Manuscript # acp-2022-678

Responses to Referee #2

The manuscript of Li et al. investigates trends of ground-level over the US for 1995 – 2019. They use a source attribution method (tagging) to attribute the ozone trends to trends of emissions of VOCs and NOx from different sectors and regions.

The topic of the manuscript is very interesting and it fits into the scope of ACP. However, the manuscript needs major revisions before it can be reconsidered for ACP.

General remarks:

1) The authors report that during winter time ozone increases due to NOx titration. They use, however, a rather coarse resolved global model. It is well known that at this resolution NOx titration is usually underestimated by the model. Therefore, I agree with referee #1 that a more detailed model evaluation is needed. The authors should also discuss how the ability of the model to capture the observed trends only partly (e.g. increasing trend in EUS in winter is strongly overestimated) influence the conclusions. Further, it would be interesting to analyse if the model is able to capture the chemical regimes in EUS and WUS correctly.

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."

The overestimation of O_3 trend in western U.S. in winter could be attributed to the overestimation of the weakened NO_x titration, related to the coarse model resolution. 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."

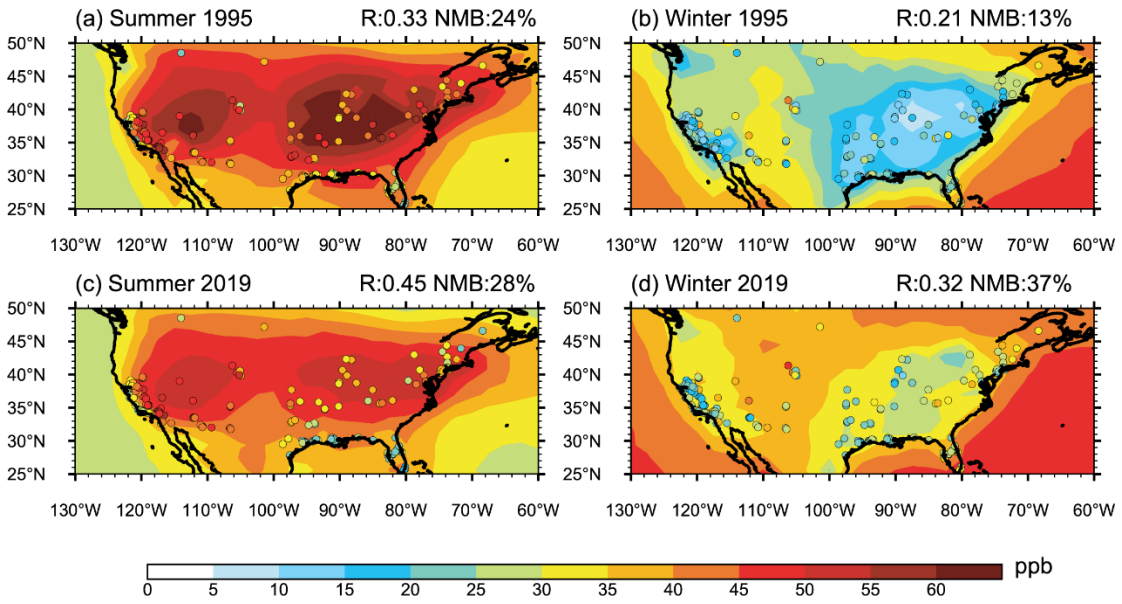

Figure S4. The simulated (contours) and observed (scatters) seasonal mean near-surface O_3 concentrations over the United States in JJA (left) and DJF (right) and in 1995 (top) and 2019 (bottom). The correlation coefficient and normalized mean bias (NMB, ∑ (Model − Observation) / ∑ Observation× 100%) are shown on top right of each panel.

