

Reviewer 1:

This paper investigates the sensitivity of the global OH and its trends simulated by a chemistry transport model to processes including water vapor UV absorption, heterogeneous reactions, the OH + NO₂ reaction rate, and oceanic CH₃CHO emissions. The model is validated against observations of multiple species. Understanding the drivers of OH concentrations and changes is an important topic. An overestimate of global mean OH is a common feature of global models, so quantifying the impact of these processes on the simulated OH is of strong interest. The methods are generally robust, but some clarifications of the methodology and conclusions are needed, as described in the comments below.

Response: We thank the reviewer for the constructive comments and positive assessment of our study. We have carefully considered all comments and revised the manuscript accordingly. Below, we provide a point-by-point response, with our responses shown in bold black text. All page and line numbers referring to the revisions correspond to the revised manuscript with tracked changes.

General Comments:

1. The introduction provides a nice overview of the recent developments related to modeling processes relevant to OH. However, it would benefit from a clearer articulation of what the open questions related to these processes are that this paper seeks to address. The conclusions could be strengthened by putting the results in the context of the open questions. In particular, the conclusions give a summary of how each of the individual processes impact OH levels in the model, but I think the paper would have more impact if it can tie that information back to the motivating issues raised in the introduction. For example, would including all these processes be expected to resolve all (or a fraction) of the OH bias seen in model intercomparisons?

Response: We thank the reviewer for this helpful suggestion and agree that the open questions motivating the study and their connection to the broader implications of our results should be stated more clearly. A key unresolved issue in global atmospheric chemistry modelling is the tendency of models to simulate relatively high tropospheric OH and, consequently, methane lifetimes that are shorter than observationally constrained estimates. We have now made this broader motivating issue more explicit in the Introduction (Pages 1–2, Lines 58–66), where we discuss the high-OH/short-methane-lifetime bias identified in previous model intercomparisons and highlight the need to better understand the processes contributing to this discrepancy.

Following the discussion of water vapour UV absorption, heterogeneous chemistry, OH + NO₂ reaction kinetics, and oceanic CH₃CHO emissions, we added the following text to clarify why these processes are assessed together (Page 3, Lines 99–101):

“Taken together, these recent findings motivate a systematic assessment of their impacts within a common modelling framework, allowing their effects on OH and methane lifetime

to be quantified consistently and their potential implications for improving global OH simulations to be evaluated.”

The objectives at the end of the Introduction have also been revised to emphasize the potential importance of these processes for reducing uncertainties and biases in simulated tropospheric oxidative capacity, while the trend analysis examines the roles of emissions and meteorology in the temporal evolution of OH (Page 4, Lines 121–126).

Following the reviewer’s suggestion, we have also strengthened the Conclusions by relating the sensitivity results to the broader motivation of the study, while emphasizing that the sensitivities may be model dependent and are not necessarily additive. The following statement has been added (Page 31, Lines 762–771):

“Together, these results suggest that improved representation of the processes examined here can help reduce the tendency of global models to overestimate OH and underestimate methane lifetimes, although these processes alone cannot fully account for this bias. The model configuration used here does not include halogen chemistry, which can influence tropospheric OH and may provide an additional pathway for reducing this bias. In this study, the large sensitivity to the OH + NO₂ reaction rate highlights the need to better constrain its kinetics, while the effects of oceanic acetaldehyde emphasize the need for improved constraints on marine emissions. For H₂O UV absorption and heterogeneous chemistry, more consistent implementation of current process understanding across global models would help reduce structural differences in simulated OH. Overall, improving OH representation requires both tighter constraints on uncertain kinetics and emissions and broader incorporation of currently understood processes into global models.”

2. The introduction motivates the study in part by highlighting the tendency of global models to overestimate global OH and underestimate methane lifetime. However, the manuscript also states on line 608 that “Airmass-weighted OH concentrations and methane lifetimes in this study are fully consistent with observational and multi-model estimates”, which suggests this bias is not present in the model used in this study. Is it the incorporation of the new processes/rates that allows the model to be consistent with observational estimates? If so, please clarify this.

Response: We thank the reviewer for raising this important point. During the development of the current model configuration, we progressively incorporated several of the processes examined in this study. Earlier evaluations of the model, before these updates were introduced, showed a substantial positive bias in simulated OH relative to ATom observations. As illustrated in Figure R1, the original model configuration (SM_STD) generally overestimated observed OH, whereas inclusion of water vapour UV absorption (SM_WVA), one of the subsequent updates, reduced this bias. The current reference configuration further incorporates heterogeneous chemistry and the updated OH + NO₂ reaction kinetics considered in this study. These developments suggest that improved

representation of these processes has contributed to the better agreement of simulated OH with observational constraints in the current model.

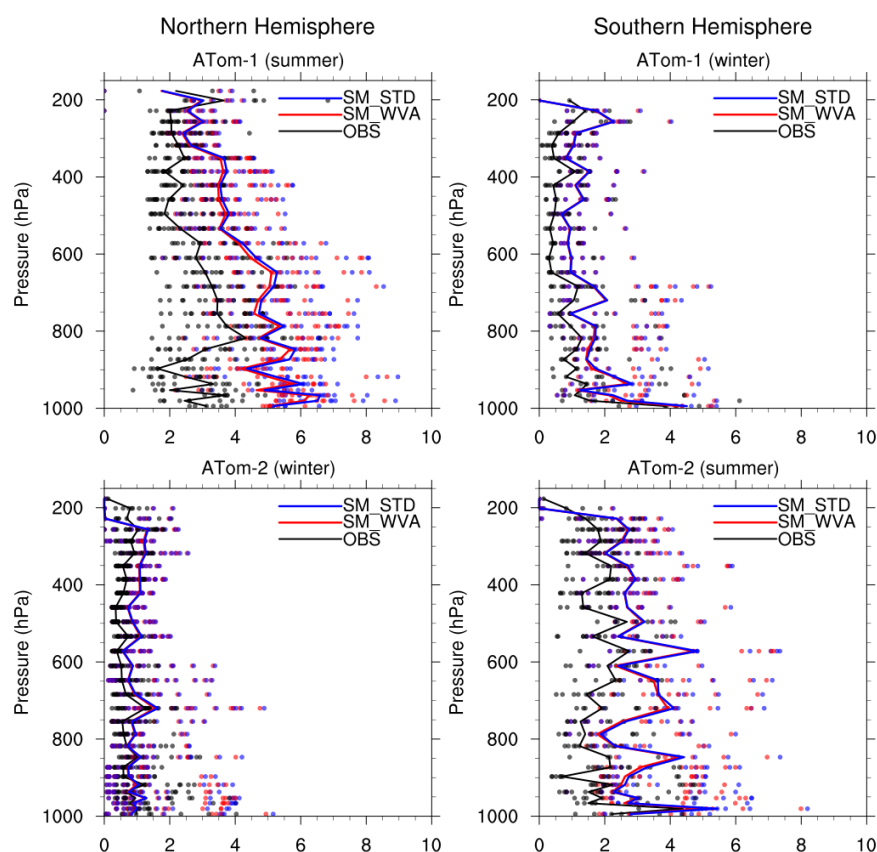
We note, however, that Figure R1 represents an evaluation performed during an earlier stage of model development. We therefore avoid attributing the present-day model–observation agreement quantitatively to individual updates based on this comparison. Instead, the sensitivity experiments presented in this study quantify the effects of the individual processes within the current model framework and demonstrate that several of them act to reduce simulated OH and increase methane lifetime, consistent with reducing the high-OH/low-methane-lifetime tendency commonly reported in global models.

