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 O_3 budget (Table S5). To further address the reviewer's suggestion, we have now added observational evaluations of O_3 for the sensitivity simulations in the Supplement. Figure S6 compares simulated surface O_3 with TOAR observations, while Fig. S7 compares simulated tropospheric column O_3 (TCO) with OMI/MLS observations.

The new comparisons show that the sensitivity experiments have relatively small effects on surface O_3 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 + NO_2 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 NOx. 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 + NO_2 + M reaction rate used in CTL relative to the JPL rate, and the panel title has been changed from “JPL OH + NO_2 + M” to “OH + NO_2 + 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 O_3 photolysis and subsequent O(^{1}D) 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, Lines662–665).

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 NO2 + 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 + NO_2 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 + NO_2 + M" to clarify that it represents the effect of the updated OH + NO_2 + 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.

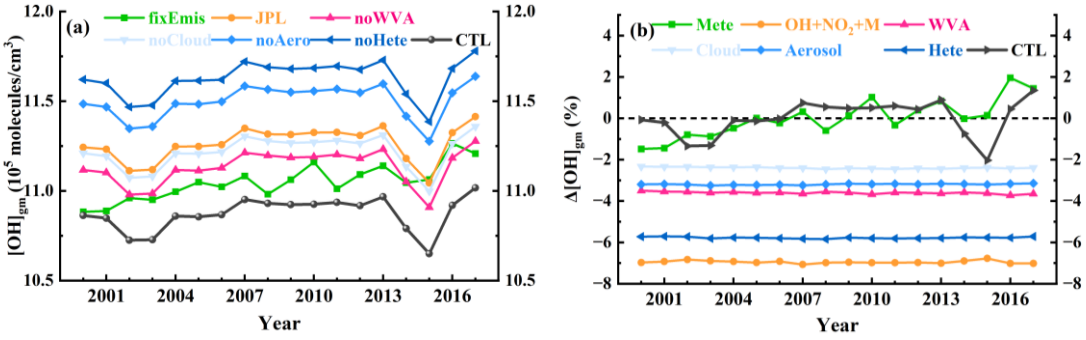

Figure R3. (a) Annual $[OH]_{gm}$ (10^5 molecules·cm^{-3}) for the CTL and sensitivity simulations, and (b) relative differences in $[OH]_{gm}$ from CTL (%) for the sensitivity simulations, with the relative anomalies of CTL and Mete also shown for comparison, over 2000–2017.

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 + NO_2 + 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 1.69×10^3 molec·cm^{-3}·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 + NO_2 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.