We thank the reviewer for their comments, which have helped to improve the manuscript. Responses to comments are shown below in red. Page and Line numbers refer to the track changes version of the resubmitted manuscript.

The manuscript acp-2020-1192 evaluates the drivers of interannual variability in hydroxyl radical (OH) throughout the troposphere. The authors primarily use model output from a 38-year chemical transport model to calculate OH variability and correlate this spatially and temporally with various climate modes, with specific focus on ENSO. They also compare results to other atmospheric chemistry models and where possible data to test the robustness of their conclusions. This is a detailed, careful, and well-presented analysis that adds to our understanding of OH science and should be published in ACP once the comments below have been addressed.

GENERAL COMMENTS

Methane feedbacks: There are known feedbacks between ENSO, methane, and OH. However, according to Section 2.2 (lines 203-204), methane in the model is a fixed boundary condition, which I understand to mean that it wouldn't vary in response to ENSO or other conditions. Therefore some component of the ENSO-driven OH variability may be missing in the simulation (this may be true for other modes too, e.g. Australian monsoon). It would be worth a brief discussion somewhere of this and the implications for the conclusions.

We now include a paragraph at the end of Section 2.2 to discuss how CH4 is treated in the MERRA2 GMI simulation, and the implications for our conclusions (Page 6, Line 269-280):

Because CH₄ is specified as a boundary condition, the model does not capture feedbacks (e.g., wetland or wildfire emissions) between CH₄ emissions and climate modes beyond the extent to which these manifest in the observed methane surface concentrations. ENSO, for example, is known to affect atmospheric CH₄ concentrations through changes in emissions from wetlands (Zhang et al., 2018;Melton et al., 2013) and biomass burning (Worden et al., 2013), although there is uncertainty in the magnitudes of these effects (Melton et al., 2013). On the global scale, however, these ENSO-induced changes in emissions do not significantly perturb background CH₄. For example, during the 1997/98 ENSO event, one of the largest on record, CH₄ grew at a rate of approximately 15 ppbv/yr on top of a background on the order of 1700 ppbv (Nisbet et al., 2016). Because of this small perturbation and the dominance of CO as the primary OH sink over much of the globe (see Section 5.0), it is unlikely that the relationship between climate modes and OH would differ significantly with the inclusion of direct methane emissions in the simulation.

Satellite comparisons (Section 3.2): As far as I can tell, the model comparisons to satellite data have NOT taken into account the retrieval process used in the satellite data (e.g., application of averaging kernels and a priori for CO / shape factors and scattering weights for NO). This introduces an element of bias into the comparisons –even if the model was 100% accurate, the values retrieved by the satellite would not match the model (because the satellite retrievals are not a perfect observing system). While this is an understandable choice for the scale of analysis in this paper, this needs to be clearly discussed. It also means that the comparisons should be considered more qualitative than quantitative (e.g. show of the quoted biases are probably not accurate), and some of the conclusions from this section should be stated less strongly.

We agree that we should have been clearer in the methodology used to compare the model and satellite. For the MOPITT CO comparison, we do convolve the model output with the averaging kernel/a priori from the CO retrieval, so quantitative comparisons between the satellite and model are possible. For OMI NO₂, we have not applied any scattering weights or shape factors. We do not anticipate this leading to major discrepancies, however. The shape factors used for the OMI NO₂ retrieval are derived from a GEOSCCM model run with a similar setup to that described here, including using the GMI chemical mechanism and MERRA2 meteorology. So, differences in the shape factors used in the satellite retrieval and those determined from the MERRA2 GMI run described in this paper will be a minimum. We include the following discussion (Page 9, Lines 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 H_2O , 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.

SPECIFIC COMMENTS BY LINE NUMBER

79-91: The relationships between a variety of climate modes and CO anomalies quantified by Buchholz et al. (2018; https://doi.org/10.1029/2018JD028438) would be relevant to include in this discussion. Also, the last sentence notes observed relationships between NAO and CO long-range transport, but other work has shown CO long-range transport also changes with other climate modes (e.g. with ENSO, see Fisher et al.).