We have therefore revised the statement in the Conclusions (Page 30, Lines 730–731):

“Airmass-weighted OH concentrations and methane lifetimes in this study are within the range of observationally constrained and multi-model estimates.”

We have also clarified the interpretation of the sensitivity experiments in the Conclusions (Page 31, Lines 757):

“Incorporating these processes reduces the simulated OH levels and alters methane lifetime, while having little effect on the OH trend.”



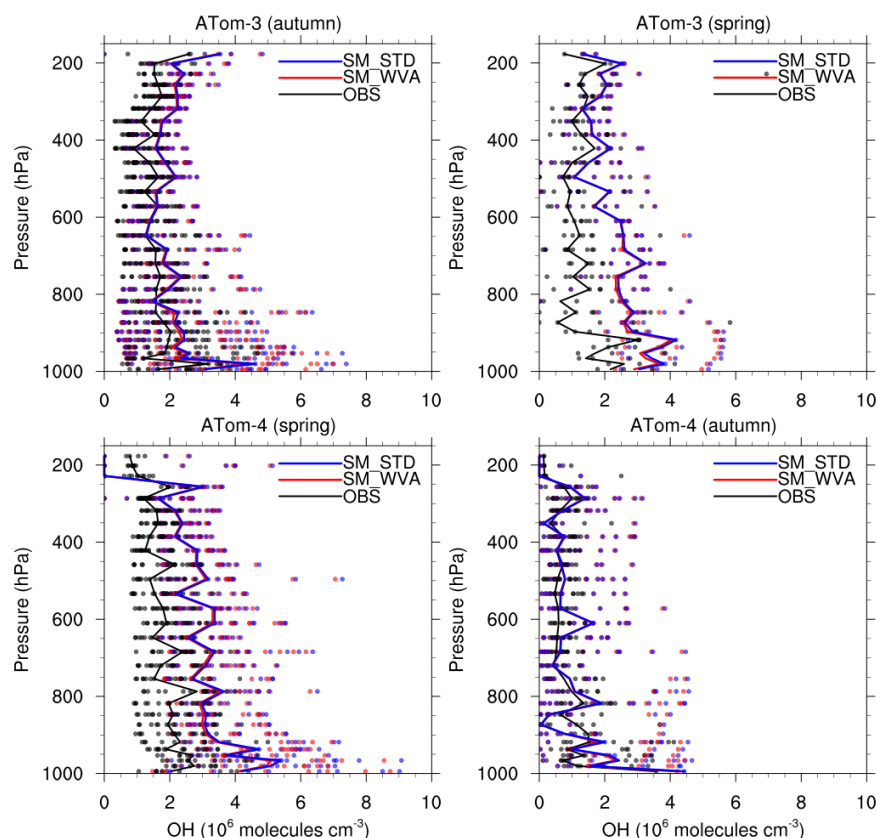


Figure R1. Median OH concentrations ($10^6 \text{ molecules cm}^{-3}$) measured during the ATom aircraft missions (black) and corresponding simulations from an earlier stage of FRSGC/UCI model development. SM_STD represents the model configuration before the process updates considered in this study, while SM_WVA includes water vapour UV absorption following Prather and Zhu (2024). Dashed black lines indicate the observed 25th–75th percentile range.

3. While the control run includes most of the processes considered in the study and the effects of the processes are quantified by removing the process in a sensitivity run, the impact of oceanic CH_3CHO emissions seems to be quantified in the opposite way, e.g. included in a sensitivity run but not the control. A couple sentences clarifying and justifying this distinction would be helpful. See also specific comment 4.

Response: We agree that the different treatment of oceanic CH_3CHO emissions compared with the other processes requires further clarification. Oceanic CH_3CHO emissions were not included in the CTL simulation because their magnitude and spatial distribution remain highly uncertain. This uncertainty is reflected in the substantial difference between the two independent estimates used here, with global annual oceanic CH_3CHO emissions of 40.8 Tg yr^{-1} from CESM2 and 62.7 Tg yr^{-1} from GEOS-Chem. We therefore performed two separate sensitivity simulations, ALD-cesm and ALD-geoc, rather than adopting a single oceanic emission estimate, allowing us to assess both the potential impact of oceanic CH_3CHO emissions on OH and its sensitivity to the prescribed oceanic source.

We have clarified this experimental design and its rationale in the revised manuscript (Page 7, Lines 224–229):

“The impact of processes already represented in the control run, such as water vapor UV absorption and heterogeneous chemistry, are quantified by removing them in the corresponding sensitivity runs. Oceanic CH₃CHO emissions remain highly uncertain, as reflected in the substantial difference between the two emission estimates considered here, and are therefore not included in the control run. We therefore consider two separate sensitivity runs, ALD-cesm and ALD-geoc, to assess the potential impact of oceanic CH₃CHO emissions and the sensitivity of this impact to the prescribed oceanic source.”

Specific Comments:

1. Line 71: I don’t understand the statement “NO_x chemistry extended the model estimate...”. Does it mean the model estimate increased?

Response: We apologize for the unclear wording in the original manuscript. We intended to state that including heterogeneous NO_x chemistry increased the simulated methane lifetime by 6.9% relative to a simulation without heterogeneous chemistry. We have revised the sentence for clarity (Page 3, Lines 78):

“Using the GEOS-Chem CTM with revised uptake coefficients, Holmes et al. (2019) showed that inclusion of heterogeneous NO_x chemistry increased the simulated τ_{CH_4} by 6.9% relative to a model run without heterogeneous chemistry.”

2. Line 167: Why is a climatology used for the aerosols? Since the paper aims to quantify the impact of heterogeneous chemistry on OH trends, it would be preferable to include interannual variability in the aerosols.

Response: We agree that including interannual variability in aerosol abundance and cloud water and ice would provide a more complete assessment of its influence on OH trends. Cloud water and ice are calculated online in the model following Eqs. (3–4), and therefore their interannual variability is represented, allowing cloud-related heterogeneous reaction rates to respond to year-to-year variations in meteorological conditions. However, interannually varying aerosol distributions are not available in the current model configuration. Although aerosol surface area is prescribed as a monthly climatology, the uptake coefficients used for heterogeneous reactions depend on temperature and humidity and therefore vary with the meteorological conditions. Thus, heterogeneous reaction rates can still exhibit interannual variability through their meteorological dependence, although variability associated with changes in aerosol abundance itself is not represented. We have clarified this limitation in the revised manuscript (Page 6, Lines 188–189):

“The monthly climatology captures seasonal variations but does not account for interannual variability, and this may limit our assessment of the effects of aerosol heterogeneous chemistry on OH trends.”

