

We thank the reviewers for their comments and have addressed them in red below. Page and Line numbers refer to the tracked changes version of the documents.

Review of Anderson et al., 2020

Major Comments:

The goal of this study is to determine the relationship between tropospheric OH and ENSO, Northern Hemispheric modes of variability, the Indian Ocean Dipole, and monsoons. The authors present an analysis of one main model (GEOSCCM) evaluated with aircraft and satellite observations, to make the case that their model can be used for this purpose, and show that their findings have some similarities with the results from four CCMi models. This work finds that multiple modes of climate variability, including ENSO, can explain OH variability over approximately 40 % of the globe. The authors also find that OH mainly changes in the boundary layer and upper troposphere, and the mid-troposphere is less impacted by different climate modes. I have several key concerns. First, the analysis of the model with aircraft and satellite observations is lacking in clarity, and possible technical approach, depending on the answers to my specific comments below. As an example, given the difficulty the model has in representing ATom OH observations in outflow from South America and New Zealand, changes over these regions should be more carefully discussed in Section 5. The authors might also discuss why they do not use any aircraft observations over land. In addition, a key finding of this paper is the finding that lightning NO_x is not a main driver of the OH-ENSO relationship, in contrast to Turner et al., 2018. The authors need to improve their support of this argument. Finally, I think the authors are missing an opportunity to address the causes of the wide variety in OH and methane lifetime across models that they point out in their introduction. While the authors say that there is agreement across models in the importance of the driving factors of OH variability, for example only three models show that ENSO is important in DJF. This manuscript is appropriate for ACP and will represent a strong contribution to the field after the major revisions to the analysis described below.

Specific Comments:

Page 2, line 58 – It is incorrect to say that formaldehyde only comes from CH_4 oxidation, please clarify what you mean here.

We have changed this sentence to read (Page 2, Line 68-70):

“Recent work has demonstrated that formaldehyde, a longer-lived species (hours) whose chemical production in the remote troposphere is often controlled by CH_4 oxidation, shows promise for inferring variability in OH columns over the remote atmosphere (Wolfe et al., 2019).”

Page 3, line 135 – Could you clarify what you mean by detrend? Are you removing the seasonal cycle?

For each grid box, we remove any linear trend in OH on a month-by-month basis to account for any changes in background values, as an increasing or decreasing background could affect the correlations. There is no need to remove the seasonal cycle as we perform separate correlations for each season. We have clarified the text (Page 3, Line 155-157) to read as follows:

“To perform the regression, we first detrend the output on a monthly basis, removing any linear trend from each variable over the 1980 to 2018 period to account for changes in the background value.”

Page 3, line 143 – Could you describe whether the climate features you take from NOAA (MEI, DMI, the Northern Hemispheric modes) are well represented in the models so that you don't have to recalculate them from the model output?

This is a good question. The Dipole Mode Index as calculated by NOAA is based on Sea Surface Temperatures (SSTs) from the Hadley SST dataset, which is the same dataset used in the CCMi model runs. While MERRA2 SSTs are assimilated from various sources, they agree well (within 0.2 degrees) with other reanalyses (Bosilovich, 2015). Because of the similarities in these datasets, the DMI should be nearly identical to analogously defined indices calculated from each of the models examined here. Likewise, the MEI is calculated using sea level pressure (SLP), SSTs, and surface level zonal and meridional winds from the JRA-55 dataset, which was also used to constrain the MRI model in this study. As demonstrated in Orbe et al (2020), there was good agreement in the

temperatures and zonal winds in the CCMI SD runs for the models constrained to the JRA-55, ERA-Interim, and MERRA reanalyses, while agreement in meridional winds was good, but not as robust. MERRA2 surface winds, both zonal and meridional, agree within less than 0.2 m/s in the tropics with both the MERRA and ERA-Interim datasets. Therefore, the MEI would agree with climate indices calculated directly from these models. Likewise, the MEI is highly correlated with other ENSO indices, such as the Nino3.4 index, calculated directly from SSTs, which as we mention above, are almost identical among the different models considered here. Finally, for the Northern Hemispheric modes, we do not have geopotential height at 500 mbar for all of the models considered here so would not be able to calculate the indices for each of the northern hemispheric modes, as defined by NOAA. For comparison, we have calculated an NAO index from MERRA2 GMI using the difference between SLPs at Iceland and the Azores. Correlation between this index and the NOAA derived NAO index agree with an r^2 of 0.79, suggesting that the NOAA index is able to describe the NAO in the model. Further evidence is given by the results shown in Figures 21a and S15, which show the relationship between OH and the various northern hemispheric modes for each model, where the areas of correlation correspond to the regions typically associated with each of the modes.

