Manuscript # acp-2022-678

Responses to Referee #1

This manuscript details a multi-year ozone tagged contribution analysis. The specific value is the attempt to explain observed trends with trends of model contributions. I think this manuscript has high value, but needs some additional analysis to be published. Below are sections that summarize comments related to model performance, methods, and editorial notes. Last, is a line-by-line section that has more specific feedback.

We thank the editor for all the insightful comments. Below, please see our point-by-point response (in blue) to the specific comments and suggestions and the changes that have been made to the manuscript, in effort to take into account all the comments raised here.

Generally, this manuscript is missing basic model evaluation in the body and supplement. The current manuscript jumps into trends of contributions and only mentions evaluation in the conclusions. In particular, only evaluation in China is ever discussed. The manuscript focuses on trends associated with titration without ever demonstrating the model reasonably captures the phenomenon. The representation of titration in the Eastern US by the model is important given that it drives the trends. Given that coarse models (2x2.5 degree) are often extremely biased at nighttime, the authors should provide some evidence that nighttime titration is reasonably simulated and/or describe how model artifacts may play a role in the trend. Overall, it seems odd that performance over China is used to suggest underestimation of long-range transport while the performance and possible errors of local contributions are omitted.

Response:

We have now added a figure and corresponding descriptions for the model evaluation. The model also tends to overestimate the weakening of NO_x titration in winter, leading to the biases in trends in winter. We also removed the performance for China. Please see our responses below.

In the methods sections, more detail is needed on several fronts. The emissions are currently under-described even though they are well referenced. Recommendations are made in the line-by-line section. Similarly, the model-observation pairing is mostly left to the reader to infer. Again, recommendations are made in the line-by-line.

Response:

Thanks for the suggestion. Please see the line-by-line responses below.

Especially in the conclusions, there are several statements where increase/decrease seem to be used incorrectly. These incorrect directional statements should be corrected. See lineby-line section for specific recommendations.

The section describing the overall trends would benefit from a table. The descriptions and parenthetical references make it somewhat difficult to easily compare. See line-by-line section for specific recommendations.

Lastly, recent application of a very similar system was made by Butler et al. 2020. That publication is referenced, but more should be done to compare these methods and results to those. From a methods standpoint, it would be nice to provide a short summary that explains how these experiments are different. From a results standpoint, you should compare the overlapping 2010 year for comparable. Are the results comparable for overlapping (2010) or proximate years? If not, do methodological differences explain discrepancies?

Response:

Thank you for your suggestion. We corrected statements and added tables to describe overall trends.

We compare the 2010 results of our simulations (Figs. S7–S9) with Butler et al. (2018, 2020) and added the following in the discussion: “Compared with Butler et al. (2018), the simulation in this study shares similar source sector contributions to the zonal average of O_3 concentrations at the surface and 400 hPa in 2010 (Figs. S7 and S8 in this study and Figs. 5 and 6 in Butler et al. (2018)). The contributions from the stratosphere and lightning NO_x are relatively higher in this study than Butler et al. (2018). This may be related to the different anthropogenic emission inventories used, causing different O_3 production/loss efficiencies by natural precursors. When comparing the contributions from different source regions to surface O_3 concentrations in North America, NO_x emissions from East Asia, South Asia, North America, and Europe contributed 2.2, 1.1, 8.3, and 0.7 ppb of the surface O_3 in North America, respectively (Fig. S9) in this study, which are also similar to those from Fig. 4 in Butler et al. (2020). Both studies show the contributions of anthropogenic NMVOCs to surface O_3 concentrations in North America are less than 10 ppb.”

* 117, please describe the depth of the first layer and the number of layers in near the surface (e.g., under 2km). This helps contextualize the model representation of titration later.

Response:

Thanks for the suggestion. We have now included such context as follows: “The height of bottom layer near the surface is about 120 m and there are about 4 layers within2 km.”

* 121, please describe how the stratospheric values are set. Are they based on climatological values? Are they scaled based on something?

Response:

Yes, stratosphere-troposphere exchange of O_3 is treated by setting O_3 to stratospheric values as their climatological means over 1996–2005 at the tropopause (Lamarque et al., 2012), which is affected by atmospheric circulation and experiences the same loss rates as O_3 in the troposphere (Tilmes et al.,2016). We have revised our description in the manuscript.

* 146, are XTR tags really neither NOx nor VOC? Are they included in both?