As to how the ability of the model to capture the observed trends only partly influence the conclusions, we have now added sentence as follows: "Lin et al. (2017) found that the contribution from increasing Asian emissions offset that from the U.S. emission reductions, resulting in a weak O_3 trend in WUS. In this study, the Asian NO_x emissions only contribute to 0.6 ppb/decade of the total positive trend in WUS in summer, much lower than the 3.7 ppb/decade decrease attributable to the domestic emission reductions, suggesting that the Asian contribution to the O_3 trends in WUS is likely underestimated in this study. The bias of O_3 simulation in China may also lead to a bias in the wintertime O_3 trend over EUS. Additionally, international shipping can have a disproportionately high influence on tropospheric O_3 due to the dispersed nature of NOx emissions (Butler et al., 2020; Kasibhatla et al., 2000; von Glasow et al., 2003), together with the weakened NO_x titration, resulting in the overestimation of O_3 trends. The fixed CH_4 concentration during simulations also biased the modeled O_3 trends in this study. The coarse model resolution also contributed to the biases. The overestimate of O_3 trend over EUS in winter, likely related to the bias in NO_x titration, implies the overestimate of source contributions to the trends in magnitude."

Although determination of the chemical regime is typically made according to the indicator ratio P_{H2O2} / P_{HNO3} (the ratio between the production rates of

hydrogen peroxide and nitric acid), our simulations did not output these two variables. Alternatively, we show here the ratio of NO_x to NMVOCs emissions to support our results as 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."

2) Even though the CEDS emissions are well documented by Hosely et al., 2018 the authors should discuss these emissions in more detail as the results of the study heavily depend on the emission inventory. How do the trends in the emissions of Hosley et al., 2018 for example compare to the trends in McDuffie et al. 2020, the CAMS or the EDGAR emissions? What is the influence of the inconsistency in the aviation emissions in CEDS (Thor et al., 2022) on the results?

Response:

Thank you for the suggestion, we have added the following discussion:

"As the results of the study heavily depend on the emission inventory, here the potential bias in emissions are also discussed. Compared with the previous CEDS version used in this study (hereafter $CEDS_{Hoesly}$), the updated CEDS inventory (hereafter $CEDS_{GBD\text{-}MAPS}$) (McDuffie et al., 2020) incorporates updated activity data. For NO_x, the global emission from $CEDS_{GBD\text{-}MAPS}$ is smaller than that of $CEDS_{Hoesly}$ after 2006 and shows a fast decreasing trend. By 2014, global emission of NO_x is about 10 % lower than the $CEDS_{Hoesly}$ estimate. These differences are mainly reflected in the industrial and residential sectors in China, followed by the transportation sector in India and Africa. For global emission of NMVOCs, which remains relatively unchanged between the $CEDS_{Hoesly}$ and $CEDS_{GBD\text{-}MAPS}$ inventories (Fig. 6 in McDuffie et al. 2020). The global NO_x emission from EDGAR v4.3.2 inventory is less than $CEDS_{Hoesly}$ (Crippa et al., 2018). This difference in NO_x emissions may reduce O_3 trends in U.S. from foreign contributions, especially from East Asia. Recent study also reported a difference in NO_x emission distribution between CMIP5 and CMIP6 related to an error in data pre-processing in CEDS, leading to a northward shift of O_3 burden in CMIP6 (Thor et al., 2023). The aviation emissions should be corrected in future studies of O_3 simulations."

3) The labels and fonts in many figures are too small. All of the labels/fonts (and also the station symbols in Fig 2.) needs to be enlarged.

Response:

Thank for you suggestion. We have made corresponding adjustments.

4) The model description misses a lot of basic information. Even though the CAM4-chem model is well known, most important information should be given

in the manuscript. As example detailed information about the chemical mechanism is missing. How is dry and wet deposition represented? What variables are nudged? Further I am missing information about the emission totals for natural emissions (lightning NOx, biogenic VOCs and soil NOx) as well as the global emissions. All of these information are important to compare results from different studies with each other. Of course not all of these things need to be discussed in detail. Some are also fine in the supplement (for example detailed information about the emissions).

Response:

Thank you for the suggestion. We have now revised the text as follows:

"The model configuration uses a comprehensive tropospheric chemistry mechanism based on the Model for Ozone and Related chemical Tracers version 4 (MOZART-4) (Emmons et al., 2010, 2012). Model configurations simulate wet deposition of gas species using the Neu and Prather (2012) scheme. Dry deposition is represented following the resistance approach originally described in Wesely (1989). 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)… The zonal and meridional wind fields are nudged towards the MERRA-2 reanalysis."

In addition, we have provided the details of the information of the total global emissions in the supplement.