3. Line 182: What lightning parameterization is used?

Response: Lightning NO_x emissions are calculated interactively as a function of convective air mass flux at 440 hPa following the parameterization of Allen and Pickering (2002) and applying the vertical profiles of Pickering et al. (1998). We have added this information to the model description (Page 6-7, Lines 199-200):

“Lightning NO_x emissions are calculated interactively in the model as a function of convective air mass flux at 440 hPa following Allen and Pickering (2002) and Pickering et al. (1998), and vary annually, providing a source of ~5 Tg[N] · yr⁻¹.”

4. Line 192: Related to general comment 2, the text here states that the control run includes all the processes detailed above. However, it later appears that the control run does not include ocean emissions of CH₃CHO. Please clarify.

Response: The original statement was imprecise and could give the impression that oceanic CH₃CHO emissions were also included in CTL. We have revised this sentence to explicitly distinguish the processes included in CTL from the oceanic CH₃CHO sensitivity experiments (Page 7, Lines 209–211):

“The control run (CTL) included the updated physical and chemical processes detailed above, including UV water vapor absorption and heterogeneous chemistry, but did not include oceanic emissions of CH₃CHO”

We have also clarified later in this section that CTL includes biogenic, biomass burning, and anthropogenic CH₃CHO emissions, but no prescribed oceanic source. As explained in our response to the general comment above, this treatment reflects the substantial uncertainty in current estimates of oceanic CH₃CHO emissions, and the rationale for this experimental design is now explicitly described in the revised manuscript.

5. Section 3 contains a lot of general model evaluation. I recommend focusing the discussion on the implications of the evaluation for OH. A summary table might be helpful.

Response: We agree that the model evaluation should be more directly connected to the subsequent analysis of OH. We have therefore revised Section 3 to emphasize the implications of model performance for simulated OH and condensed some of the more general discussion.

For O₃, we now discuss more explicitly how biases in its abundance and temporal evolution may affect primary OH production and simulated OH trends. We also clarify that the generally good representation of the major spatial and temporal characteristics of tropospheric O₃ supports the subsequent OH analysis, while the identified regional biases remain a source of uncertainty (Pages 11–13, Lines 341, 368–369, and 381–383).

For NO₂, we have strengthened the discussion of its connection to OH through HOx recycling and how biases in NO₂ may affect simulated OH. We also emphasize that the generally good agreement between observed and simulated NO₂ trends, particularly over polluted regions, supports the subsequent assessment of the influence of changing NOx emissions on OH trends (Page 14, Lines 392–393 and 405–407).

For CO, we have retained and emphasized the discussion of how the model–observation mismatch in CO trends may affect the simulated temporal evolution of OH over 2000–2017.

We considered the reviewer's suggestion of a summary table. However, because these implications are species-specific and closely linked to the corresponding evaluation results, we have incorporated them directly into the relevant sections rather than adding a separate table. We believe this provides a more direct connection between the model evaluation and its implications for OH while avoiding duplication of information already presented in the text and figures.

6. Line 288: Is this the correlation of the timeseries of annual mean modeled O₃ with OMI/MLS O₃ at each grid box?

Response: Yes. The correlation is calculated between the annual mean modelled and OMI/MLS tropospheric O₃ column time series at each grid box. We have clarified this in the Figure 2 caption.

7. Line 293: Could mismatches between modeled and OMI/MLS tropopause be involved?

Response: Yes, differences between the modelled and OMI/MLS tropopause could also contribute to the lower correlations in these regions. We have added this as an additional source of uncertainty in the revised manuscript (Page 11, Lines 346).

8. Line 294: How is the seasonal cycle involved? I thought the correlations were for the annual mean. Please clarify.

Response: We apologize for the confusion. The reference to the seasonal cycle was a mistake; the correlations are calculated using the annual mean O₃ time series at each grid box and do not involve the seasonal cycle. As part of the revision to condense the general model evaluation in Section 3, this sentence has been removed, and the description of the correlation calculation has been clarified accordingly.

9. Lines 316-318: The text states “Overall, our simulated results represent the distributions and interannual variations of O₃ well”, but Fig. 1e, f shows substantial differences between the modeled and OMI/MLS tropospheric O₃ column trends. This could potentially impact simulated OH trends.

Response: We agree that the regional differences between the modelled and OMI/MLS O₃ trends could affect the simulated OH trends. Accurately reproducing tropospheric O₃ columns and their long-term trends remains challenging because of uncertainties in both model simulations and satellite retrievals, including those associated with tropopause determination. The manuscript also includes evaluations against surface O₃ observations and direct OH and HO₂ observations, providing additional constraints on model performance relevant to OH. We have revised the text to distinguish the relatively good agreement in the spatial distribution and interannual variability of tropospheric O₃ from the larger regional differences in its long-term trends, and to explicitly acknowledge their potential implications for simulated OH trends. The revised text now reads (Page 13, Lines 381–383):

“Overall, the model captures the major spatial characteristics and interannual variability of the tropospheric O₃ column reasonably well, although regional differences are evident in the trends. These differences may contribute to uncertainty in simulated OH trends.”

10. Section 3.1-3.2: It might make sense to either reorder the discussion of section 3.1 to discuss O₃ last, or else move section 3.2 before section 3.1, to put the discussion of satellite O₃ next to the discussion of surface O₃.

Response: We agree that the satellite and surface O₃ evaluations should be discussed consecutively. We have therefore reordered Sections 3.1 and 3.2, with the surface O₃ evaluation now presented in Section 3.1, followed by the satellite evaluation of O₃, NO₂ and CO in Section 3.2 (Page 10-16).

11. Table 3: Some of the differences in [OH]_{gm} between simulations are pretty small. Could you add the range of [OH]_{gm} for individual years or else the standard deviation over all years so that the difference between simulations can be compared to the magnitude of year-to-year variability?

Response: Thank you for the suggestion. We have now added the standard deviation of the annual mean [OH]_{gm} over 2000–2017 to Table 3 to provide a measure of the year-to-year variability. The standard deviations are very similar across the simulations, ranging only from 0.09 to 0.10 × 10⁵ molec·cm⁻³, indicating that the magnitude of interannual variability in [OH]_{gm} is largely unchanged among the sensitivity simulations (Page 19, Table 3).

12. Line 469: Can this result be shown on Fig. 5?

Response: We appreciate the suggestion. However, Figure 5 already presents the A_{OH} observations, their 25th–75th percentiles, and the corresponding control run results for both hemispheres. Adding another run result would make the figure more crowded and potentially more difficult to interpret. We therefore prefer to retain the current presentation of Figure 5.

Figure 7f shows that the faster OH + NO₂ rate coefficient used in the control run reduces OH throughout the troposphere, with a somewhat larger effect in the upper troposphere. Since the control run already shows slightly higher OH than observed in Figure 5, using the slower JPL rate coefficient would further increase simulated OH and thus likely increase this positive bias. We have clarified this point in the revised manuscript (Page 22, Lines 541–543).