We have summarized this work in the paper (Page 4, lines 183-190):

“The MERRA2 GMI (Section 2.2) and CCMI models (Section 2.3) included here are constrained or nudged to reanalyses data (MERRA, MERRA2, JRA-55, and the ERA-interim) which assimilate observed meteorology. The meteorological variables used to calculate the DMI and MEI, including sea surface temperature, sea level pressure, and zonal and meridional winds, agree well or are identical among the different reanalyses (Orbe et al., 2020; Bosilovich et al., 2015). Thus, climate modes in these models correspond to the NOAA indices. Likewise, indices for the NAO calculated from surface pressure from the models correlate well (r^2 of 0.79 or greater) with the NAO index calculated by NOAA.”

Page 4, line 168 – Again please describe the method here a little better. What linear fit are you subtracting? What trend are you removing? Why do you have to divide by the standard deviation?

We have removed reference to dividing by the standard deviation as this does not affect our results, although this is necessary for some forms of extended EOF analysis not performed here. We have edited the description as follows (Page 5, line 214-216):

“To perform the analysis, OH fields for each grid box were detrended by subtracting a linear fit to the time series over the 1980 to 2018 period to account for changes in background associated with long-term trends in OH.”

Page 5, line 202 – It might be clearer to say that “over the tropical Pacific area, emissions vary by up to 50 % over the time period studied.”

We have made this change.

Page 6, line 268 – Are you including the land crossings, or only using ocean data? The OH points that degrade the model/observation correlation seem like they must have some defining characteristic – if not over land, than maybe some other type of plume?

We include all OH and CO observations from ATom, which we now clarify in the paper. We describe the model outliers more thoroughly in the response to Page 7, Line 285 below.

Page 6, line 275 – Per my comment above, would it not be more appropriate to average the ATom observations to the model output resolution, rather than interpolate to ATom? Otherwise, this is not an apples to apples comparison. There is no way the model will compare well to 5-min data when it is output at hourly resolution

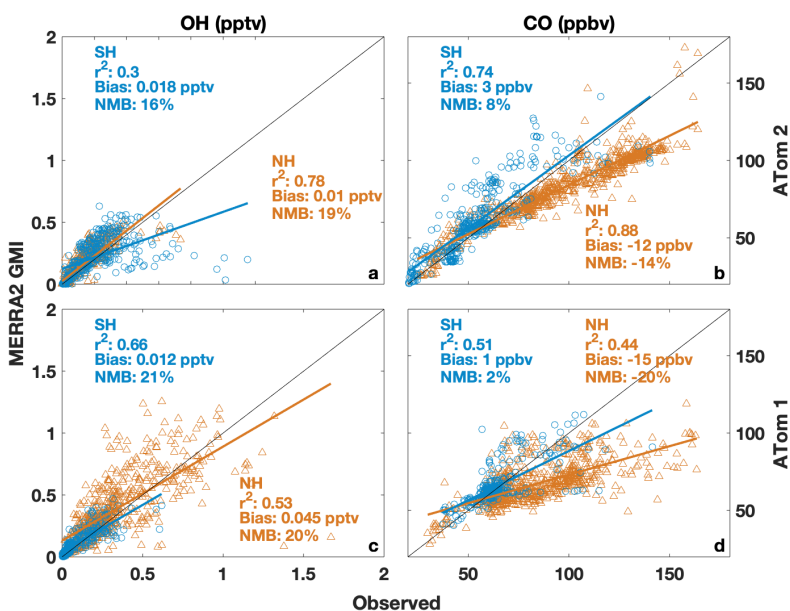
We spent some time debating the best way to do a measurement/model comparison, and each way presents its own set of complications. If the observations were made at a constant location, we would agree with your suggested approach. Because of the campaign sampling strategy, however, averaging the ATom observations to an hourly time scale averages over space rather than time and is still not an apples-to-apples comparison. For example, over an hour period, an ATom flight could complete approximately two vertical profiles spanning from ~250 hPa to ~950 hPa. Averaging data over this period could involve multiple air masses with distinct features, none of which would

be represented by an average. These averaged data would therefore not be comparable to the model output for the averaged latitude, longitude, and pressure of the sampling interval. Similar problems would be present if the plane were only travelling horizontally. While we could use observations averaged over a shorter time scale near the model output time (observations made within 2.5 min of the model output time, for example) this would greatly limit the number of data points for comparison. We also note that this method of comparison has been used for OH in the remote atmosphere in previously published work (e.g., Nicely et al, 2016).

Nicely, J. M., et al. (2016). "An observationally constrained evaluation of the oxidative capacity in the tropical western Pacific troposphere." *Journal of Geophysical Research: Atmospheres* **121**: 7461-7488.

Page 6, line 279 – I would prefer Fig S2 to be part of Figure 1. It seems arbitrary which is shown in the main text, and they are both important, particularly since Fig. S2 doesn't show the same OH outliers in the SH as Figure 1. It would also be useful if they were on the same scale.