Table S2. Global total emissions of NO_x, NMVOCs and CO for different sectors in 1995 and 2019.

	year	AGR	ENE	IND	RCO	SHP	TRA	SLV	WST	BMB	SOIL/BIO	AIR
NO_x	1995	1.23	9.19	4.24	3.16	5.11	11.34		0.39	4.48	7.98	0.67
TgN/yr	2019	1.61	7.83	5.25	2.53	6.02	11.26		0.74	4.00	7.98	1.19
NMVOCs	1995	4.86	24.99	8.27	31.16	2.59	35.97	23.32	2.70	64.28	664.87	
TgC/yr	2019	7.60	35.51	11.63	28.93	3.15	25.30	31.90	2.76	61.99	664.87	
CO	1995		15.97	42.42	113.09	0.21	102.52		3.81		68.51	0.19
TgC/yr	2019		28.17	40.41	97.63	0.33	55.63		7.25		68.51	0.31

5) The authors need to clarify the units they use. They use ppb as unit which sounds like (volume/mass?) mixing ratios but use the term concentration throughout the manuscript. Please clarify if you consider mixing ratios or concentrations. Also, for the emissions totals the authors should specify what NOx and NMVOCs are (see below for more details).

Response:

We have now clarified parts per billion (ppb, volume ratio in this study) and described NO_x and NMVOC with TG N and TG C, respectively. The mixing ratio is sometimes expressed as concentration in many studies, so we prefer to keep it as it is.

6) In the model simulations CH4 mixing ratios are kept fix at 1750 ppb. This represents ~ 1990 levels (https://gml.noaa.gov/ccgg/trends_ch4/). Until 2019 CH4 levels have been increased to ~ 1880 ppb which is a increase of ~7--8 %. This increase influences ozone production and Butler et al., 2020 found very inhomogeneous changes of the contributions by CH4 increases. Therefore, I suggest to perform an additional simulation with an CH4 increase.

Response:

This is a good suggestion to quantify the contribution of increasing CH_4 to O_3 trend. However, it takes several months for one long-term simulation, which beyond our current computational resource. We have added a note that "It is noticed that the fixed CH_4 mixing ratio during simulations also biased the modeled O_3 trends in this study, which deserves further investigation with the varying CH_4 levels in future studies."

7) The authors should reconsider the choice of tagging labels. In my opinion the region North America should have been spitted into US and Canada. Further, important information are lost because the shipping emissions have the ROW tag in the "region tagging" runs. Why are they not tagged as shipping/oceanic in the "regional tagging" runs? Further, I wonder why the results of many "unchanged" sectors changed between the "sector" and "region" tagging runs. Shouldn't the results for the tags "STR", "LGT", "AIR", "SOIL" be identical in Fig 5. and 7? If only anthropogenic sources get either sector or region tags the results of the natural sources should not change? In my view this is a very critical inconsistency which needs to be clarified (maybe I also don't understand the approach correctly). In addition, the authors should motivate the special category for CO in more detail. Many information are lost by lumping all CO emissions in one category.

Response:

For the choice of tagging labels, we referred to the HTAP Tier 2 receptor regions. As to the ROW tag in the "region tagging" runs, according to Fig. 1, which includes emissions from the Arctic and Antarctic as well as southern Africa, Oceania, and northern Asia, not just the oceans. We would like to add more tagging labels, but the tagging system crashed when more tags were added, which should be addressed in the next version of the code.

Thanks for pointing the tag issue, the results for the tags "STR", "LGT",

"AIR", and "SOIL" should be similar between sector and regional runs. The larger discrepancies for these tags in last version are due to an error in the division of emissions used in our model and it has been corrected now.

For the CO tagging, we have clarified as "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."

8) The manuscript lacks a detailed discussion of the results in comparison to other global source attribution studies. Are the results in accordance with other studies? For example are results of O3 from STR, lightning or biogenic sources comparable with other studies? Much more comparison with previous work is needed (see below for some references; in addition also Guo et al. 2017 .and all the studies from the HTAP framework (https://htap.org/) can be of interest here). In my opinion also the introduction needs to be improved (see detailed comments below).

Response:

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

We have now compared the 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."

9) I am wondering about the trend of O3 due to aircraft emissions. Usually most aircraft emissions take place in the (upper) troposphere and not near ground-level. Therefore I wonder if there is a trend of O3 from aviation (check values in the upper troposphere) or if there is an increase in downward transport. If so, it would be interesting to separate effects due to increased emissions and due to changes in dynamics.