13. Line 532 and Fig. 9: Can the color scale of the figure be extended so that it doesn't max out over such a large region of Asia? Also, what explains the location of the OH maximum in the annual mean? Is it related to the terrain?

Response: Thank you for this suggestion. We have revised the colour scales in Fig. 9 to better represent the spatial variability over regions with high OH. Specifically, the range for TCOH has been extended from 0–30 to 0–45, and that for the TCOH trend from -1.5 – 1.5 to -3 – 3. The revised figure now resolves the high TCOH values over East Asia more clearly rather than saturating over a large area.

The annual-mean TCOH maximum over East Asia is primarily associated with the pronounced summertime enhancement rather than being directly caused by terrain. As shown in the revised Fig. 9, TCOH over East Asia reaches approximately 42×10^{12} molecules·cm⁻² in summer, substantially higher than in winter. This enhancement is consistent with stronger summertime photochemical activity associated with higher solar radiation, temperature, and water vapour, together with abundant NO_x emissions that promote OH production. Terrain and associated circulation may influence the detailed spatial distribution, particularly around the Tibetan Plateau, but our results do not indicate that terrain is the primary driver of the broad East Asian maximum.

The TCOH trend also shows its strongest enhancement over East Asia in summer, with local values reaching approximately 5.0×10^{11} molecules cm⁻² yr⁻¹, although the revised colour scale is limited to $\pm 3 \times 10^{11}$ molecules cm⁻² yr⁻¹ to retain sufficient spatial contrast elsewhere.

14. Section 4.4 last paragraph: How do these trend results compare to those of Sourì et al., 2024 (<https://doi.org/10.5194/acp-24-8677-2024>)?

Response: Thank you for this suggestion. A direct quantitative comparison with Sourì et al. (2024) is not straightforward because the two studies use different OH metrics and analysis

periods. Nevertheless, the spatial patterns and signs of the regional trends can be compared qualitatively.

There are several broad similarities. Both studies show strong positive OH trends over India, consistent with increasing NO_x and associated photochemical changes. Sourì et al. (2024) found the strongest NO₂-driven increase in tropospheric OH over India, consistent with the pronounced positive OH-column trend in our simulation. In contrast, Sourì et al. (2024) found declining OH over the North China Plain associated with NO_x reductions after approximately 2011, whereas our simulation for 2000–2017 shows a positive OH-column trend over much of China. This difference may partly reflect the different analysis periods and representation of recent Chinese NO_x emission controls; notably, Sourì et al. (2024) reported that their unconstrained simulation did not reproduce the observed reduction over China until OMI information was incorporated.

A further difference occurs over central Africa, where our simulation shows decreasing OH, whereas Sourì et al. (2024) reported a strong positive trend that could not be explained by the five drivers considered in their analysis. In our simulation, the decrease persists when anthropogenic emissions are fixed, suggesting an important contribution from meteorological variability, although the modelled positive CO trend may enhance the simulated OH decrease. Overall, the comparison highlights both broad consistency in some regional OH trends and the sensitivity of regional trends to the analysis period, emission evolution, model constraints, and the OH metric used.

We have added this comparison to Section 4.4 (Page 27, Line 621-626).

15. Line 547: This would be a good place to also mention the possible effects of the model mismatches with the OMI/MLS O₃ trend.

Response: Thank you. We have revised the text to acknowledge that the underestimated positive trend in tropospheric O₃ column may also contribute to the overestimated OH decreases in this region (Page 27, Lines 631–632).

16. Table 6: Can you add the uncertainty in the trends to the table? This would help show how statistically different the trends in the different simulations are from each other.

Response: Thank you for the suggestion. We have added the 90% confidence intervals of the Theil–Sen trend estimates to Table 6 (Page 27). The confidence intervals provide a direct indication of the uncertainty in the estimated trends and show that, while the fixed-emission experiment exhibits a distinctly stronger trend, the trend differences among most of the other sensitivity simulations are small relative to their uncertainties.

17. Line 562-563: Can you say which meteorological differences drive the trend?

Response: The fixed-emission experiment isolates the combined influence of interannual meteorological variability, but does not allow the contributions from individual meteorological factors to be separated. We have therefore clarified that changes in temperature, water vapour, clouds and photolysis may contribute to the simulated OH trend, while their individual contributions cannot be quantified from the present experiments (Page 28, Lines 649–652).

18. Fig. 10: Does “Mete” in the legend correspond to the fixed emission run? If so, please clarify this in the caption.

Response: Yes, “Mete” corresponds to the fixed-emission (fixEmis) simulation. We have clarified this in the Figure 10 caption (Page 28).

19. Line 581: The statement “effectively reduce this overestimation without substantially altering the OH trend, thereby improving model performance” seems to imply that not altering the OH trend is important to preserving model performance. But do we know the model OH trend is correct?

Response: Thank you for pointing this out. We agree that the original wording could imply that preserving the simulated OH trend represents an improvement in model performance, whereas the accuracy of the modelled OH trend remains uncertain. We have therefore revised the text to clearly distinguish the improvement in simulated mean OH abundance from the relatively small effects of these processes on the simulated OH trend (Page 29, Lines 677–680).

20. Line 583-584: Why does heterogeneous chemistry on clouds suppress the trend? Is there a trend in the model clouds?

Response: Thank you for raising this point. Unlike the prescribed aerosol surface area density, the cloud ice surface area density is calculated using Eqs. (3)–(4) and therefore varies with meteorological conditions from year to year. To examine this explicitly, we calculated the global total cloud ice surface area for each year during 2000–2017. As shown in the new Figure R2, substantial interannual variability is present, with highest values in 2016.

Since heterogeneous chemistry on cloud ice acts as a net sink of OH, the increase in cloud ice surface area provides more surface for heterogeneous reactions and therefore contributes to the slightly weaker positive OH trend when cloud heterogeneous chemistry is included (Table 6).

We have revised the corresponding discussion in the manuscript (Page 29, Lines 686–691) to clarify this point:

“As shown in Table 6 and Figure 10, water vapor UV absorption reduces global mean OH by 3.6% and slightly weakens its increase. Heterogeneous chemistry on clouds decreases OH by 2.4%. Because cloud ice surface area density varies with meteorological conditions and shows a small increase during 2000–2017, cloud heterogeneous chemistry may also suppress the positive OH trend slightly. Aerosol heterogeneous chemistry has a somewhat larger effect on mean OH, reducing it by 3.2%, but has little influence on the OH trend here because the prescribed aerosol surface area density does not capture interannual changes in aerosol abundance. The combined heterogeneous effect reduces global mean OH by 5.8%, with the change in the OH trend arising primarily from cloud heterogeneous chemistry.”