We have added a reference to Buchholz et al. as follows (Page 2, Line 89-100):

In addition to this biomass burning relationship with ENSO, Buchholz et al. (2018) also noted relationships between tropical fire regions and the IOD as well as with the Tropical South Atlantic and Southern Annular modes.

Likewise, we have modified the final sentence of the paragraph to read (Page 2, Line 102-104):

Finally, climate modes can alter the long range transport of CO to the Arctic, through increased outflow from Europe (Li et al., 2002;Creilson et al., 2003;e.g. Duncan, 2004) and Asia (Fisher et al., 2010) for the NAO and ENSO, respectively.

143-162: I suggest including a table that lists all the modes and monsoons included in the analysis and a column that indicates their source (for the modes) or the fact that they are calculated (for the monsoons). Right now I think it's a bit hidden that the monsoons are model-dependent while the modes are not (and I wonder if this has any influence on the fact that you tend not to find agreement between the models in the monsoon analysis in Section 6).

We have included the suggested table in the paper and reproduce it here.

Table 1: Summary of the climate modes and monsoons considered in this work. The index used to characterize the mode, as well as the source of the index, is also indicated.

Mode Type	Index	Mode Type	Index
El Niño Southern	Multivariate ENSO	North Atlantic	EOF of geopotential height at 500 mbar from NCEP reanalysis (NOAA)
Uscillation	Dinala Mada Inden	Oscillation	
Dipole	(NOAA)	East Atlantic	
Asian Monsoon	Model-specific index calculated from the difference of zonal winds in monsoon specific regions	Pacific North American	
South American		East Atlantic	
Monsoon		Western Russian	
North American Monsoon		Scandinavian	
South African Monsoon		West Pacific	
North African Monsoon		East Pacific North Pacific	
Australian Monsoon		Tropical Northern	
Western North		Hemisphere	
Pacific Monsoon		mennisphere	

203-204: More details of the methane concentration implementation would be useful here. What is the source for the methane values? At what latitudinal and time resolution are they input to the model? Are the specified concentrations applied throughout the troposphere, or only at the surface then allowed to advect freely?

We have updated the text to more fully explain the methane constraints in the model. The text now reads (Page 5-6, Lines 250-267):

Methane concentrations are specified at the surface for 4 different latitude bands $(90^{\circ}\text{S} - 30^{\circ}\text{S}, 30^{\circ}\text{S} - 0^{\circ}, 0^{\circ} - 30^{\circ}\text{N}, 30^{\circ}\text{N} - 90^{\circ}\text{N})$ at monthly resolution and advected throughout the troposphere. Methane data are from the NOAA Global Monitoring Division (GMD) surface network (Dlugokencky et al., 1994) and monthly values are interpolated from annual means.

229-231: What biomass burning emissions are used in the CCMI models, and are they consistent with the GEOSCCM biomass burning emissions?

We now include a brief discussion of the biomass burning emissions from CCMI and how they compare to the GFED4s inventory used in MERRA 2 GMI (Page 6, Lines 307 - 313):

Biomass burning emissions are from Granier et al. (2011), which incorporate a modified version of the RETRO inventory from 1980 – 1996 and GFEDv2 from 1997 – 2010 and are based on Lamarque et al. (2010). Monthly-averaged CO emissions from this inventory in Indonesia, where biomass burning emissions are strongly affected by ENSO (e.g., Duncan, 2003a), are highly correlated ($r^2 = 0.79$) in time with the GFED 4s inventory used in the M2GMI simulation. Likewise, monthly-averaged CO emissions over Indonesia from the two inventories agree within 35%, on average.