Response：

We have now added a figure and the following to illustrate the O_3 trend contributed by aircraft emissions as “Although aircraft NO_x emissions slightly increased, but O3 attributed to aircraft NOx 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). It can be confirmed that the NO_x from aircraft contributes to the increase in O_3 concentrations at 250 hPa in high latitude regions of the Northern Hemisphere during 1995–2019 (Fig. S6).”

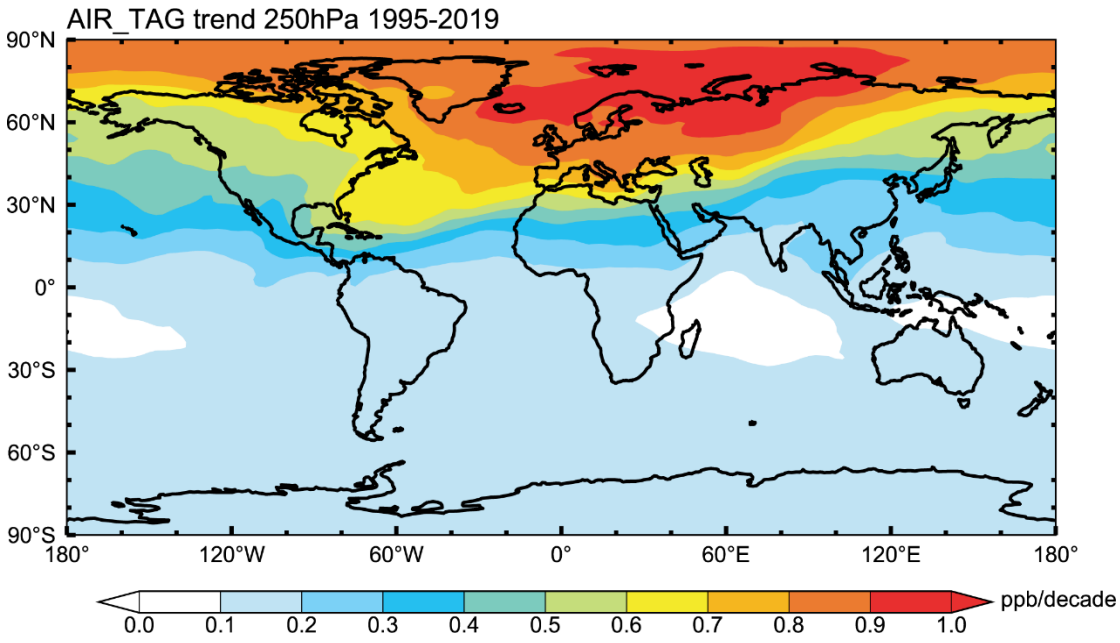

Figure S6. Annual O_3 trends contributed by aircraft at 250hPa from 1995-2019.

Detailed comments:

p4l70-p4l87: This section needs some corrections. The perturbation approach and labeling techniques are two different methods answering different scientific questions. The perturbation approach provides (potential) impacts. Tagging provides contributions. There is many literature discussing this issue which can be checked for more details– some are: Grewe et al. 2010, Emmons et al. 2012, Clappier et al. 2017 and Tunis et al., 2020.

Response:

We have now corrected the description as “One method of obtaining an O_3

source-receptor relationship is to zero out or perturb emissions from a given source region or sector in sensitivity simulations along with a baseline simulation, which gives information about the response of O_3 to changes in precursor emissions." "The tagging approach produces information about the contribution of precursor emissions to the total amount of O_3 (Butler et al., 2020). The perturbation and tagging methods are two different methods answering different scientific questions, with the first for the impacts and the last for the contributions (Grewe et al. 2010, Emmons et al. 2012, Clappier et al. 2017 and Thunis et al., 2019)."

p4l91: This is not correct. There are approaches applied on the regional scale which use chemical indicators (Dunker et al., 2012, Kwok et al., 2015). However, there are also approaches on the regional scale which do not use chemical indicators (e.g. LupaÅ□cu and Butler, 2019; Mertens et al. 2020)

Response:

Thanks for the suggestion. We revised the description to "In some regional models, O_3 apportionment is based on the ratio of chemical indicators to determine the regime of O_3 generation (e.g., VOC-limited or NO_x-limited regimes) and then attribute the generation of O_3 to the tag carried by a certain precursor (VOCs or NO_x), which however cannot simultaneously attribute O_3 production to NO_x and VOCs, respectively (Dunker et al., 2002; Kwok et al., 2015), while some models do not use the chemical indicators (Lupaşcu and Butler, 2019; Mertens et al., 2020)."