We had included ATom 2 as a figure in the main text because, for the most part, the work focuses on relationships during DJF, and limited the ATom 1 figure to the supplement to help control length of an already long paper. We have combined the two figures, which we show here, making sure that the axes have the same scale for both ATom deployments.



Page 7, line 285 – I see that the outliers are somewhat explained here as being driven by continental outflow from South America and New Zealand. Does this imply errors in model NO_x in these regions? Or missing model OH recycling from biogenic VOC oxidation? Can ATom shed some light on this? Just stating that the correlation improves when those points are removed does not improve confidence in the ability of the model to simulate OH.

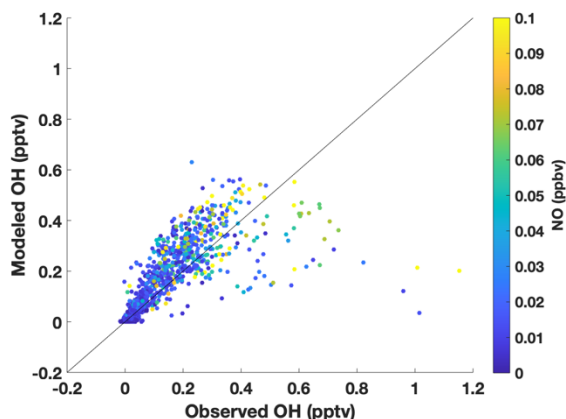
This is a good question. It is difficult to evaluate the cause of differences between observations and the model because hourly resolution output for MERRA2 GMI is only available for a small number of species, which do not include NO_y constituents or any VOCs. We have looked at the correlations in Figure 1a colored by different observed species, such as NO which is shown below, but there is no obvious relationship between the data points underestimated by the model and any of these species. Particularly off the coast of South America, it is possible that this underestimate is related to incorrect isoprene chemistry and emissions in the model, as there is a known high bias in isoprene emissions in the model, resulting in unrealistically low OH over parts of South America. We note this in the text as follows (Page 8, Line 374-378):

The limited model output at hourly resolution does not allow for a determination of the cause of this disagreement in continental outflow regions. In the case of South America, however, a known high

bias in modeled isoprene, resulting in extremely low OH over the Amazon, is consistent with the disagreement between the simulation and observations.

Because the measure/model disagreement is limited to these continental outflow regions and the majority of the correlations discussed in the paper are in the remote atmosphere, in regions unaffected by fresh continental emissions, we do not explore the causes of the disagreement in more detail. We also note, that outside of these outflow regions, the model agrees with the measurements within measurement uncertainty. We have updated the text to read (Page 9, Line 400-403):

This lack of agreement does not significantly affect the results discussed in this work, as the majority of the relationships found between OH and modes of climate variability discussed in Sections 4 and 5 are centered in the remote atmosphere.



Page 7, line 293 – To my knowledge, it has not been shown that fixing the CO bias improves model OH biases. It is true that the low CO bias in the NH is well known, and the model OH bias is well known, but not that the OH bias is due to low CO. Please clarify your point here.

We did not intend to imply that the OH bias is exclusively due to the model underestimate in CO, merely that it could be a contributing factor, since it is the dominant sink on a global scale. Indeed, our own results suggest there must be additional issues as the OH bias exists in both hemispheres, despite the hemispheric asymmetry of the CO bias. We have revised the sentence to read (Page 9, Line 391-393):

“This NH low bias in CO is a well-known problem in global chemistry models (Naik et al., 2013; Stein et al., 2014; Travis et al., 2020) and could be a contributing factor in the overestimate in OH, as CO is the dominant global OH sink.”

Fig. 2 caption, please explain what you mean by “satellite lifetime?”

We have edited the text to clarify that we use data averaged from the beginning of available data for a given satellite (e.g. 2003 for AIRS) through 2018, when the simulation ended. The updated text reads:

Figure 1: Tropospheric column CO (left), $H_2O_{(v)}$ (middle), and NO_2 (right) from MOPITT, AIRS, and OMI, respectively (top row), and MERRA2 GMI (middle row) for DJF. For the satellite retrievals and model, data are averaged over the time range described in the text for each instrument. The fractional difference between MERRA2 GMI and the satellite is shown in the bottom row.

Page 7, line 324 – Again it would be easier to read this if Fig. S3 was part of Fig. 2. It seems important enough to the discussion to warrant being in the main text.

As with the ATom 1 comparison, we originally chose to include the JJA satellite comparisons in the supplement to limit paper length and because the majority of the paper analysis focuses on DJF. But we understand your point so are now including the JJA satellite comparison in the main text as Figure 3.

Page 7, line 324 – You say that “overestimates of column CO, averaging 45%, in the SH corresponding with areas of biomass burning.” Is that really true in SH winter (Fig. S3)?