325: I'm somewhat surprised to see the overestimates in the SH in JJA linked to biomass burning, as this is not peak burning season in the SH, and the spatial pat-terns within the SH continents don't look like the areas of primary dry season burning. Is there other evidence to support the conclusion that this is a biomass burning signal?

We agree with you, and the other reviewer, that biomass burning is likely not the cause of the JJA disagreement. A more likely explanation for the difference is due to errors in isoprene emissions and chemistry. We now say (Page 10, Line 4347-449):

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.

404: How are El Niño "events" defined? And what is the timescale used? I think in some places the terminology is "events" and in other places it is "years" – are there sub-annual events, or is each year classified in its entirety? This also needs addressing in the Figure 6 caption, which describes a difference between "El Niño events and neutral years" – if events are sub-annual, how does this work?

We agree that the term neutral year is confusing and now use "neutral event". In fact, El Niño, La Niña, and neutral events are all defined on the seasonal scale. We now formally define our terminology as follows (Page 4, Line 171-173):

We use monthly values of the ENSO multivariate index (MEI) (Wolter and Timlin, 2011) obtained from https://psl.noaa.gov/enso/mei and averaged to seasonal time scales. Here, ENSO-related events are defined according to the seasonally averaged MEI, where MEI > 0.5 is an El Niño event, MEI < -0.5 is a La Niña event, and an MEI value between 0.5 and -0.5 is a neutral event.

In addition we updated the caption to Figure 7 (formally Figure 6) to reflect the new terminology and changed language throughout the text from neutral years to neutral events.

478: Does the Percent Variance in the table (and quoted throughout the text) refer to the spatial variance? It would be useful if this were clarified somewhere in the text, as it's a little confusing to see such large disconnect between the percent variance explained and the r².

Yes, we have updated all references to variance in the text to reflect that we are talking about the spatial variance.

513-514: "Analysis of vertical winds..." – it's not clear to me how Fig. S6 supports this conclusion (and I find it hard to tell what is going on in Fig. S6). To clarify would probably require more discussion – but as the paper is already very long with a huge number of figures between the main text and the supplement, I would suggest just cutting this one.

We have removed the figure and appreciate the desire to keep the paper a more manageable size.

544: "the 2nd EOF for the PBL" – it would be useful to have an equivalent figure to Figure 7 included in the supplement to show the 2nd EOF for the PBL, since it is the most correlated EOF here.

We agree that this is a good idea. We include the suggested figure, shown below, as Figure S6.



Figure 1: The 2nd EOF of PBL OH from MERRA2 GMI for DJF (a), MAM (b), JJA (c), and SON (d)

In addition, we have added the following text to the paragraph discussing EOFs in the PBL (Page 18, Line 770-771):

The spatial pattern of the 2nd EOF for PBL OH varies markedly across seasons (Fig. S6), with the largest signal over the tropical Pacific during DJF and MAM and over Indonesia in SON.

583-584: This sentence needs a reference

We have included a reference to Oman et al. (2011).

654-656: I am confused and think more discussion is warranted here. The statement is that the difference in the role of lightning NO emissions could be due to "differences in the handling of biomass burning emissions in the two models", but only the MERRA2GMI biomass burning emissions are explained. How were the biomass burning emissions handled in the Turner et al. study? Second, why would the lightning NO emissions change in response to the biomass burning emissions? Or is the argument that the Turner et al. study was missing variability in biomass burning and so incorrectly attributed the variability to lightning NO? Please explain.

We agree, with both you and the other reviewer, that we did not clearly articulate how the results of our work differ from that of Turner et al. 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 positive correlation with lightning NO and the MEI but an anticorrelation in NO (as well as other NO_y species). Outside of the equatorial region, both lightning NO emissions and NO concentrations are anticorrelated with the MEI, suggesting decreased NO during El Niño, which is consistent with the decreased OH concentrations, and increased NO during La Niña. So our work is in agreement with Turner outside the equatorial region. 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.