P5l97: I think this heavily depends on how the boundary conditions are implemented (see literature above).

Response:

Here we are trying to say the regional model can not separate the regional contributions from several source regions outside the domain. We have now clarified the description.

P5l100ff: There are also global approaches which use a sector wise attribution or a combination of sector wise and regional attribution (Emmons et al. 2012, Grewe et al. 2017, Butler et al. 2018). Butler et al. 2018 includes a comprehensive overview of different approaches which the authors could check.

Response:

Yes. Here we would like to highlight that some global models directly tag the O_3 production rather than the precursor emissions. These literatures have been cited in the manuscript.

P6L120: Is O3 is nudged at the tropopause towards 'stratospheric' values?

What happens with the stratosphere tagged tracer?

Response:

We have revised the description to “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).”

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.

P7l152ff: See also general comments above. Why not tagging shipping emissions separately as Ocean (see Butler et al., 2020). What about aviation in this list. If I understand the analysis correctly aviation has been tagged as sector in the regional runs?

Response:

“ROW” includes emissions from the Arctic and Antarctic as well as southern Africa, Oceania, and northern Asia in addition to oceans. Due to the limitation of the number of tagging (causing model crash), we merged shipping emission into ROW tag.

Yes, aviation has been tagged as sector in the regional runs. Because aircraft emissions covers both land and ocean regions, it is not so reasonable to determine the regional contribution of aviation emissions only by the emissions over the region.

P7l164: See also general remarks. Is this the explanation why CO is lumped?

Response:

We have clarified as “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.”

p7l168: What about emissions of SO2 and NH3?

Response:

They are also from CEDS. Now added.

P8l177: Please specify the lightning NOx total emissions?

Response:

Specified as "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} as Lamarque et. al. (2012)."

P9l208-p9l225: See also general remarks. For summer EUS many stations show an increase of O3. It seems that this is not captured by the model? For winter EUS stations some stations show no or even a decreasing trend. These trends are not captured by the model. Please comment.

Response:

Yes, in summer some studies found O_3 increased in Los Angeles and some sites in the central United States, decreased in Nevada and Utah (Cooper et al., 2012; Lin et al., 2017). The coarse grid resolution of the global model cannot accurately capture the differences between cities or sites, but the relatively stronger O_3 reduction trend in the central WUS region (Nevada and Utah) is still well reproduced on the regional scale. It is the same reason for winter. We have added the explanation in the manuscript.

Fig 3 : Why do VOC emissions of the ENE sector increase while NOx emissions decrease? Please comment and compare the trend of ENE with other emission inventories. Please specify if emissions are Tg(N), TG(NO) etc. (also for VOCs)

Response:

Thank you for your suggestion. We now specify NO_x as TG N and VOC as TG C and modify the corresponding figures.

Over the past 10–20 years in the US, the reduction in coal-fired power plant emissions have resulted in emission reductions in NO_x (Krotkov et al., 2016; Duncan et al., 2013; Castellanos and Boersma, 2012; de Gouw et al., 2014). Over this same time period, however, oil and gas production in key regions in the US has more than tripled between 2007 and 2017 (EIA, 2020), which resulted in VOC emissions increasing.

P10l235: Fig 4 and 5 should be reordered; same for 6 and 7.

Response:

Reordered.

P10l246: A more detailed analysis of the change of the O3 production efficiency would be very valuable here.

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.”

P10l247: See also general remarks: Information about the trends of global emissions (e.g. shipping etc.) would be very valuable here.

Response:

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.