You are correct. A more likely source of the CO overestimate in this region is a known model bias in isoprene, which would lead to an overestimate in *in situ* CO production from biogenic species. The text now reads (Page 10, 447-449):

“These areas of high bias over South America likely result from the high bias in isoprene emissions, as discussed in Section 2.2, that would lead to unrealistically high *in situ* production of CO.”

Page 8, line 341 – Are you re-calculating the OMI NO₂ columns using the model *a priori* to give you an apples to apples comparison, per Lamsal et al., 2014 (doi:10.5194/acp-14-11587-2014)?

We have not. We incorrectly indicated in the paper that we used version 3 of the OMI product, when we actually used version 4 (Lamsal, 2021). The *a priori* used for the OMI retrievals is from a GMI replay run using MERRA2 meteorology and the same emissions used for the simulation described here, and as such, according to Lok Lamsal, it is not necessary to re-calculate the OMI columns. We have updated the text and relevant references to reflect that we were using version 4 data. And, in response to comments from the other reviewer we have added the following paragraph discussing the satellite/model comparison (Page 9, Line 421-431):

“For comparison of the satellite retrievals to MERRA2 GMI, we use monthly fields of the model variables output at the satellite overpass time. For CO, where averaging kernel and *a priori* information are available for the Level 3 MOPITT data, we convolve the model output with these variables so that direct comparison between satellite and model are possible. While shape factors and scattering weights for the OMI NO₂ retrieval are unavailable for the Level 3 data, shape factors for the OMI NO₂ retrieval are determined from a similar setup of the GEOSCCM model, also employing the GMI chemical mechanism and MERRA2 meteorology. Applying the satellite shape factors to the simulation discussed here would therefore not result in significant changes in the modeled NO₂. Finally, for AIRS H₂O, averaging kernel information was unavailable for the Level 3 data, so numerical comparisons between satellite and model should be regarded as more qualitative than quantitative.”

Page 10, line 408 – You say, “The relative importance of the individual reactions is similar during neutral and La Niña years (not shown) and is in agreement with previous model studies (e.g. Spivakovsky et al., 2000).” I am confused since you show in Fig. 4 that ENSO is correlated with TCOH, mainly in the tropics. How can this be if the relative importance of R1-4 is the same during neutral and La Nina years?

While the relative importance of the individual reactions is the same during El Niño, La Niña, and neutral events (e.g., the NO + HO₂ reaction is always the dominant reaction for OH production in the tropical UFT, regardless of ENSO phase), the production rate from that reaction can still vary with ENSO phase. So, while the OH production rate from this reaction is lower over much of the tropics during an El Niño event than during other phases, OH production from this particular reaction is still larger than for the other reactions. Similar reasoning applies to the other atmospheric levels.

We have added the following text to help clarify this point (Page 13, Line 556-559):

While the production rates along these pathways vary with the ENSO phase, as discussed in Sections 5.2 and 5.3, the relative importance of the individual reactions is similar during neutral and La Niña years (not shown) and is in agreement with previous model studies (e.g. Spivakovsky et al., 2000).

Page 10, line 413 – You do not use the term ‘UFT’ in the supplement (Fig. S4). Please be consistent in your terminology.

Thanks for pointing this out. This was left over from a previous set of terminology, and we missed updating this figure. We have updated the labels in the figure (now Fig. S2).

Page 12, line 456 – Does this mean that the variability shown in Fig 6a is driven mainly by changes in DJF as shown in Fig. 7a? If so, should Fig. 6 be shown seasonally similar to Fig. 7?

Figure 6a shows the anomalies in TCOH in DJF, while Figure 7a shows the spatial pattern of the first EOF of TCOH for DJF. Because the spatial pattern of the two figures is nearly identical (along with the high correlation between the MEI and temporal component of the first EOF), this indicates that the first EOF is likely ENSO-related for DJF. Similarly, agreement between the spatial pattern of the 1st EOF and El Niño anomaly plots of TCOH for the other

seasons would suggest that they are ENSO-related as well. We have added a supplementary figure (Fig. S5) showing the anomalies for the other seasons. As is expected from the r^2 values of the correlation of the MEI with temporal component of the EOFs, the spatial pattern of the first EOF for SON agrees more closely with the El Niño anomaly plot than do JJA and MAM which show lower correlation.

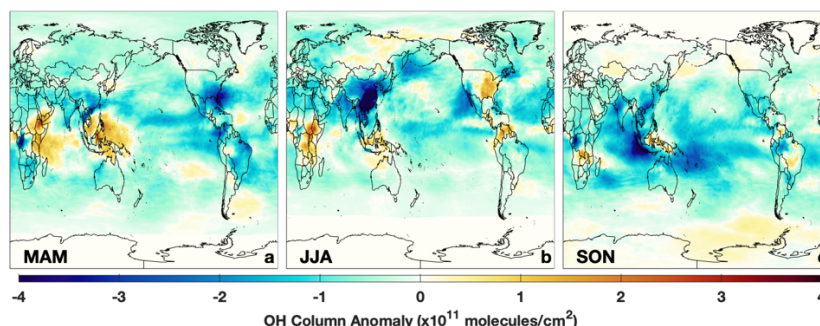


Figure S2: Same as panel a of Figure 7 but for MAM (a), JJA (b), and SON (c).