Response:

This is a special kind of tagging, and its use is usually due to the fact that we cannot attribute it well to the source of the currently running tagging system, none of the reactants belong to the O_x chemical family resulting in no tags can be passed to the O_3. Some examples are as follows: When NO_x is being tagged, the reactions of HO_2 with certain organic peroxy radicals produce O3_X_XTR. A reaction during VOC tagging is the production of the specially tagged species HO_2_X_XTR from the reaction between OH and H_2O_2 (Butler et al., 2018). So XTR exists in both.

* 159, It says CO and CH4 are not tagged by individual sources? Does that mean just by regions? Or, all CO is lumped? The wording is currently unclear. Particularly interested for CO.

Response:

We have clarified as “We does not tag CH_4 by individual sources and its contribution is lumped, because CH_4 is often considered separately from NMVOCs. It has a relative long lifetime in the troposphere and it is well mixed in the troposphere due to its exceptionally low reactivity, which can contribute to O_3 formation at any location in the troposphere where photochemical conditions are favorable (Fiore et al., 2008). CO also has a longer lifetime and lower reactivity than most NMVOCs, separately tagging of CO is more conducive to distinguish its contribution to O_3 from other NMVOCs. Therefore, the lumped total CO is separately tagged in the sector attribution simulations, but the CO is not specifically tagged in the regional attribution simulations due to the computational limit.”

* 164, as you note, this limitation of CO seems odd.

Response:

We have removed this part. Please see our response above

* 168, It would be useful to note here (in addition to later) that TgN and TgC are shown in the appendix.

Response:

Thank you for your suggestion. Added.

* 173, This seems like an important methodological shift. Can the authors highlight whether conclusions are robust to analysis from 1995-2015 or 1995-2019?

Response:

The biomass burning only accounts for a very small amount of total NO_x and NMVOCs emissions (Fig. S1). Therefore, the biomass burning emissions interpolated from SSP2-4.5 forcing scenario should not affect the results.

* 175, Can you clarify what "present-day" means here? Is this a climatology based on a range of "present-day" years or a specific year?

Response:

Clarified as "… and are kept at the present-day (2000) climatological levels during simulations."

* 177, Please elaborate on Price parameterization. I think you are saying online parameterization based on simulated cloud top heights. There are also climatologies based on Price, so it is good to be clear.

Response:

We added a further explanation of the parameterization: "Lightning emissions of NO_x are estimated using online parameterization based on simulated cloud top heights from Price et al. (1997), which is scaled to provide a global annual emission of 3–5 Tg N yr^{-1}."

* 180-185, Please clarify whether the simulation is being sampled only at observation sitedays or averaged seasonally and then sampled at sites.

Response:

The simulation is averaged seasonally and then sampled at the grid boxes of sites. For observations, seasonal mean for any site that has less than 50% data availability in any month of a season is not calculated. O_3 trends at sites is shown only when the data availability is greater than 85% during the analyzed period. Since the observational data are quality-controlled, we don't expect the seasonal average for simulations can largely influence the comparison, but the coarse model resolution may contribute to the biases when comparing with the

observations. We have now added this bias in the discussion.

* 200, The results should start with some estimate of model performance over the target analysis areas. At least, 1) a map of the model with obs scattered on it for an early year and a late year and 2) a description of how basic performance stats change over time. Because this paper focuses on the JJA and DJF, I would expect the model performance to have a similar separation. This will help the readers contextualize results.

Response:

Thank you for the suggestion. We have now added the distribution of observed and modeled surface O_3 in the United States for summer and winter in 1995 and 2019 in Figure S4. We have also included a model evaluation section as:

"Figure S4 compares the simulated near-surface O_3 concentrations with those from observations in 1995 and 2019, respectively. In general, the model overestimates O_3 concentrations in the U.S. in both summer and winter by 10–40%. It can capture the O_3 seasonality that high concentrations in summer and low concentrations in winter. The spatial distributions can also be roughly captured by the model, with statistically significant correlation coefficients between simulations and observations in the range of 0.21–0.45. From 1995 to 2019, the O_3 concentrations in the U.S. decreased in summer and increased in winter presented in observations. The model can produce the sign of the changes, but has large biases in magnitudes, which will be discussed in the following section."