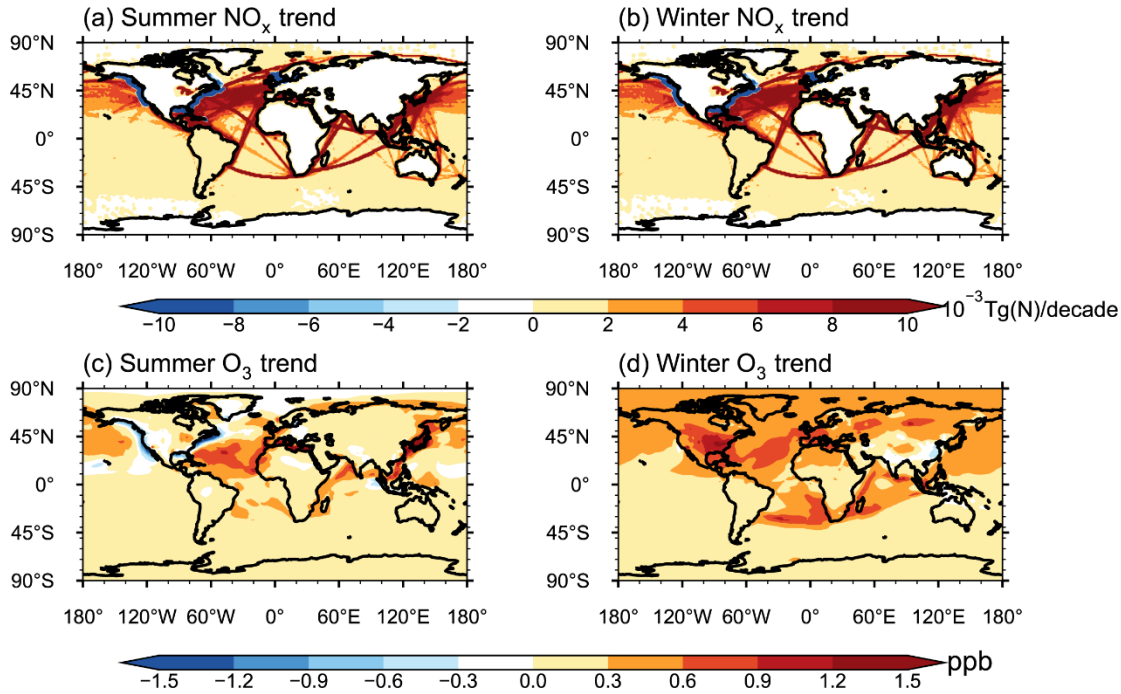

Figure S5. Trends of shipping emissions of NO_x and O_3 trends contributed by shipping emissions in JJA and DJF from 1995 to 2019.

P10L257ff: Could you please explain the argumentation here in more detailed? I think additional analysis would help here to make this point more clear.

Response:

O_3 decreases in summer over the U.S. due to the reductions in NO_x emissions. At the same time, the VOCs do now have significant decrease, that leads to a decrease in O_3 production efficiency by VOCs. We have revised the sentence as follows: "This does not actually mean that CH_4 and biogenic NMVOCs themselves contributed to the overall O_3 trend through changing the precursor levels since they were kept constant during simulations; rather, mainly due to the reductions in NO_x emissions, O_3 production efficiency by reactive carbon species decreases, leading to decreasing trends of O_3 contribution by CH_4 and biogenic NMVOCs."

P11l278ff: This sentence is very long. I suggest to split it up.

Response:

Changed.

P12l295: See general remarks. Why did you applied one combined tag for North America and not separate tags for US and Canada?

Response:

The choice of tagging labels we refered to HTAP Tier 2 receptor regions and the tagging system will crash when more tags are added.

P13l318: I don't understand the sentence. Please explain. Thanks!

Response:

The combined natural source means LGT, STR and SOIL. In winter, with the reductions in domestic anthropogenic NO_x emissions, the weakened NO_x titration process leads to an increase in the O_3 production efficiency from these sources.

P13L334: The figure suggest that none of the results are significant (by the way; with which method did you check for significance. Please explain in detail were appropriate).

Response:

Based on our MET experiments, it is significant in the central U.S. in winter (Fig. 8b). We calculate linear least-squares regressions for O_3 and years,

considering the P<0.05 to be significant.

Section 3.4: See general comments above. More details/analysis are missing here. In my opinion especially an analysis of the contributions would be very valuable here. How large is for example the trend of ozone from the stratosphere due to changes in the dynamics.