In addition, we have added the following clarification to the text (Page 15, Line 641-642): “Likewise, the spatial patterns of the first EOF of TCOH for these seasons are similar to the composite figures showing OH anomalies during El Niño (Fig. S5).”

Page 14, line 514 – I don’t understand the plot of satellite NO₂ against the MEI for Fig. 8. You say that “OMI data are insufficient,” but show a plot anyway. If you aren’t going to talk about it, maybe don’t bother showing it. If you are filtering out noise from the NO₂ retrieval, and this leaves you only with NO₂ over land and outflow regions, why do you show ocean values for NO₂ in Fig. 2?

We originally omitted this data because the correlations over the ocean demonstrate significantly more noise for OMI NO₂ than for the other species and are much more frequently below our significance level ($r > 0.5$). We have decided to take your suggestion and include the OMI NO₂ regression, however, because we do find that, despite these limitations, there is still information to be found in the regressions. We have updated the figure, shown below, and include the following text (Page 17, Line 713-720):

Correlations between OMI NO₂ and the MEI suggest similar relationships as found in the MERRA2 GMI simulation, although the correlations are not as robust as for the other satellite variables examined here. This is likely because tropospheric NO₂ column over the ocean are frequently at or below the instrumental average noise (5×10^{14} molecules/cm²). As with the simulation, OMI suggests broad regions of anti-correlation between ENSO and NO₂ in the equatorial Pacific and Gulf of Alaska as well as a region of positive correlation in the extra-tropical NH Pacific. These results demonstrate that, with enough temporal and spatial averaging, OMI is capable of capturing the variability of tropospheric NO₂ even in remote regions with low concentrations.

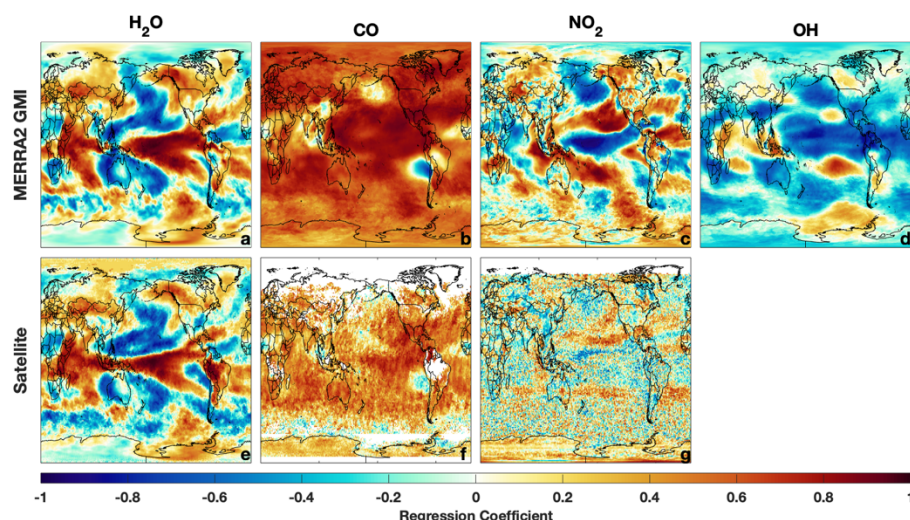


Figure S6 – Should the caption say, “model” NO₂ to differentiate from satellite?

We have removed this figure based on the suggestion of the other reviewer.

Page 14, line 535 – Is there a plot somewhere for the sentence “Changes in the PBL during La Nina...?”

We have added a reference to Fig. 10b to the paper. We have also updated the text to emphasize that these are localized changes. (Page 18, Line 760).

Page 14, line 547 – You might want to consider the mass-weighted concentration of OH more consistently throughout the paper, and possibly the methane lifetime as well. This might provide an additional perspective on how different climate indices change the atmospheric oxidation capacity and address the variability in methane lifetime across models that you describe in the introduction.

We have changed most of the references to percent changes in OH concentration to changes in mass weighted OH concentration when discussing global means. Although we do make occasional references to global or regional changes in OH, our aim is to focus more on the spatial relationship between OH and climate modes. We have included a brief discussion of methane lifetime, which we explain more fully in response to your comment regarding page 24, Line 832.

Page 15, line 545 – According to Table 1, the r^2 with ENSO in JJA is less than 0.25, this should be pointed out.