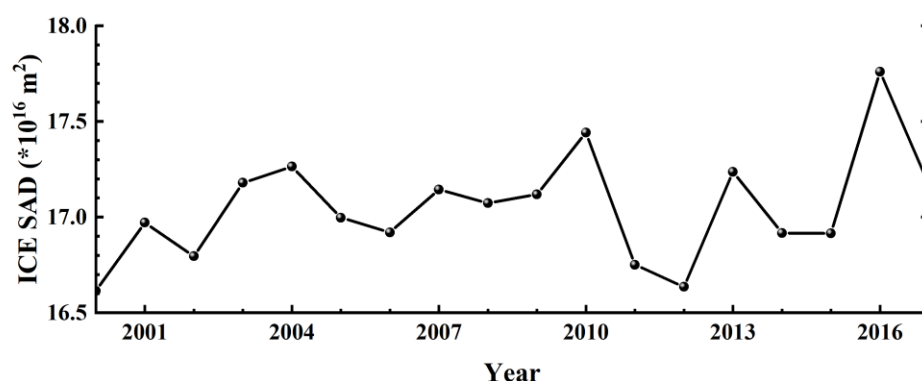


Figure R2. The global total surface area density of cloud ice (10^{16} m^2) from 2000 to 2017.

21. Line 584: Isn't the little influence of aerosols on the OH interannual variability expected from the use of climatological aerosols? Accurately quantifying this effect on interannual variability would require including aerosol interannual variability in the simulation.

Response: We agree. As noted in our response above, the prescribed aerosol surface area density does not include interannual variability. Therefore, the small influence on OH interannual variability is reasonable in the current simulations, and fully quantifying this effect would require interannually varying aerosol surface area densities. We have acknowledged this limitation in the revised manuscript (Page 29, Lines 688–690).

“Aerosol heterogeneous chemistry has a somewhat larger effect on mean OH, reducing it by 3.2%, but has little influence on the OH trend here because the prescribed aerosol surface area density does not capture interannual changes in aerosol abundance.”

22. Line 255-257: Explain how changing a reaction that contributes only 1% of OH loss changes the global mean OH by 7%.

Response: Although the $\text{OH} + \text{NO}_2$ reaction accounts for only about 1% of the globally integrated OH loss, its effect on OH is amplified through the coupled HOx–NOx chemical system. A change in this reaction not only alters the direct OH sink but also perturbs radical recycling and secondary OH production. In the JPL simulation, the decrease in $\text{OH} + \text{NO}_2$ loss

from 2.3 to 2.1 Tmol yr⁻¹ is accompanied by an increase in total OH production from 208 to 217 Tmol yr⁻¹, mainly through changes in O(¹D) + H₂O, HO₂ + NO, and HO₂ + O₃. Thus, the ~7% change in global mean OH reflects the nonlinear adjustment of the HOx–NOx chemical system rather than the change in this direct OH loss pathway alone. We have clarified this point in the revised manuscript (Page 23, Lines 564–565; Page 29, Lines 694–696).

“Despite its small contribution to the global OH loss budget, this change perturbs the coupled HOx–NOx chemistry and enhances radical recycling, leading to a larger response in the steady-state OH abundance.”

and

“The updated OH + NO₂ reaction rate used in the control run also reduces global mean OH by about 7%, while having only a small effect on the simulated trend. Although the OH + NO₂ reaction contributes only ~1% to total OH loss, this sensitivity reflects nonlinear adjustments in HOx–NOx chemistry and radical recycling.”

23. Last paragraph of Section 4: This paragraph would be stronger if it highlighted what new insights into the OH variations are obtained from this comparison. If this comparison is more for model validation, then I recommend moving it further up in the text.

Response: Thank you for this helpful suggestion. The purpose of this comparison is to place the relative temporal evolution of OH simulated by the FRSGC/UCI CTM in the context of previous model and inversion estimates, rather than simply to provide additional model validation. We have revised the paragraph to make the new insights from this comparison clearer. The model results show relatively smooth year-to-year variations and a weak long-term trend, while several temporal features, such as enhanced OH around 2012–2013 and lower OH during 2014–2015, are also evident in other estimates. This agreement suggests that some features of the simulated OH variability are relatively robust across different approaches. At the same time, the substantially different amplitudes, particularly the larger fluctuations in the MCF-based inversions, indicate considerable uncertainty in the magnitude of global OH interannual variability. Overall, the comparison places the weak OH trend simulated by the model within the range of temporal behavior reported by previous studies, while highlighting greater uncertainty in the magnitude of interannual variability. We have revised the discussion accordingly (Pages 29–30, Lines 699–719).

24. Last 2 sentences: Can you add something more specific about which processes most need improved representation? Also, does improving representation in this context mean further research to understand the processes, or incorporating the current understanding of the processes into more models?

Response: Thank you for this helpful suggestion. We have revised the Conclusions to specify which processes most need improved representation and to clarify what we mean

by “improved representation.” Our results highlight the OH + NO₂ reaction kinetics and oceanic acetaldehyde emissions as priorities for further constraints, while for H₂O UV absorption and heterogeneous chemistry, more consistent incorporation of current process understanding across global models is needed. We therefore clarify that improving OH representation involves both further constraints on uncertain kinetics and emissions and broader implementation of processes that are already reasonably understood. We also note that the model configuration used here does not include halogen chemistry, which can influence tropospheric OH and may provide an additional pathway for reducing the high-OH bias. Thus, the processes examined in this study should not be regarded as an exhaustive set of factors contributing to uncertainties in simulated OH.

The final part of the Conclusions has been revised accordingly (Page 31, Lines 762–771):

“Together, these results suggest that improved representation of the processes examined here can help reduce the tendency of global models to overestimate OH and underestimate methane lifetimes, although these processes alone cannot fully account for this bias. The model configuration used here does not include halogen chemistry, which can influence tropospheric OH and may provide an additional pathway for reducing this bias. In this study, the large sensitivity to the OH + NO₂ reaction rate highlights the need to better constrain its kinetics, while the effects of oceanic acetaldehyde emphasize the need for improved constraints on marine emissions. For H₂O UV absorption and heterogeneous chemistry, more consistent implementation of current process understanding across global models would help reduce structural differences in simulated OH. Overall, improving OH representation requires both tighter constraints on uncertain kinetics and emissions and broader incorporation of currently understood processes into global models.”

25. Supplement Fig. S6: I couldn't find a reference to this figure in the text. How does it differ from the right side of Fig. 9?

Response: Thank you for pointing this out. Figure S6 (now Figure S8) shows the OH trends from the fixed-emission experiment, whereas the right column of Figure 9 shows those from the control simulation with varying emissions. The comparison helps distinguish the effects of anthropogenic emission changes from those associated with meteorological variability. Over India and China, the positive OH trends are much smaller in the fixed-emission experiment, supporting an important role for changes in anthropogenic emissions, particularly NO_x and other O₃ precursors. In contrast, OH decreases over tropical Africa in both the control and fixed-emission experiments, suggesting a stronger role for meteorological changes in this region. We have now explicitly referred to Figure S8 in the corresponding discussion in the main text (Page 27, Lines 629–630).

26. Some papers listed in the references do not appear to be cited in the text.

Response: Thank you for pointing this out. We have carefully checked the reference list against the citations in the main text and removed references that were not cited. The reference list has been updated accordingly.

