the strong coupling between the atmospheric chemistry of these species.

Anonymous Referee #2

1 Overview:

Review of "Trends in global tropospheric hydroxyl radical and methane lifetime since 1850 from AerChemMIP" by Stevenson et al.

I apologize for the delay in my review. Stevenson et al. present an analysis of changes in OH abundance and methane lifetime from 1850 to present using simulations from a model intercomparison (CMIP6/AerChemMIP). Specifically, they use output from 3 models: GFDL-ESM4, CESM2-WACCM, and UKESM1. The three models simulate stable OH concentrations prior to 1980 and an increase post 1980. The work then uses a set of sensitivity simulations to diagnose the processes that control the time evolution of OH. Overall, I think the work is both useful and interesting. My main comments relate to the presentation of the interpretation. Specifically, the discussion regarding conflicts with observational MCF constraints and the brevity of the final discussion (there's only half a page of discussion after laying a solid groundwork in the methods). I feel like this could be expanded to make the work more useful to others. I would suggest minor revisions for the work.

2 Comments:

2.1 Discussion of MCF constraints

The authors seem to be arguing that these model-derived forward simulations of OH are more reliable than reconstructions.

It was not our intention to present the results this way, and we don't think the model results are more reliable than the reconstructions. We attempted to present the model results and give the OH reconstructions as context, in order to facilitate comparison. We now include uncertainties in the reconstructions from Rigby et al. (2017), which help clarify this comparison.

I'd be wary of framing it this way as this paper has ZERO observational constraints.

This is not quite true – global mean surface methane concentrations are prescribed to evolve following observed levels. Hence the calculated OH values in our paper are consistent with the evolution of observed global mean methane.

On their face, the results differ from observationally constrained OH estimates and this is the interpretation from the authors (Line 3 in the abstract); however, I'm not convinced they really differ. If the authors were to include the uncertainty estimates from the Rigby et al. (2017) paper, for example, they would likely find that it bounds their results (the uncertainties are included in the supplemental data from the Rigby paper). So I think some of the "disagreement" they see is within the uncertainties.

We broadly agree with this (see details below).

Additionally, the OH changes here do seem to agree quite well with the results from Turner et al. (2017) up until 2005. One could argue there is a divergence post-2005, but the authors don't really seem to discuss this at all.
The authors seem to argue that the entire post-1980 rise differs from the MCF-derived estimates. This is curious to me.
I feel that line 3 of the abstract ("The model-derived OH trend since 1980 differs from trends found in several studies that infer OH from inversions of methyl chloroform measurements; however, these inversions are poorly constrained and contain large uncertainties that do not rule out the possibility of recent positive OH trends.") and some of the main text discussion of the MCF reconstructions needs to be rephrased.

See below – we now have included the uncertainty estimates from Rigby et al. (2017) into a revised Figure 2 and will adjust the text accordingly. We agree with the reviewer that the AerChemMIP modelled OH trends are (just about) within the uncertainty range derived by Rigby et al. (2017).

The authors seem to have missed two important papers from Joe McNorton as well:
McNorton et al. (2016; https://doi.org/10.5194/acp-16-7943-2016) and McNorton et al. (2018; https://doi.org/10.5194/acp-18-18149-2018).
There are two other recent papers that should also be referenced and briefly discussed:
He et al. (2020; https://doi.org/10.5194/acp-20-805-2020) and Nguyen et al. (2020; https://doi.org/10.1029/2019GL085706). He et al. (2020) also used the GFDL model to simulate methane from 1980 to present and find a similar time evolution of OH.
Nguyen et al. (2020) look at the impact of chemical cycling on methane and OH.

These papers are all very relevant and we will incorporate discussion of them into the revised text. McNorton et al. (2016) performed inverse modelling using a 3-D Chemistry-Transport Model (CTM) constrained by MCF data, and found that OH increases contributed significantly to the slowdown in the CH_4 growth rate between 1999 and 2006, and that the post-2007 increases in CH_4 growth rate were poorly simulated if OH variations were ignored. McNorton et al. (2018) extended this work with further constraints from GOSAT CH_4 and $\delta^{13}CH_4$ and found that the post-2007 CH_4 growth rate surge was most likely due to a combination of a decrease (-1.8 ± 0.4 %) in global OH and increases in CH_4 emissions, although an alternative inversion that assumed fixed OH indicated slightly larger increases in CH_4 emissions. He et al. (2020) used the GFDL-AM4 model, which is the atmospheric component of the GFDL-ESM4 used in this study, and found an upward trend in global OH since 1980 similar in magnitude to our results. Like Gaubert et al. (2017), Nguyen et al. (2020) found that decreasing global CO

concentrations since the 2000s have important influences on CH_4 flux inversion results, because of the strong chemical coupling between CO, CH_4 and OH.