Response:

We supplement the trend of O_3 contribution from each emission source to the U.S. in winter in Fig. 8 and added more details as follows: "Variations in the circulation facilitate O_3 transport from upper altitudes to the surface, as well as foreign contributions from Asia, which is consistent with the finding in Lin et al. (2015). The O_3 increasing trend in winter over the U.S. attributing to stratospheric injection and Asian NO_x emissions due to dynamics are both 0.2±0.1 ppb/decade (Fig. 8e). Therefore, changes in anthropogenic emissions are the main factor affecting O_3 trends."

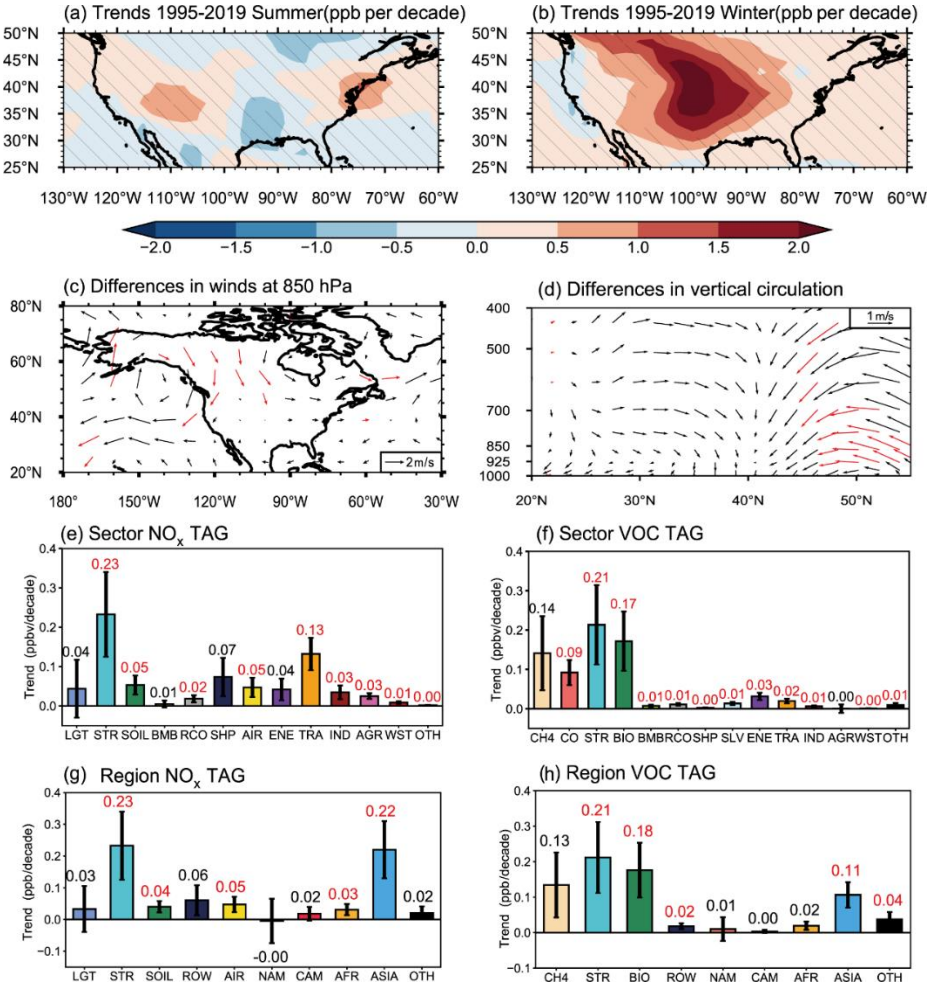

Figure 8. Linear trends (ppb/decade) of simulated (a) JJA and (b) DJF mean near-surface O_3 concentrations during 1995–2019. Differences between the

first (1995–1999) and last (2015–2019) five years during 1995–2019 (last–first) in DJF mean (c) 850 hPa horizontal winds and (d) meridional winds and vertical velocity averaged over 90–105°W. Areas without hatches in (a) and (b) and red arrows in (c) and (d) indicate statistical significance with 95% confidence. All results are from the MET experiments. Linear trends (ppb/decade) of near-surface O_3 concentrations in winter over the U.S, contributed by the NOx (e, g) and reactive carbon (f, h) emissions from various source sectors (e, f) and regions (g, h). The increasing and decreasing trends marked with red and blue color numbers, respectively, indicate statistical significance with 95% confidence. Contributions from source regions EAS, SAS and SEA are combined to ASIA. Some sources having small contributions are combined and shown as OTH.