Reviewer 2:

Hou et al. present an evaluation of OH trends from year 2000 to 2017 using a chemistry transport model and their drivers. The studies includes a thorough evaluation of the model with observations. Another novelty of the study is the exploration of the role of introducing newer atmospheric processes, updated rate constants and new oceanic emissions on OH and methane lifetime. The paper is generally well-written, highly relevant to the scope of ACP, and the exploration of new processes / rate constant/ emissions is extremely relevant and valuable to the modelling community. I have some comments which I hope the authors will consider, but other than that, recommend publication.

Response: We thank the reviewer for the positive assessment of our manuscript and for the constructive comments. We have carefully considered all comments and revised the manuscript accordingly. Our point-by-point responses are provided below in bold. All page and line numbers referring to the revisions correspond to the revised manuscript with tracked changes.

General comments:

1. The authors have presented a thorough evaluation of drivers of OH, namely ozone and CO. I was wondering if the authors ever explored how the inclusion/exclusion of the different OH processes affects model performance for these drivers too? Although I understand it gets a bit circular sometimes (the drivers affect OH, but OH also potentially affects the drivers). It just may be of interest to the community to also see how the different processes suggested could affect model performance across multiple metrics.

Response: We thank the reviewer for this helpful suggestion. The manuscript already includes the impacts of these processes on the global tropospheric O₃ budget (Table S5). To further address the reviewer's suggestion, we have now added observational evaluations of O₃ for the sensitivity simulations in the Supplement. Figure S6 compares simulated surface O₃ with TOAR observations, while Fig. S7 compares simulated tropospheric column O₃ (TCO) with OMI/MLS observations.

The new comparisons show that the sensitivity experiments have relatively small effects on surface O₃ over the northern mid-latitude regions examined (Fig. S6). In

contrast, the effects on TCO are more evident, particularly in the tropics (Fig. S7). Removing heterogeneous chemistry increases the positive TCO bias, indicating that its inclusion in CTL helps reduce the model overestimate of TCO. The JPL simulation, which uses the slower OH + NO₂ reaction rate coefficient, produces a larger positive TCO bias than CTL. These results have now been discussed in the revised manuscript (Page 11, Line 328-330 and Page 13, Line 374-380).

2. I have a similar question as the other anonymous reviewer: why do the authors deem that preserving the OH trend across different processes is a goal if there are uncertainties in the trend itself?

Response: We thank the reviewer for pointing this out. We agree that the wording “fully consistent with observational and multi-model estimates” was too strong. The airmass-weighted OH concentration and methane lifetime in our reference simulation already fall within the range of previous observationally constrained and multi-model estimates. The sensitivity experiments are therefore intended to quantify how the processes considered here affect OH and methane lifetime, rather than to correct a specific OH bias in the FRSGC/UCI model. Nevertheless, several processes, including water vapour UV absorption and heterogeneous chemistry, reduce OH and therefore act in the direction expected to alleviate the high-OH/low-methane-lifetime tendency commonly reported in global models.

We have therefore revised the statement in the text and Conclusions (Page 29, Line 677-680 and Page 31, Line 762-771). We have also clarified that these processes mainly affect the magnitude of simulated OH and methane lifetime, while their effects on the simulated long-term OH trend are relatively small. Given the uncertainty in OH trends, this limited trend sensitivity is not interpreted as evidence of improved model performance or as a goal of the sensitivity experiments.

Specific comments:

1. Title: Not sure if it would be good to put the time period in the title? Also, a key novelty is the exploration of physical mechanisms not currently employed in most models, so I was wondering if the title could somehow reflect this? Just some suggestions to make the title more specific and also highlight novel aspects, since the title currently reads as quite general.

Response: We thank the reviewer for this helpful suggestion. We agree that highlighting the process-level sensitivity analysis makes the title more specific and better reflects the novelty of the study. We have therefore revised the title to:

“Tropospheric OH and Its Trends: New Insight into Atmospheric Processes and Implications for Methane Lifetime.”

2. Abstract: ‘substantial increase in OH’: I guess I’m not really certain what would constitute as a ‘substantial’ increase? Also, ‘dynamic emissions’ - would suggest rewording this to say ‘time-varying emissions’?

Response: We agree that attributing the OH increase simply to “meteorological conditions alone” was too broad. In the fixed-emission simulation, meteorological changes can affect OH through several pathways, including changes in temperature, water vapour, clouds and photolysis, transport, and meteorology-dependent lightning NO_x. As these individual contributions were not isolated in separate sensitivity experiments, we have revised the Abstract to avoid attributing the increase to a specific meteorological mechanism (Page 1, Line 26-27). We have also replaced “dynamic emissions” with “time-varying emissions”.

3. Line 39: The authors mention ‘future atmospheric composition’, whereas the studies cited (Stevenson 2020, Lelieveld 2016, Chua 2023) are historical. Perhaps some studies like Voulgarakis et al. (2013), Liu et al. (2024) or Chua et al. (2026) could be useful as some examples of future scenario OH / methane lifetime drivers, though I guess if the broader point is about exploring the ‘interactions between natural and anthropogenic emissions and climate change’, then it could be fine to remain as is.

Response: We apologize that this was not clearly expressed. We have added Voulgarakis et al. (2013), Liu et al. (2024) and Chua et al. (2026), which explicitly examine future changes in OH and methane lifetime, to better support this statement (Page 2, Lines 41–42), and the corresponding references have been added to the reference list.

4. Table 4: Why is there a table entry that is bolded?

Response: The bold font was originally used to highlight the major reaction changes in the sensitivity experiments for ease of analysis. To avoid confusion, we have removed the bold formatting from Table 4 in the revised manuscript.

5. Line 270: Would suggest not using the term ‘long-term’ for the study period of 17 years.

Response: We agree and have removed the term “long-term”. Corresponding changes have also been made throughout the manuscript for consistency.

6. Line 275: 'airmass' rather than mass?

Response: We agree and have changed “mass” to “airmass” as suggested.

7. Figure 7:

(1) Panel (a) title units should say 'molec cm⁻³' not 'moles cm⁻³'.

(2) Currently, the caption which says 'difference in OH (in %) in the troposphere due to (b) removing water vapour UV absorption (WVA) ...', but perhaps it would be helpful for the reader to remove the word 'removing', since what is being shown is actually the isolated WVA effect from CTL minus noWVA. Same suggestion for the other instances of 'removing' for the other processes.

(3) As for JPL, the panel f title (JPL) and also the caption description is a bit confusing, since if I'm not mistaken what's really being plotted is the effect of the updated rate constant versus the JPL rate constant?

Response: We appreciate the reviewer for pointing out these unclear expressions. We have corrected the unit in panel (a) from “moles·cm⁻³” to “molec·cm⁻³” and removed “removing” from the caption. We have also clarified that the percentage differences are calculated as (CTL – experiment)/CTL × 100%. For panel (f), this represents the effect of the updated OH + NO₂ + M reaction rate used in CTL relative to the JPL rate, and the panel title has been changed from “JPL OH + NO₂ + M” to “OH + NO₂ + M” to avoid confusion.

8. Line 485: Regarding the phrase 'strong solar radiation' here: suggest rephrasing. I'm guessing that the authors mean something like, greater amount of UV radiation available to produce O¹D due to reduced UV absorption? If so, maybe something to that effect would be good here.