Collectively, all these earlier studies that have attempted to interpret the observed trends in methane and related species find that OH is sensitive to CO, NO_2, O_3, as well as CH_4. These studies have spanned box models to sophisticated 3-D CTMs, and all appear to be under-constrained in deriving trends in OH. To date, studies have used subsets of the available observational data (e.g., one or more of MCF, CH_4, $\delta^{13}CH_4$, CO, ^{14}CO, NO_2, and O_3), but not yet all available relevant data. The OH trends presented in this study are from state-of-the-art Earth System Models driven by up-to-date emissions estimates from CMIP6, and are consistent with observed trends in CH_4, however other species (e.g., CO, O_3 and NO_2) are allowed to freely evolve. It is unclear if the OH trends simulated by these CMIP6 models are realistic, however, it is clear that the way these models simulate OH is very important for projecting future trends (and understanding past trends) in CH_4.

2.2 Processes controlling the OH changes

It would be nice if the authors had one additional schematic type figure that summarizes their findings. There are quite a few acronyms and competing effects that make it confusing at times. Naik et al. (2013) paper had some nice bar charts showing the relative contribution of different factors to the PI-PD OH changes. This really helped follow the argument and understand what the different scenarios are doing. It seems like this would be particularly helpful to the casual reader.

As it stands, Figures 5 and 6 are the ones that diagnose the processes controlling the long-term OH changes in the model. But I can imagine many readers having a difficult time figuring out what they are supposed to take away from those figures. As it stands, they are an acronym soup.

Personally, I feel that the manuscript would greatly benefit from a final synthesis figure that summarizes the changes described in the abstract and a few additional paragraphs in the discussion section describing this.

We have constructed a new figure (Figure 8) that summarizes the drivers of OH changes – this also includes a residual term, that we think mainly reflects climate change effects:

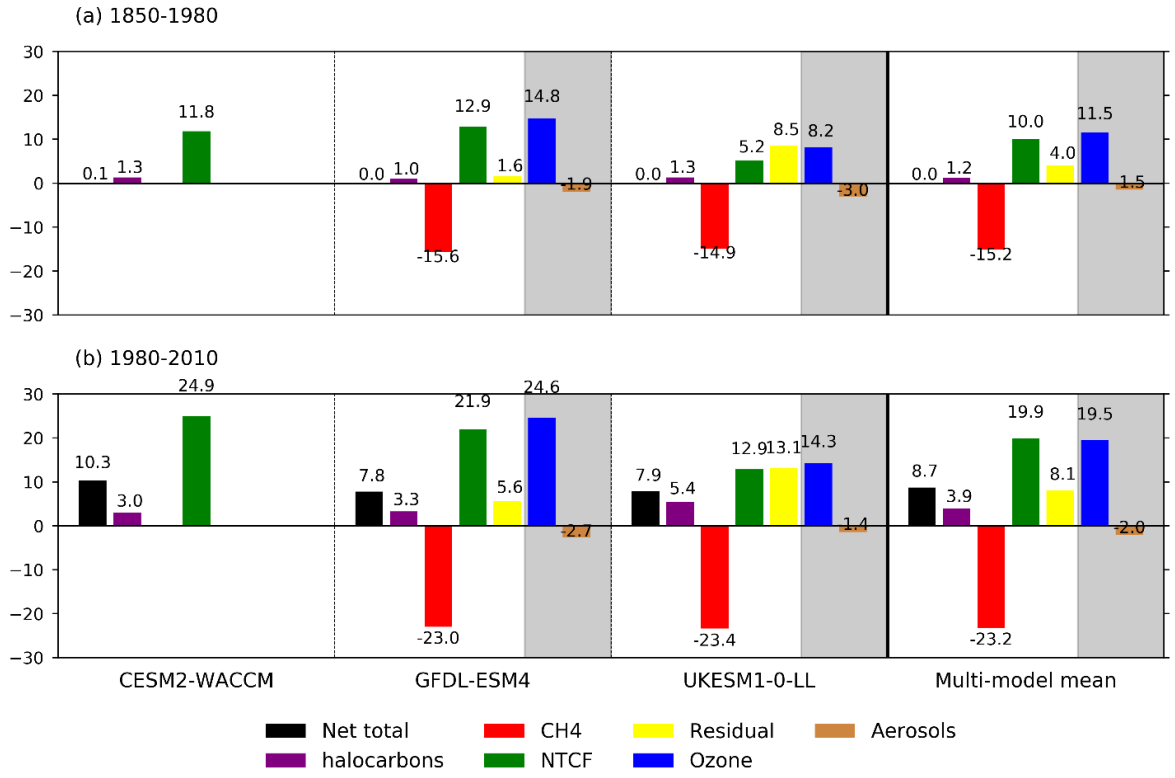

Figure 8. Summary of drivers of OH changes (%), relative to 1850, for the three models and their multi-model mean over: (a) 1850-1980; and (b) 1980-2010. (NB we have used decadal means: 1850 refers to (1850-1859); 1980 is (1975-1984); and 2010 is (2005-2014). The shaded areas show the split of the NTCF signal (green) into ozone precursors (blue) and aerosols (brown), where models have performed both the histSST-piNTCF and histSST-piO3 experiments. The residual values (yellow) are the differences between the total change (black, from the histSST simulations) and the sum of the changes from CH4 (red), NTCF, and halocarbons (purple). We interpret the residual terms as being due to climate change, in addition to any non-linear interactions between forcings.

3 Specific comments:

Lines 180–185 (Inserted the relevant lines from the discussion paper here)

"Naus et al. (2019) further investigated the inversion methods used by Rigby et al. (2017) and Turner et al. (2017), confirming that the derivation of OH from MCF and CH4 is a strongly under-constrained problem, and found that estimated OH trends with a range of different magnitudes and signs are equally valid solutions from the available data."

and 280–283

"Naus et al. (2019) found that the uncertainties inherent in inversion of MCF and other proxy measures of OH are sufficiently large that OH trends derived from them are less constrained than previously thought, and that positive recent OH trends are compatible with the MCF measurements."

: I'm confused here, I thought the Rigby et al. (2017) and Turner et al. (2017) paper showed that the problem was under-constrained. If I recall, the Turner paper showed they could fit the data without changing OH and that there were a number of valid solutions. It's not clear what the Naus et al. (2019) paper added?

The Naus et al. (2019) study nicely illustrates the uncertainties discussed and presented in Rigby et al. (2017). We retain discussion of Naus et al. (2019) in the revised paper, whilst acknowledging that Rigby et al. (2017) quantified uncertainties earlier.

Lines 198–200

"The published inferred trends from different inversion methods show a range of different trends, but there is little resemblance to the upwards trends simulated by the models over this time period."

and 277–280

"The strong recent increase is at odds with several studies that use MCF and other proxies to reconstruct OH trends (e.g., Figure 2 inset); however, these show a wide range of trends."

This is the discussion that I would disagree with. The model results don't seem that different from the model results (especially if you include error bars from Rigby). You might be able to argue differences post-2005, but 1980-2005 seem be in pretty good agreement. The He et al. (2020) paper also look at this.

We will revise this discussion, based on a new version of Figure 2 (below), incorporating uncertainties from Rigby et al (2017).