715: In Fig 18 and other similar figures, I'm not sure of the validity of including the MERRA2 GMI model that is the focus of the paper in the model count. What would be more useful would be to see how many of the independent CCMI models reproduce the relationship seen in MERRA2 GMI. I think this could be done by changing these figures to have only the 4 models in the color scale, but then using e.g. hatching to overlay where MERRA2 GMI had a significant correlation. This would be a clearer comparison of the robustness of the MERRA2 GMI relationships already identified.

Altering Figure 18 (now Fig. 19) to highlight agreement among the different CCMI models is a good idea. We have changed this figure, along with the Figures 19 (now Fig. 21), S16 (now S15), and S17 (now S16) to only include the CCMI models. We have included stippling to indicate the grid boxes where MERRA2 GMI also shows significant correlations between the different variables. We include a sample figure here.



. Number of Models

799-800: "TCOH is anticorrelated with the DMI" - any ideas why?

This is a good question. The positive phase of the IOD is characterized by increased convection and precipitation in the western Indian Ocean and over Eastern Africa as well as a Walker-type circulation over the Indian Ocean basin with anomalous surface easterly winds. This suggests that the region of anticorrelation between TCOH and the DMI in this western Indian Ocean region could result from similar mechanisms as over regions in the Pacific with increased convection during El Niño events. We have examined the correlation between the DMI and the different OH production reactions for each layer as was discussed for ENSO throughout the manuscript. Production from the NO + HO2 reaction is anti-correlated with the DMI in the region in most layers despite a positive correlation with lightning NO emissions, although the r value does not meet our test of statistical significance. So this is suggestive that the Walker-type circulation is potentially driving decreases in NO_X and therefore OH in the region, although further work is needed to prove this definitively. We have updated the text as follows (Page 27, Line 1196-1214):

During the positive phase of the IOD, the Indian Ocean basin exhibits a Walker-type circulation with anomalous surface easterly winds and increased convection in the region that exhibits anticorrelation between OH the DMI. This region is also characterized by an anticorrelation between the DMI and OH production from the NO + HO₂ reaction despite a positive correlation

with lightning NO emissions, analogous to the relationship between NO and ENSO in the equatorial Pacific. This suggests that the anticorrelation between TCOH and the DMI in the eastern Indian is being driven by changes in NO transport from this Walker-type circulation. More work is needed, however, to prove this relationship, as the correlations between OH production from the NO + HO₂ reaction and the DMI do not meet the statistical significance criteria.

TECHNICAL COMMENTS BY LINE NUMBER

29: "Reductions in OH due to ENSO" – please change ENSO to El Niño as ENSO would imply OH is reduced during both phases.

We have made this change.

195: Please specify the version of MEGAN used.

The version of MEGAN used in this model run actually predates version numbers. We have reworded the text to make this point more clear (Page 5, Line 240-242):

Biogenic emissions are calculated online using the method described in Guenther et al. (1999) and Guenther et al. (2000), an early form of the Model of Emissions of Gases and Aerosols from Nature (MEGAN).

263: I would suggest turning this into a 4-panel figure that combines the existing Figure 1 with the existing Figure S2 – the ATom 1 comparison is also useful to see without having to flip to the supplement.

This was also suggested by the other reviewer. We have updated the figure as recommended.

286: "When data from these regions are omitted" – it would be useful to identify the "omitted" data points in the plots shown in Fig. 1 (e.g. with a different shape, filled circle, etc.). We now indicate the data points that are part of the outflow region in the SH with blue stars.

292-293: It would be worth referencing the more recent work of Travis et al. (2020;https://doi.org/10.5194/acp-20-7753-2020) here.

We have added this reference.

319: "boreal winter" - specify months in parentheses.

We have made this change.

376: "In JJA, the climate modes" – please clarify what this refers to...Is it just the NHmodes? All climate modes (NHmodes + ENSO + IOD)? Or all drivers (NHmodes + ENSO + IOD + monsoons)?