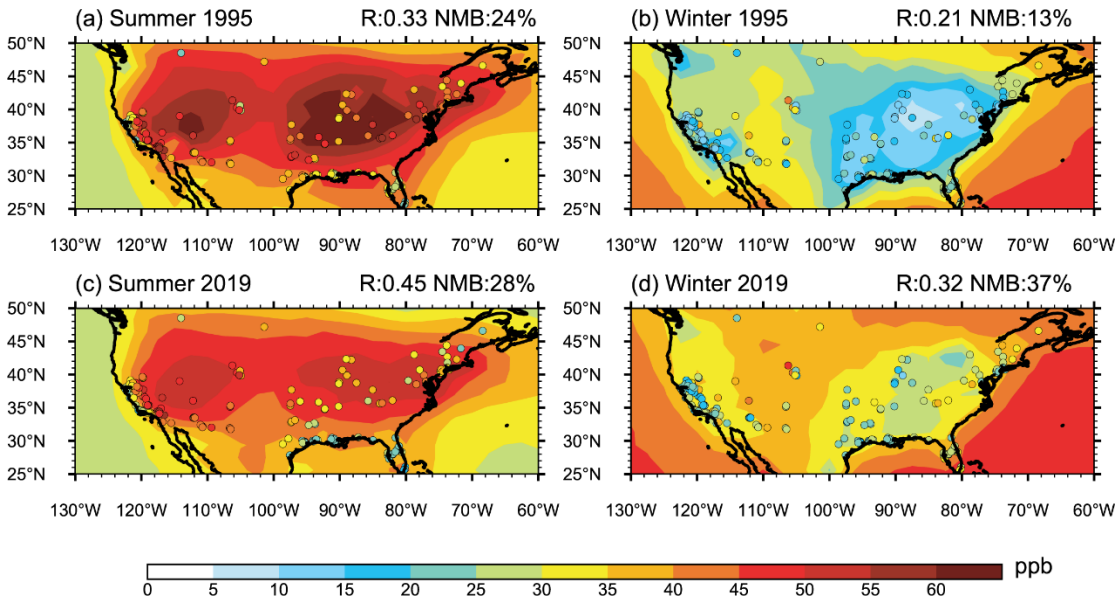

Figure S4. The simulated (contours) and observed (scatters) seasonal mean near-surface O_3 concentrations over the United States in JJA (left) and DJF

(right) and in 1995 (top) and 2019 (bottom). The correlation coefficient and normalized mean bias (NMB, ∑ (Model − Observation) / ∑ Observation× 100%) are shown on top right of each panel.

* 205, Please add lightning NOx in the supplemental figures.

Response:

The model did not output the lightning emission, but the emissions of NO from lightning are scaled to provide a global annual emission of 3–5 Tg N yr^{-1} as Lamarque et. al. (2012).

* 214, related to 180-185, are these trends based on the model only at observation sites or based on averages of the regional "box"

Response:

They are based on the regional grid boxes. The coarse model resolution may contribute to the model biases when comparing with the observations. We have now added this bias in the discussion.

* 214: Looking at Figure 4b, there is a lot of heterogeneity in the western summer trends. The Western cities are fairly isolated leading to misrepresentation by coarse global models. Can you discuss what would happen if you only looked at CASTNet or rural monitors? Or just the IPCC sites?

Response:

I think you were referring to Figure 2b. For some heterogeneity in the summer trends, other studies also found O_3 increasing in Los Angeles and some cities in the central United States, and decreasing in Nevada and Utah based on observations at rural sites (Cooper et al., 2012; Lin et al., 2017). That may explain the strong decreasing trend over western U.S. produced by the model. We have revised the description as that “The decreasing trend over WUS in summer and increasing trend over EUS in winter, however, are largely overestimated in the model, partly attributed to the coarse model resolution.”

* 216-223, I found it difficult to keep the text organized in my mind. I recommend adding a table here.

Response:

We have added Table S1 to show the values.

Table S1. O_3 trends (ppb/decade) over eastern U.S. and western U.S. in DJF and JJA from observations and model simulations.

Season	Source	eastern U.S.	western U.S.
DJF	Observation	2.1 ± 0.29	2.2 ± 0.23
DJF	Model	6.1 ± 0.40	3.2 ± 0.28
JJA	Observation	-3.0±0.41	-0.5 ± 0.42
JJA	Model	-3.0±0.29	-2.3 ± 0.20

It would also be good to add some clarify on what "well produce" means Based on a 95% certainty, the CI are not overlapping for Eastern winter or Western summer. The CIs for Western winter are barely overlapping and only after rounding. The model seems to clearly reproduce the trend only for Eastern summer.