The 0.25 correlation is actually for the tropospheric column, while the line you reference is discussing the PBL, where correlation is much lower ($r^2 = 0.07$). We have updated the text in the EOF discussion in both the column (Page 15, Line 535-537):

In JJA, ENSO influence on OH is much weaker, with a correlation between the 1st EOF and TCOH of $r^2 = 0.25$, consistent with the seasonal cycle of ENSO.

And the PBL discussion (Page 18, Line 771-773):

In general, the r^2 with ENSO is 0.5 or higher and the mode contributes approximately 10% of the total variance, although correlation in JJA ($r^2 = .07$) is negligible.

Page 15, line 551 – Europe looks like it has decreases not increases, but it is hard to tell with the way the plot is centered. Please double check.

We’ve updated the text to reflect that the sign of the EOF is not constant over Europe, as it is positively signed over portions over the UK, Scandinavia, and other parts of Northern Europe, and negatively signed over other portions of the continent. We now say (Page 18, Line 780) “...net emissions reductions (the United States, portions of Europe, ...)”.

Page 17, line 588 – Do you mean “These increases in jO1D”, not “These increases in O1D?” Also, does the model show that decreased stratospheric ozone is the driver behind the increase in photolysis?

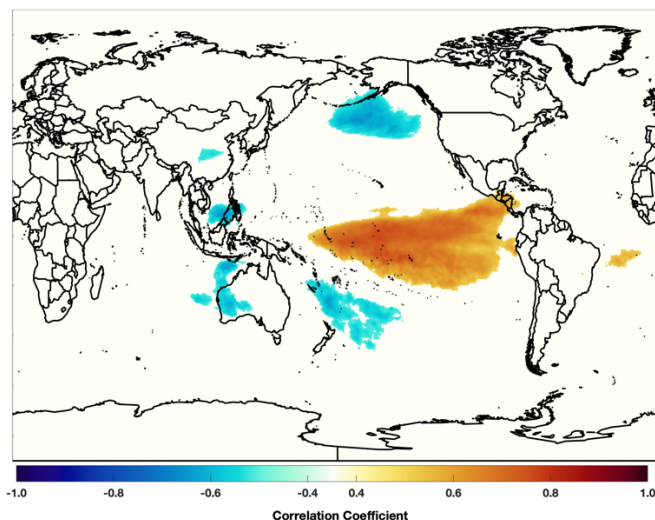
Yes, we meant JO1D. The model does show decreases in total stratospheric column O₃ during El Niño events, which we now note in the paper. Showing an explicit linkage between the decrease in column O₃ and JO1D is beyond the scope of the work. We have refined our language to indicate more uncertainty around the linkage (Page 20, Line 841-844):

The MERRA2 GMI simulation shows reduction in total stratospheric column O₃ of 2-5% in the tropics during El Niño, consistent with previous work (e.g., Randel et al., 2009), which could contribute to the increase in JO¹D, although more work is needed to establish this link.

Page 17, line 621 – It is surprising to me that if stratospheric ozone impacted JO1D in the PBL, it wouldn't also be important in the UFT. Please clarify.

There is positive correlation between JO¹D and ENSO in the UFT, as demonstrated by the figure below, which would suggest increased OH production from this reaction in the upper troposphere. The overall importance of the O¹D + H₂O reaction, however, is much lower in the tropical UFT, where it contributes only about 10% of total OH production, than it is in the PBL, where it contributes 70% or greater of total OH production. Because of this minor importance in the UFT, any changes in production from the O¹D + H₂O reaction are dwarfed by changes from secondary production by the NO + HO₂ reaction, which comprises ~80% of total OH production. We have added the following text to the paper to clarify this point (Page 20, Line 873-889):

While JO¹D does increase in the UFT during El Niño events, as does production from the O¹D + H₂O reaction in some regions, the relatively small contribution of this reaction to the total OH production in the UFT (Fig. Error! Reference source not found.a) does not significantly perturb OH in this layer.



Correlation between JO¹D in the UFT with the MEI.

Page 18, line 638 – I don't think the reference to Fig. S7 is correct, did you mean to refer to a different figure?

Based on the suggestion of the other reviewer, he have removed this figure.

Page 18, line 645 – H₂O(v) is Fig. 8a, not Fig. 8c.

We have made this change.

Page 18, line 646 – It seems that there is a figure, Fig. 16, that you should refer to here in your discussion of lightning.

We have added a reference to what is now Figure 17 on Page 22, Line 919.