Response: We agree that “strong solar radiation” was unclear. We have revised the text to clarify that neglecting water vapor UV absorption allows more UV radiation to contribute to O₃ photolysis and subsequent O(¹D) production, thereby enhancing primary OH production (Page 22, Lines 559–560).

9. Line 491: 'Overall, the sensitivity experiments highlight the key atmospheric processes influencing OH levels and quantify their respective impacts.' : perhaps it would be worthwhile to highlight that the OH budget analysis of the sensitivity experiments reveal the chemical pathways through which the different processes explored in this study affects OH levels?

Response: We appreciate this helpful suggestion. We have revised the text to highlight that the OH budget analysis reveals the chemical pathways through which the processes examined in this study affect OH production and loss (Page 23, Lines 568–569).

10. Section 4.5 title: ‘Key processes’: I guess the processes being isolated are key to the study, but I’m not sure if they are ‘key’ processes per se compared to other processes? To avoid being misleading, I would suggest having more specific language to constrain the scope of the discussion here to be limited to the specific processes discussed in this study.

Response: We appreciate this helpful suggestion. We have replaced “Key Processes” with “Selected Atmospheric Processes” to better constrain the discussion to the specific processes examined in this study (Page 27, Line 634).

11. Line 540: What is a ‘coherent’ region?

Response: We agree that “coherent” was unclear. We have replaced “large, coherent regions” with “spatially extensive regions” to more clearly describe the spatially widespread decreases in OH (Page 27, Lines 619–620).

12. Line 559-560: ‘...but estimates from atmospheric chemistry transport models typically suggest an increase over this period (Stevenson et al., 2020; Zhao et al., 2020)’: Just a technicality that Stevenson et al. (2020) used chemistry-climate models /Earth system models, not chemical transport models, so perhaps it would be useful to revise the statement to be more specific.

Response: We appreciate this clarification. We agree that “atmospheric chemistry transport models” does not accurately describe the different approaches used in the cited studies. Stevenson et al. (2020) used chemistry-climate/Earth system models, while Zhao et al. (2025) estimated OH variations using an observation- and model-driven approach with chemical mechanisms based on MOZART and GEOS-Chem. We have therefore revised the wording to the more general term “model-based studies” (Page 28, Lines 644):

“...but estimates from several model-based studies suggest an increase over this period (Stevenson et al., 2020; Zhao et al., 2025).”

13. Line 570-571: 'slight increase in global mean OH from 2006 to 2007 and decreases in 2002 and 2015 in the control run.': Actually it seems like there are a lot more features than those highlighted, for example the spike in 2010, dip in 2011, spike in 2012, etc.

Response: We agree that Figure 10a shows additional interannual features beyond those originally highlighted. We have revised the text to better describe the overall interannual variability, while highlighting the pronounced decreases in 2002 and 2015 and increases in 2007 and 2016 (Page 28, Lines 662–665).

14. Line 571: 'anomalously low': not sure based on the Figure S3 that CO emissions are 'anomalously' low for year 2007, though there is clearly a dip from 2006 to 2007, so perhaps rephrasing to emphasise that it is a dip rather than an anomalous low.

Response: We agree that “anomalously low” was too strong. We have revised the text to describe the decrease in fire-related CO emissions from 2006 to 2007 more accurately (Page 28, Lines 666).

15. Line 576: I was wondering if it would be straightforward to, for example, calculate a correlation coefficient between OH anomaly and CO emission anomaly to substantiate the point?

Response: We appreciate this nice suggestion. We have calculated the correlation between annual anomalies in global mean OH and fire-related CO emissions over 2000–2017 and find a strong negative correlation ($R = -0.75$). We have added this result to further support the influence of interannual variability in fire-related CO emissions on OH, while noting the uncertainty in the CO emission inventory used in this study (Page 29, Lines 671–674).

16. Paragraph from Line 578 to 587: I'm curious why the authors highlight water vapour UV absorption and heterogeneous reactions specifically in the first sentence of the paragraph as processes that can reduce positive bias of OH without affecting trend, but the last sentence of the paragraph then also says that the updated reaction rate coefficient of $\text{NO}_2 + \text{OH}$ also does the same thing. Unless I'm missing something, I would suggest restructuring the paragraph to more generally say in the first sentence that all the processes explored in this study reduces positive bias of OH without affecting trend, then describing the various specific processes in the rest of the paragraph.

Response: We appreciate this helpful suggestion. We have restructured the paragraph to first clarify that all the processes examined here primarily affect the mean OH abundance, while their effects on the simulated OH trend are relatively

small. We then discuss the effects of water vapor UV absorption, heterogeneous chemistry, and the OH + NO₂ reaction rate separately. We have also clarified that the limited trend sensitivity should not be interpreted as evidence of improved model performance, given the uncertainty in OH trends (Page 29, Lines 677–680).

17. Line 580: “Most chemistry-climate models tend to overestimate OH”: it would be helpful to reintroduce a relevant citation here.

Response: We agree and have added relevant citations (Shindell et al., 2006; Fiore et al., 2009; Naik et al., 2013) to support the statement regarding the high-OH tendency in global models (Page 29, Lines 679–680).

18. Line 583: ‘decreases OH by 2.4% and slightly weakens its increase’: reading it without context is confusing since these seem contradictory, but contextually it is interpretable. Suggest slightly rephrasing to something like ‘decreases OH by 2.4% and slightly weakens the increasing trend’ or something like that.

Response: We agree that the original wording could be confusing. We have rephrased the sentence to clearly distinguish the effects on mean OH abundance from those on the OH trend (Page 29, Lines 688–689).

19. Figure 10:

(1) From panel a, does meteorology potentially also play a role in driving interannual variability?

Response: We agree that meteorology also contributes to the interannual variability in OH. While its contribution to the OH trend is discussed in the preceding paragraph, we have added a sentence noting that clear interannual variability also remains in the fixed-emission simulation, further indicating a role for meteorological variability in year-to-year changes in OH (Page 28, Lines 664–665).

(2) Similar comment to Figure 7 especially for the JPL label in Panel b, in that if I’m not mistaken, the ‘JPL’ label actually stands for the effect of using the new updated rate constant versus using the JPL rate constant, so using ‘JPL’ as a label is a bit misleading.

Response: We agree that the “JPL” label could be misleading. We have changed the label in Figure 10b from “JPL” to “OH + NO₂ + M” to clarify that it represents the effect of the updated OH + NO₂ + M reaction rate used in CTL relative to the JPL rate.

(3) Would suggest adding a label referring to Zhao et al., 2025 for MOZART and GEOS-Chem runs in the figure panel a.

Response: We appreciate this suggestion. To avoid adding additional labels to the already crowded panel, we have instead added the relevant references to the Figure 10 caption. The caption now identifies the MOZART and GEOS-Chem results as being from Zhao et al. (2025), together with the sources of the other estimates shown in panel (c).

(4) In the caption for panel b, currently it reads ‘the average relative differences in [OH]gm from the control run (%) from 2000 to 2017’. Currently, because the figure mixes time series with the bar chart, the current phrasing could be interpreted as ‘the difference in the change in OH in year 2017 versus 2000 due to the different processes’. I would suggest rephrasing it to say something like ‘the average relative difference in the annual mean [OH]gm averaged over the period 2000-2017 for the respective model runs’, or something like that.