Response:

We have revised the description as “The model reproduces the observed O_3 trend over EUS in summer and roughly captures the O_3 trend over WUS in winter (Table S1). The decreasing trend over WUS in summer and increasing trend over EUS in winter, however, are largely overestimated in the model, partly attributed to the coarse model resolution.”

For me, the titration performance in the East raises questions about the West. The model seems to dramatically overestimate the reduced titration in the East. Given the population density of the East, the titration is likely more widely spread. Due to the population sparsity of the West, the overestimated titration is likely diluted. How does this impact the conclusion about well representing winter in the West?

Response:

Thank you for pointing it out. We agree with the reviewer that the overestimation of O_3 trend in western U.S. in winter could be attributed to the overestimation of the weakened NO_x titration. We have now added a note that “The model also tends to overestimate the weakening of NO_x titration in winter, leading to the biases.”

* 236, I am surprised to see STR (stratosphere) in both NOx and VOC. Is that via XTR?

Response:

The STR tag is neither from NO_x nor VOCs emissions. In both NO_x and VOCs tagging, initial conditions for O_x species in the stratosphere were tagged with STR. In addition, the photolysis of O_2 and N_2O ultimately produces the O_3,

which is all tagged as STR. They can be transported downward via atmospheric circulation and contribute to the near-surface O_3 concentrations.

* 241, can you add error bars to the figure?

Response:

Yes, we have added it.

* 243-247, I think this is a very interesting finding! If the atmosphere is increasingly NOx sensitive, that should have important implications for VOC tagging in later years. Can you discuss that a bit more?

Response:

Thanks for your suggestion, we have added the following: “Note that, during 1995–2019, the molar ratio (mol N /mol C) of emitted NO_x to NMVOCs reduced from 0.11 to 0.07 in the WUS and from 0.14 to 0.07 in the EUS, confirming the enhanced NO_x-sensitive condition during the analyzed time period.”

* 247-251, What role does the location of monitors play in the conclusion here? Is there a strong spatial gradient to the SHP contribution? This is important because the populations tend to be skewed toward near the ocean. In an ideal world, it would be interesting to see a few maps (1995 and 2019) of contributions trends that have a strong spatial gradient.

Response:

Yes, the SHP contribution trends has a strong spatial gradient. We have added a figure below to describe the trend of shipping emissions and O_3 contribution, then modified the shipping-related part as follows:

In recent decades, emissions from international shipping have increased rapidly (Eyring, 2005; Müller-Casseres et al., 2021), but have declined near the coast of the United States. Due to a strong chemical sink associated with photolysis of O_3 with subsequent production of hydroxyl radical (OH) from water vapor in summer (Johnson et al., 1999), the effect of increased emissions of the far-shore ocean on the continental United States was blunted. But the increase in shipping emissions inland tends to increase O_3 concentrations in eastern U.S.

In winter, the decrease in near-shore shipping weakened the NO_x titration, together with the weakened O_3 chemical sink from water vapor in winter, leading to large increasing trends of O_3 by 0.8±0.1 and 1.0±0.1 ppb/decade, respectively, in the WUS and EUS during 1995–2019.