Page 19, line 652 – I don't understand your discussion of Turner et al. (2018). If I understand Turner et al. Fig. 4, correctly, there is more lightning during La Nina, but less during El Nino and thus La Nina is positively correlated

with increases in OH. If I understand your Fig. 15 correctly, if NO goes down as MEI goes up, then NO goes up as the MEI goes down, meaning more lightning during a La Nina. This actually agrees with the findings of Turner et al. Please clarify if I am missing something here. If I am wrong, you will definitely need to better clarify your statement about biomass burning and be more specific about how interannual variability in emissions could have changed your results vs. Turner et al., 2018. For example, maybe you need a correlation plot of biomass burning emissions with MEI.

We have overhauled the discussion of Turner *et al* as we agree it was not clearly written. We had intended to convey the idea that additional mechanisms, besides lightning NO production, likely lead to the modeled relationship between NO_y species and ENSO in the equatorial Pacific, where the model shows a narrow equatorial band where NO positively correlates with the MEI (Fig. 17) that overlaps spatially with the region where overall NO (as well as other NO_y species) is anti-correlated (Fig. 16). Outside of the equatorial region, both lightning flash rates and the overall NO concentrations are anti-correlated with the MEI, suggesting decreased NO during El Niño, which is consistent with the decreased OH concentrations, indicating increased NO during La Niña, as shown to be the case when one integrates the LIS/OTD flash count product over the entire tropical region available (Turner et al., 2018). We have updated this discussion as follows (Page 23, Lines 947-965):

This tri-pole correlation pattern between MEI and lightning, evident in both the satellite and model (Fig. 17) is in contrast to the relationship with NO (Fig. 16a) and other reactive nitrogen (NO_y) species in the UFT. While the anti-correlation in NO is consistent with the changes in lightning NO emissions in some regions, in the equatorial Pacific band, NO decreases during El Niño events despite an increase in lightning NO emissions. This apparent discrepancy occurs because even though lightning NO increases by 100% or more over the equatorial Pacific during El Niño events in the model, the absolute difference is orders of magnitude lower than the accompanying changes over land. We conclude that the resulting NO perturbations over the equatorial Pacific latitudes are dominated by mechanism other than the local lightning response, such as changes in the Walker Circulation and the associated transport of air originating over the continents. This mechanism is supported by the similar regression pattern of longer-lived species, such as HNO₃ (Fig. **Error! Reference source not found.**c) and PAN (not shown), to NO in the UFT supports this idea, showing that transport of reactive nitrogen from other source regions, particularly lightning over South America, is likely reduced during El Niño events.

Our findings are broadly consistent with Turner et al. (2018), who found that increases in lightning NO emissions drive increases in OH during La Niña and, conversely, decreases in lightning NO emissions lead to OH decreases during El Niño. The results presented here suggest that in addition to this influence of lightning locally, other mechanisms, such as atmospheric transport of NO_y species, also likely contribute to the relationship between ENSO and OH in the equatorial Pacific.

Page 19, line 667 – It seems that part of the argument is given here. Again, I would suggest that a biomass burning correlation plot or something similar would strengthen your argument. It does seem unlikely though that biomass burning would have such a large impact on the UFT, this definitely needs more discussion.

We agree that further analysis of the role of biomass burning would be helpful. Unfortunately, no biomass burning tracers, such as HCN or tagged CO, were output by the model, so the only biomass burning specific data available are the emissions. So, while we do think this is an interesting question, determining the relative impact of biomass burning on UFT OH during El Niño would require substantial new modeling work that is beyond the scope of this paper.

Page 21, line 740 – What is going on such that EMAC has no correlation in the column, but a very strong correlation in the UFT?

We have avoided addressing the causes of inter-model differences as well as the drivers in the individual CCMI models because a model intercomparison is beyond the scope of this paper. Other work has already made strides in exploring inter-model differences in OH in the CCMI models (e.g. Nicely, 2019 and Zhao, 2019) and we refer the reader to those (Page 6, Line 287). That being said, it's not inconsistent that the first EOF of OH in the UFT is correlated with the MEI and the first EOF of TCOH is not. Because different reactions control OH production at different levels in the atmosphere, it is possible that, in EMAC, ENSO affects OH production from the dominant NO + HO₂ reaction in the UFT while ENSO does not affect OH production reactions in the other atmospheric levels. In

addition, the UFT is only a small portion of the tropospheric column, spanning from 300 hPa to the tropopause, so changes in this level might not significantly perturb column amounts. We have added the following sentence to the paper (Page 25, Line 1108-1111):

While further work is needed to understand the cause of the relationship between OH and ENSO in the UFT in EMAC, results from MERRA2 GMI suggest a role for changes in production via the $\text{NO} + \text{HO}_2$ reaction.

Figure 18 – While you focus on the agreement in your discussion of this figure, the discrepancies are actually quite large considering that all 5 models do not agree on the relationship of ENSO to TCOH in most cases. While you say it is beyond the scope of the paper on Page 22, line 748, given your analysis on the drivers of the ENSO to TCOH relationship from MERRA2 GMI, can you suggest areas of focus that might help us understand the huge model to model variability on OH and methane lifetime that you discuss in the introduction (e.g., Voulgarakis et al., 2013; Nicely et al.,; Zhao et al., 2019)?

