

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.

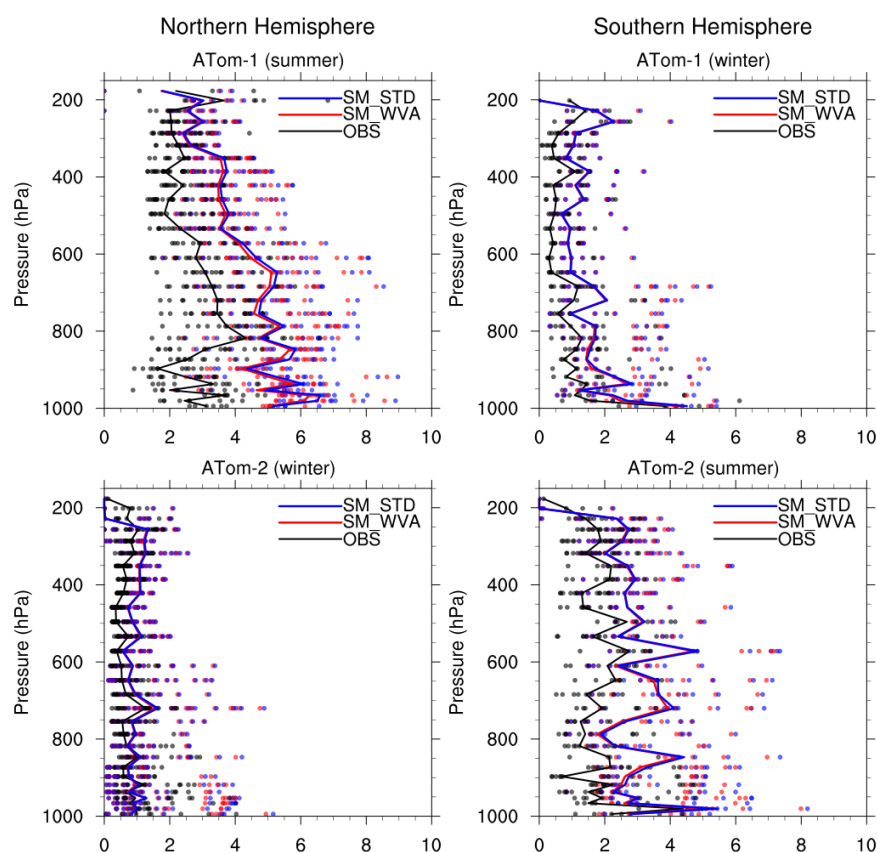
We note, however, that Figure R1 represents an evaluation performed during an earlier stage of model development. We therefore avoid attributing the present-day model–observation agreement quantitatively to individual updates based on this comparison. Instead, the sensitivity experiments presented in 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.”



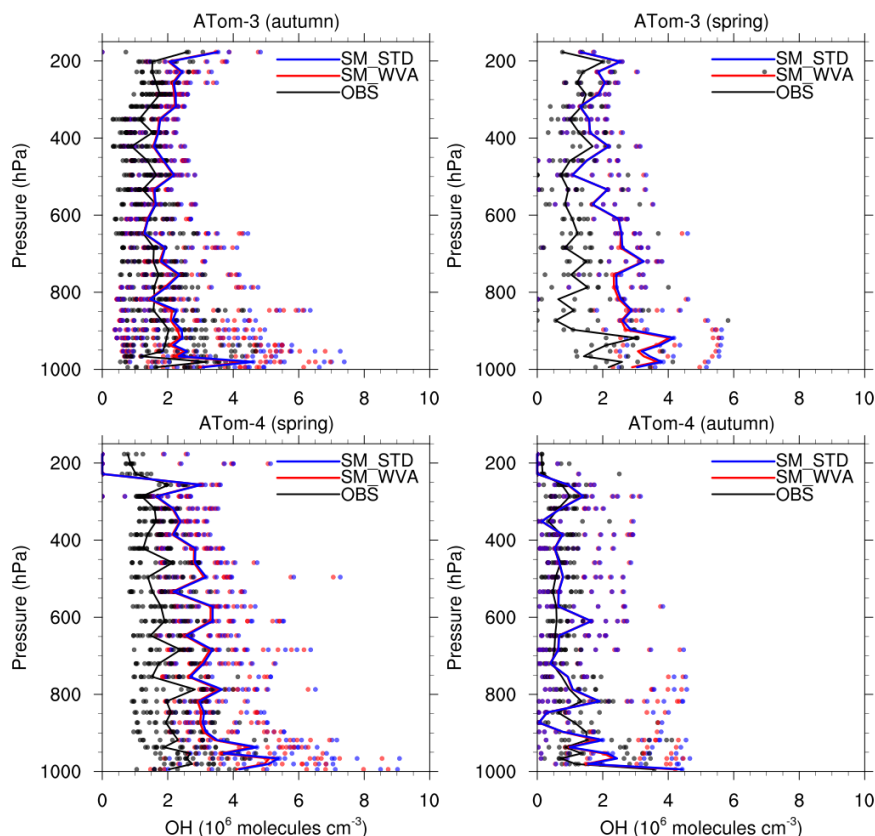


Figure R1. Median OH concentrations (10^6 molecules- cm^{-3}) measured during the ATom aircraft missions (black) and corresponding simulations from an earlier stage of FRSGC/UCI model development. SM_STD represents the model configuration before the process updates considered in this study, while SM_WVA 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. Souri 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, Souri 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, Souri 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 Souri 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.”

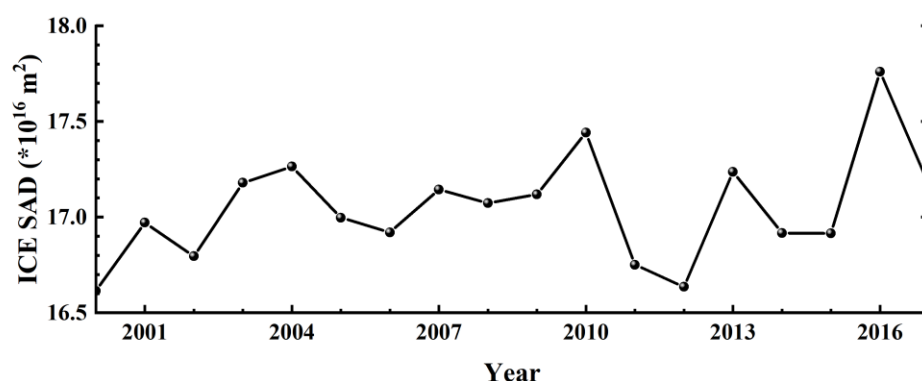


Figure R2. The global total surface area density of cloud ice (10^{16} m^2) from 2000 to 2017.

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.