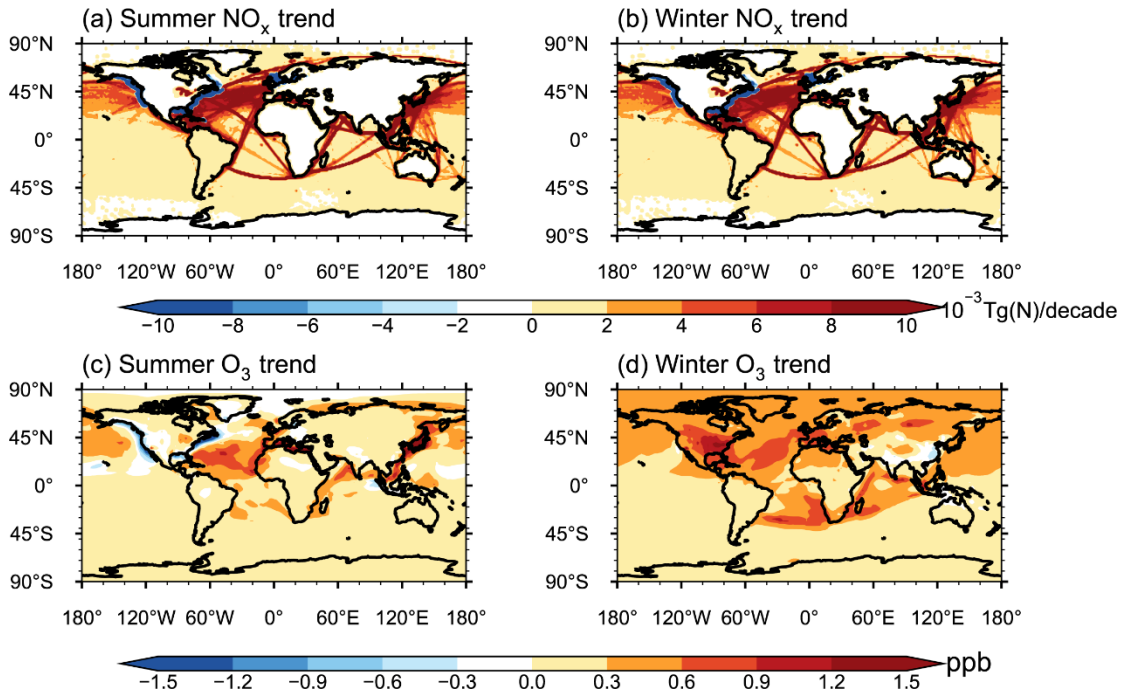

Figure S5. Trends of shipping emissions of NO_x and O_3 trends contributed by shipping emissions in JJA and DJF from 1995 to 2019.

* 259, I find this to be a particularly interesting finding that has implications for the estimation of climate/air quality co-benefit assessments. I wish it was expanded a bit in the conclusions.

Response:

Thank you for the suggestion. We have expanded it in the conclusions as follows:

"Due to the reductions in NO_x emissions, the O_3 production efficiency by reactive carbon species also decreased, leading to the decreasing contributions to O_3 from reactive carbon species in summer during 1995–2019. Even though biogenic NMVOCs emissions and CH_4 concentrations were fixed during simulations, their contributions also decreased related to the weakened O_3 production efficiency by these precursors."

* 272, I find this confusing. Most of this sentence makes perfect sense to me. It is introduced, however, in the context of reduced VOCs. At aircraft heights, you say that only NOx increase. Does that mean that there are no aircraft VOCs? If so, are you suggesting that VOCs at 6-10km were meaningfully reduced and that contributed to the large aircraft trends?

Response:

We apology for the confusing. We were trying to say only the aircraft sector increased and other anthropogenic sectors decreased. By the way, the aircraft mainly emits NO_x rather than NMVOCs. We have rephased the description as

that “Although aircraft NO_x emissions slightly increased, but O_3 attributed to aircraft NO_x emissions shows positive trends as large as 0.4±0.0 and 0.6±0.0 ppb/decade in WUS and EUS, respectively, because aircraft emissions are injected directly into the upper troposphere and lower stratosphere in a low ambient NO_x condition and have a much higher O_3 enhancement efficiency than surface emissions (Hodnebrog et al., 2011).”

* 280-282, Similar comment to earlier. The spatial nature of this enhancement is important. It'd be great to see a map of the contribution and trends.

Response:
Added and please see the response above.

* 283, I don't think this is strictly speaking true. Your lightning emissions are parameterized based on simulated clouds. Can you clarify that this is only true for VOC?

Response:
NO_x emission from the soil is fixed in our simulation, and lightning NO_x varies but is not likely to have a clear trend. All natural VOCs emissions are hold constant during the simulations. We have revised the descriptions as “although most natural emissions do not change during the simulations, …”.

* 289, Butler et al compared January and July in Figure 5. It isn't clear to me that the contribution maximized in DJF vs MAM.

Response:
We have now revised the text as follows: “when stratospheric contribution to the near-surface O_3 is relatively high”.

* 295, Specify anthropogenic and/or that you are excluding soil NOx. Soil NOx in summer has a large anthropogenic component and the contribution from soil is likely "domestic" (e.g. Lapina et al. 2014).

Response:
We revised it to “domestic anthropogenic NO_x emissions (excluding those from soil)”.

* 316, This is definitely interesting... I'm struck however by the dramatic overestimate in the trend, which might be related to the models representation of vertical mixing in winter.

Response:
Thank for your suggestion. We have added this possible explanation in the

conclusion as “The overestimate of O_3 trend in the EUS might be related to a potential biased model representation of vertical mixing in winter”.