As discussed above, we do not want to delve too deeply into the world of inter-model comparisons, but your suggestion here does make sense. We have added the following text to the end of Section 5.5, discussing possible sources of disagreement among the models (Page 26, Line 1128-1132):

Given the results from the MERRA2 GMI analysis, investigating ENSO-related changes in UFT NO, both from lightning and transport, could provide insight into these inter-model differences. Further, Nicely et al. (2020) showed that JO^1D was the largest driver in differences in the methane lifetime in the CCM1 models, suggesting the potential importance of this variable in inter-model differences in the OH-ENS0 relationship in the PBL and lower troposphere.

Page 22, line 767 – It seems like Figure S13 is more appropriate for the main text.

We have moved this figure to the main text as Figure 20.

Page 22, line 768 – Here you do talk about mass-weighted OH. I will just reiterate that it could be extremely useful to calculate changes to mass-weighted OH, or even better, changes to the methane lifetime in El Niño vs. La Niña.

As suggested above, we have increased the inclusion of the mass-weighted OH metric. We discuss the relationship between ENSO and methane lifetime in a response below.

Page 22, line 789 – Can you check the model output during JJA to see whether there is really no interannual variability? It does seem more likely that the model is not accurately capturing the chemical variability within the monsoon anticyclone.

We do not find significant interannual variability in OH above India during the monsoon season, but a full evaluation of this would require defining the location and bounds of the monsoon anticyclone for each year, which we feel is beyond the scope of this work. We have omitted the first part of the sentence, and now simply say (Page 27, Line 1185-1186): “The lack of correlation demonstrated here suggests that the model is not accurately capturing the chemical variability within the monsoon anticyclone”.

Page 23, line 819 – Could this not also be that most models fail to capture the mechanism behind the monsoon impact on OH? If it is due to convectively lofting pollution above the monsoon clouds, then it seems very likely that all models have difficulty with this. Could you look to see whether your model has expected elevated levels of pollution during the monsoon in the UFT that might drive increased OH as described by Lelieveld et al., 2018?

Yes, the models could be missing important transport mechanisms and have included this in the text, which now reads (Page 28, Line 1234-1236) “...the lack of correlation among the models suggests either that those changes are not highly variable from year to year or that not all models capture the mechanisms behind monsoon influence on OH, such as convective lofting of OH precursors.” To examine this more deeply would require a more in-depth analysis (e.g., defining monsoonal circulation) that will further lengthen the paper.

Page 24, line 832 – Could you say something as well about the average decrease in the methane lifetime (using the methane concentrations from the model)?

We have now included a discussion of methane lifetime, but in section 5.4, where we discuss ENSO-related changes in OH in the LFT and MFT. As mentioned earlier, globally averaged changes in mass-weighted tropospheric OH are only ~2% during El Niño events, when compared to neutral events, which corresponds to an approximately 1%

increase in methane lifetime during El Niño events. While we demonstrate throughout the paper that ENSO drives OH variability over much of the globe, when viewed with global annual mean metrics, the variability effectively averages out. So, we do not expand on the CH₄ lifetime beyond the following text (Page 24, Lines 1057-1063):

“The comparatively smaller changes in LFT OH during El Niño events limit the effect of ENSO on the interannual variability of the CH₄ lifetime. Global mean, mass-weighted tropospheric OH decreases by 2.2% during El Niño events, corresponding to only a 1% decrease in the CH₄ lifetime. While changes in OH concentration are most pronounced in the UFT and PBL, CH₄ lifetime is mostly dictated by OH in the LFT due to the temperature dependence of the OH + CH₄ reaction rate. This limited effect on CH₄ lifetime highlights the importance of investigating the spatial OH variability as global mean metrics can obscure important year-to-year changes.”

Page 24, line 844 – This statement about lightning emissions is very important, and definitely needs more support in the prior text. I don’t think that “increased convective lofting of low NO air from near the surface and advection of air with lower reactive nitrogen than during neutral years” has been well supported.

We have updated the text to reflect the clarifications we discussed earlier in reference to our characterization of Turner et al and the relationship between lightning NO and transport of NO in the tropical Pacific. The text now reads (Page 29, Line 1276-1281):

In much of the region, decreases in lightning NO production correspond to decreases in total NO, and thus OH. In the equatorial region, however, increases in lightning NO production are offset by other processes, potentially including transport due to changes in the Walker Circulation. Further work is needed to determine the relative importance of these two factors in controlling OH in the region during El Niño and La Niña events.

Technical Corrections

Page 7, line 288 – “a NMB”, not “an NMB”

Page 15, line 546 – I think it should be “ENSO-related”.

We have made both of these corrections.