We have updated the text to read (Page 12, Line 511) "In JJA, the combination of the different climate modes and monsoons has the smallest spatial coverage..."

407-408: "We focus our analysis on DJF...." – the text would be clearer if this sentence came earlier, before the current sentence starting on line 405 "Figure 5 shows..."

We agree and have changed the order of the sentences.

425: Duplicated 5.1.1, this should just be 5.1

We have made this change.

483-484: "with greater than" - please change to "with NO2 greater than"

We have changed panel g of Figure 8 based on a comment from the other reviewer so now omit this sentence.

518-519: "The dominant OH sink..." – this sentence would fit better with the earlier discussion of Figure 5 and the dominant OH source pathways.

We have moved this sentence to the suggested section (Page 13, Line 568-570).

527: "Same as Figure 6" -> "Same as Figure 6ab"

We have made this clarification.

536: "smaller than in the UFT (Fig. 10)" – it is odd to mention the UFT in this context before it has been discussed. I suggest removing this part and just keeping the comparison to TCOH here.

We have removed the reference to the UFT in this sentence.

557: "Figure 11 shows..." – the wording is unclear both here and in the caption. I think in both cases, the correlation is with the MEI, but in one case it is MEI vs. OH production from H2O+O1D and in the other it is MEI vs. total OH production rate. If this is correct, I would suggest clarifying the sentence to start "Figure 11 shows the correlation of MEI against both OH production from this reaction and the total OH production rate for the PBL."

We have updated the sentence in the text as well as the figure caption for Figure 11 (now Figure 12) to clarify that we are showing the regression between MEI and the reaction rates.

577: Here and elsewhere, JO1D \rightarrow J(O1D)

We have changed the notation here, throughout the text, and in relevant figures.

638: "Fig. S7" - should be "Fig S6." (although I have suggested earlier that this figure be cut)

We have removed this figure from the paper.

We have corrected the grammar here.

644-651: There should be a reference to Fig. 16 somewhere in the text of this section.

We now reference Fig. 16 (now Fig. 17) on page 22 line 919.

708-709: "the CCMI models" – it would be useful to remind the reader here how many CCMI models are being included (4 I think?); e.g. "the 4 CCMI models..." We have updated the sentence to reflect the number of CCMI models used in this study.

we have updated the sentence to reflect the number of Cervit models used in this study.

765-767: I find this long sentence difficult to interpret. I would suggest splitting into two sentences, one about the NAO and one about the PNA.

The sentence now reads (Page 26, Line 1146-1149):

During the positive phase of the NAO, defined as the index being greater than 0.4, TCOH increases by up to 25% in the northern Atlantic. Similarly, during the positive phase of the PNA, TCOH decreases by 10 - 20% in the northern Pacific (Fig. **Error! Reference source not found.**).

774: "vary markedly among the different monsoons (Fig. 4)" – I find this very hard to see from Fig. 4. I would suggest a separate figure in the supplement that shows just the monsoon correlations to aid this discussion. Same goes for the reference to Fig.4b&d in line 786.

We have included a new figure (Figure S11), shown below, that shows just the correlation of the different monsoons with TCOH by season as suggested.



Figure S2: For each season, regions that show a significant correlation (absolute value of r > 0.5) between TCOH and indices for the different monsoons: Australian (Aus.), South African (SAf), South American (SAm), Indian, North American (NAm), North African (NAf), and the Western North Pacific Monsoon (WNPM).

811: Suggest swapping the order of panels b and c to reflect the order in which they are discussed in the text.

We have made this change.

Throughout: If I understand correctly, the main model is sometimes referred to as GEOSCCM and sometimes referred to as MERRA2 GMI - please make the name of the model consistent throughout.

We have clarified the text to indicate that MERRA2 GMI refers to a simulation using the GEOSCCM model with a specific setup. We have removed most references to GEOSCCM except where necessary.