Response: We agree that the original wording could be misinterpreted. We have revised the caption to clarify that panel (b) shows the relative differences in annual mean [OH]gm between CTL and the respective sensitivity runs, averaged over 2000–2017.

(5) Did the authors also look at line plot time series for all the model runs, not just for CTL and no emissions runs? Maybe it doesn’t reveal anything new or interesting, but I was wondering if the different processes show similar peaks and dips during the period, and if the processes contribute to interannual variability too.

Response: We have examined the annual [OH]gm time series for all sensitivity simulations, as shown in the figure below (Figure R3). Except for the fixed-emission simulation, the sensitivity runs show very similar interannual variations to CTL, with the major peaks and dips occurring in the same years. The relative differences from CTL also remain nearly constant over 2000–2017, indicating that the processes examined here primarily affect the mean OH abundance, with only small effects on its interannual variability. In contrast, fixing emissions produces a more evident change in the OH trend and interannual variability. As this analysis does not reveal substantial additional features beyond those already discussed in the manuscript, we show the figure here for clarification but have not added it to the revised manuscript.

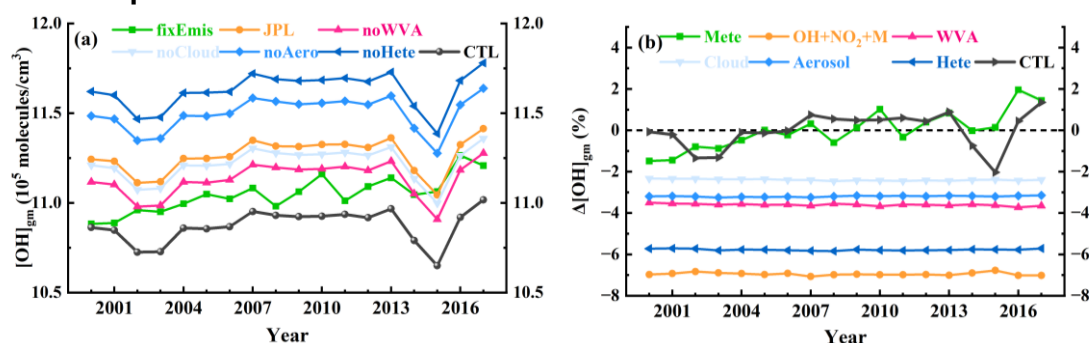


Figure R3. (a) Annual $[\text{OH}]_{\text{gm}}$ ($10^5 \text{ molecules}\cdot\text{cm}^{-3}$) for the CTL and sensitivity simulations, and (b) relative differences in $[\text{OH}]_{\text{gm}}$ from CTL (%) for the sensitivity simulations, with the relative anomalies of CTL and Mete also shown for comparison, over 2000–2017.

20. Line 589: The reference to Zhao et al. (2025)'s MOZART and GEOS-Chem runs could be a bit misleading since technically Zhao et al. (2025) used the same modelling framework for the two different chemical mechanisms taken from MOZART and GEOS-Chem. I would suggest adding a brief summary of what Zhao et al. (2025)'s runs are doing here.

Response: We appreciate this clarification. We have revised the text to clarify that Zhao et al. (2025) estimated OH variations using an observation- and model-driven approach with chemical mechanisms based on MOZART and GEOS-Chem, rather than using MOZART and GEOS-Chem as two separate models (Page 29, Lines 701–702).

21. Line 612: 'A slower reaction rate for the termolecular reaction between OH and NO₂ leads to a 7% increase in global tropospheric OH that is largest in the upper troposphere and at high latitudes.': Suggest rephrasing to talk about the use of a faster reaction rate leads to a decrease in global tropospheric OH, since that is more consistent with the phrasing used in the rest of the paper.

Response: We agree and have rephrased the sentence to describe the effect of the faster OH + NO₂ + M reaction rate used in the control run relative to the slower JPL rate, consistent with the terminology used elsewhere in the manuscript (Page 30, Lines 734–736).

22. Line 622: 'meteorological conditions (contributing ~60%) but also reflect a mismatch in the modelled CO column trend (~16%)': It would be good to see these specific percentage numbers somewhere in the results section too? Not sure if I see how these specific numbers were derived.

Response: We appreciate this comment. The ~60% estimate was derived from the OH trends in the fixed-emission and CTL simulations, while the ~16% estimate was an approximate calculation based on the difference between simulated and observed CO column trends and the contribution of OH + CO to total OH loss. We agree that these estimates do not provide a rigorous quantitative attribution. We have therefore removed the specific percentages and revised the text to describe

the effects of meteorological conditions and the uncertainty associated with the simulated CO trend qualitatively (Page 31, Lines 746–748).

23. Line 625: It would be good to specify ‘annual’ trend for the number quoted here to prevent confusion with the previous sentence which quoted the summer trend number.

Response: We agree and have clarified that the value of $1.69 \times 10^3 \text{ molec}\cdot\text{cm}^{-3}\cdot\text{yr}^{-1}$ refers to the annual trend in global mean OH (Page 31, Lines 751).

24. Line 630: Doesn’t the OH + NO₂ reaction rate change also not change the trend by much, the same as with the heterogeneous process and water vapour UV absorption? (similar comment as for Paragraph line 578-587).

Response: We agree. We have revised the text to clarify that the OH + NO₂ reaction rate, like water vapor UV absorption and heterogeneous chemistry, affects OH levels but has only a small effect on the simulated trend (Page 31, Lines 757).

25. Conclusions: It would be good to see some sort of discussion about a broader recommendations to the community based on the findings from this study.

Response: We appreciate this helpful suggestion. We have expanded the Conclusions to include broader recommendations for improving OH representation in global models, focusing on uncertain kinetics and emissions and the consistent implementation of relevant atmospheric processes (Page 31, Lines 762–771).

26. Citations:

Chua, G., Naik, V., & Horowitz, L. W. (2026). Role of future climate change, air pollution control and methane mitigation in driving hydroxyl radical (OH) and methane lifetime. *Geophysical Research Letters*, 53 (9), e2025GL120136. doi:<https://doi.org/10.1029/2025GL120136>

Liu, M., Song, Y., Matsui, H., Shang, F., Kang, L., Cai, X., et al. (2024). Enhanced atmospheric oxidation toward carbon neutrality reduces methane’s climate forcing. *Nature Communications*, 15(1), 3148. <https://doi.org/10.1038/s41467-024-47436-9>

Voulgarakis, A., Naik, V., Lamarque, J.-F., Shindell, D. T., Young, P. J., Prather, M. J., et al. (2013). Analysis of present day and future OH and methane lifetime in the ACCMIP simulations. *Atmospheric Chemistry and Physics*, 13(5), 2563–2587. <https://doi.org/10.5194/acp-13-2563-2013>

Response: We appreciate these helpful references. They have now been cited where appropriate in the revised manuscript and added to the reference list.