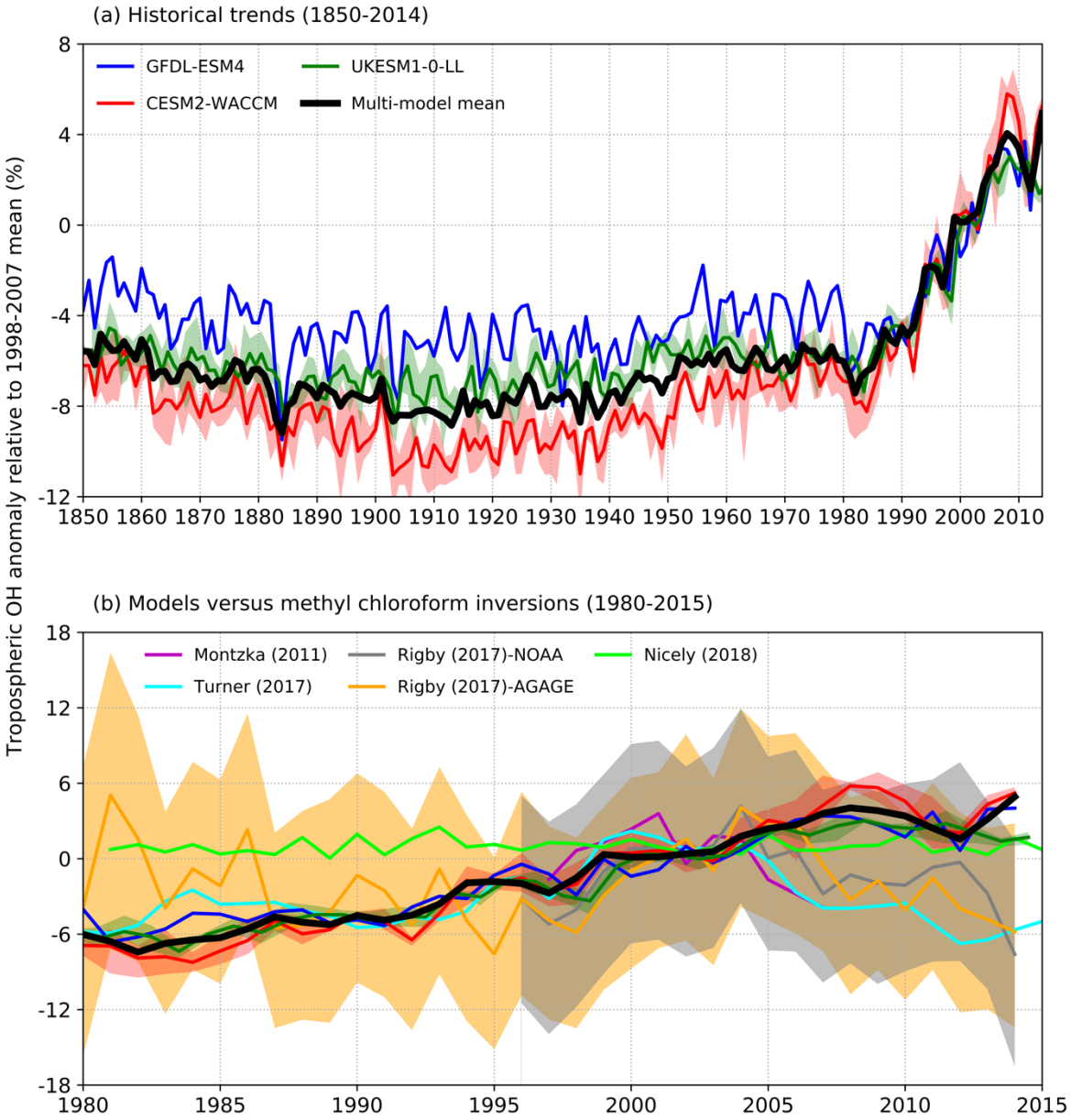

Figure 2 (a) Time evolution of global annual mean tropospheric OH (1850-2014), expressed as a percentage anomaly relative to the 1998-2007 mean (and ensemble spreads, where available) for GFDL-ESM4 (blue), UKESM1-0-LL (green), and CESM2-WACCM (red), and the multi-model mean (black). (b) Observation-based inversions of global annual mean tropospheric OH for 1980-2015, including ±1 standard deviation uncertainties for the results from Rigby et al. (2017), with model results from panel (a) overlain.

The new Figure 2 shows that the AerChemMIP modelled trends are (just about) within the uncertainties of the observation-based estimates of OH. So as the reviewer notes, they are consistent. The model trends do however go from being at the lower end of the uncertainty range in 1980 to the upper edge of the range in 2014. We will adjust the text and discussion accordingly.