* 326-327, The idea that South Asia, and Southeast Asia East Asia "equally contribute" is a somewhat surprising finding. Many previous refereed articles show a decreased transport efficiency from India (S. Asia) to the US compared to East Asia. Similarly, Butler et al 2020 showed significantly larger East Asia contribution than South Asia. Can you highlight why your results would be so different?

Response:

Here the result shows their contribution to the O_3 trends not to O_3 concentrations. The reason for this phenomenon may be due to China's rapid reduction of NO_x emissions in industry and energy in recent years. But we also noticed that the model can not well simulate the O_3 trends in China, which deserves further investigation in our future work.

* 335-350, This discussion really highlights the oversimplicity of linear trends. The authors do a good job noting this is likely associated with transport. It would be good if they connected it to known meteorological cycles. A quick look shows that ENSO cycles are likely accounted for by the time averaging, but the Pacific Decadal Oscillation (PDO) is not.
This highlights why the trend is likely not significant. It is likely made up of ups and downs seen in the PDO. A 5-year average of the NCEI PDO index shows that the winters of these two periods are of opposite signs (despite inter annual variability). This is in part because in mid 1998, the PDO index shifted. This leads to a smaller difference between summers than winters. You could also reference the Lin et al. paper about the position of the jet stream.

Response:

Thanks for your suggestion. We have now included such context as follows:” The horizontal and vertical transport of O_3 together contribute to the near-surface O_3 increases in winter during 1995–2019 associated with the changes in large-scale circulations. The anomalous atmospheric circulation is likely linked to the location of the midlatitude jet stream, which is influenced by ENSO cycle (Lin et al., 2015).”

* 342, I think it is a mistake to call the comparison of two five year periods "anomalous".

Response:

This is the difference between two 5-year averages. So we guess the “anomalous” could be used.

* 355-357, I think you got the signs of change wrong here. You showed decreasing in the summer (controls) and increasing in the winter (lessening titration).

Response:

Yes, we have corrected it now.

* 355-357, You showed that it could only replicate the decreasing trend in the eastern summer and the increasing trend in the western winter. You showed that the trends for Eastern winter and western summer were *not* well captured. The trends were significantly different. So, it is wrong to say that it did well in the conclusion.

Response:

We corrected the expression as follows: “This model can capture the O_3 decreasing trend over the EUS in summer and increasing trend over the WUS in winter during this time period, but largely overestimates the decreasing trend over WUS in summer and increasing trend over EUS in winter.”

* 359-361, You need to be clear when you are talking about the model regions and when you are talking about the observed sites as sampled by the model. Are these trends at select sites? Are these trends representative of the larger region? Or the population weighted concentrations?

Response:

We have clarified as that “In summer, our simulation results show that…”.

* 364, This is a little less clear to me. The VOCs were also reduced. How do you distinguish between reduced VOC trends and reduced NOx OPE impacts on VOC trends.

Response:

Because the biogenic emissions contribute the largest to the O_3 decreasing trend in summer, but they are kept constant during the simulation. Therefore, the O_3 decrease in summer is dominated by the reductions in NO_x emissions.

* 391, This was a 3.7 ppb/decade decrease (not increase).

Response:

Corrected.

* 392-393, The authors are offering only one of several equally permissible explanations. As you note, one is that the Asian trends are underestimated. Another is that the coarse model overestimates the decrease associated with

domestic reductions. Another is that stratospheric variability is underestimated. Another is that the trend in ships contributions are underestimated. What makes the authors confident that this is the only hypothesis to highlight?

Response:

We agree many reasons can lead to the overestimate of the decreasing trend over WUS in summer by the model. Lin et al. (2017) found that the contribution from increasing Asian emissions offset that from the U.S. emission reductions, but our results showed the Asian contribution only offset a small amount of the domestic emission reductions. So we suspect that the "the Asian contribution to the O_3 trends in WUS is likely underestimated in this study". We did not say the overestimate of the decreasing trend over WUS is due to the bias in Asian contribution alone.

* 393-396, 1) The authors should show this model performance if their conclusions rely upon it. 2) The local peaks in China will depend on near surface vertical structure while the continental scale outflow may not. So, you could only say that it "consistent with the low contribution from Asian sources" since you cannot say that it definitively explains anything.

Response:

To avoid misunderstanding, we have not removed this sentence from the manuscript.

* 396-397, It is unreasonable to think that model evaluation of China is worth discussing, while model evaluation over the US is not.

Response:

Yes, we have now added model evaluation over the US. Please see the response above.

Reference:

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