P14l355: I don't agree with the statement that the model captures the trends 'well'. Please rephrase.

Response:

We change the expression to "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."

P15l374ff: What is the reason for the increase of shipping and aviation – emissions or dynamics? Please analyse in more detail.

Response:

According to the newly added Fig. 8e and Fig. 5, the O_3 increase attributing to shipping and aviation NO_x emissions due to dynamics are 0.07 and 0.05 ppb/decade, respectively, much lower than 0.76/0.99 and 0.43/0.55 ppb/decade in the default simulation. Therefore, changes in emissions are the main factor.

Reference:

Castellanos, P. and Boersma, K. F.: Reductions in nitrogen oxides over Europe driven by environmental policy and economic recession, Sci. Rep.-UK, 2, 265, https://doi.org/10.1038/srep00265, 2012.

Clappier, A., Belis, C. A., Pernigotti, D., and Thunis, P.: Source apportionment and sensitivity analysis: two methodologies with two different purposes, Geosci. Model Dev., 10, 4245–4256, https://doi.org/10.5194/gmd-10-4245-2017, 2017.

Crippa, M., Guizzardi, D., Muntean, M., Schaaf, E., Dentener, F., van Aardenne,

J. A., Monni, S., Doering, U., Olivier, J. G. J., Pagliari, V ., and Janssens-Maenhout, G.: Gridded emissions of air pollutants for the period 1970–2012 within EDGAR v4.3.2, Earth Syst. Sci. Data, 10, 1987–2013, https://doi.org/10.5194/essd-10-1987-2018, 2018.

de Gouw, J. A., Parrish, D. D., Frost, G. J., and Trainer, M.: Reduced emissions of CO2, NOx, and SO2 from US power plants owing to switch from coal to natural gas with combined cycle technology, Earths Future, 2, 75–82, https://doi.org/10.1002/2013EF000196, 2014.

Duncan, B. N., Y oshida, Y ., de Foy, B., Lamsal, L. N., Streets, D. G., Lu, Z., Pickering, K. E., and Krotkov, N. A.: The observed response of Ozone Monitoring Instrument (OMI) NO2 columns to NOx emission controls on power plants in the United States: 2005–2011, Atmos. Environ., 81, 102–111, https://doi.org/10.1016/j.atmosenv.2013.08.068, 2013.

Dunker, A. M., Yarwood, G., Ortmann, J. P., and Wilson, G. M.: Comparison of Source Apportionment and Source Sensitivity of Ozone in a Three-Dimensional Air Quality Model, Environ. Sci. Technol., 36, 2953–2964, https://doi.org/10.1021/es011418f, 2002.

EIA: US Energy Information Administration: Drilling Productivity Report, available at: https://www.eia.gov/petroleum/drilling/, last access: 7 April 2020.

Emmons, L. K., Hess, P. G., Lamarque, J.-F., and Pfister, G. G.: Tagged ozone mechanism for MOZART-4, CAM-chem and other chemical transport models, Geosci. Model Dev., 5, 1531– 1542, https://doi.org/10.5194/gmd-5-1531-2012, 2012.

Fiore, A. M., Dentener, F. J., Wild, O., Cuvelier, C., Schultz, M. G., Hess, P., Textor, C., Schulz, M., Doherty, R. M., Horowitz, L. W., MacKenzie, I. A., Sanderson, M. G., Shindell, D. T., Stevenson, D. S., Szopa, S., Van Dingenen, R., Zeng, G., Atherton, C., Bergmann, D., Bey, I., Carmichael, G., Collins, W. J., Duncan, B. N., Faluvegi, G., Folberth, G., Gauss, M., Gong, S., Hauglustaine, D., Holloway, T., Isaksen, I. S. A., Jacob, D. J., Jonson, J. E., Kaminski, J. W., Keating, T. J., Lupu, A., Marmer, E., Montanaro, V., Park, R. J., Pitari, G., Pringle, K. J., Pyle, J. A., Schroeder, S., Vivanco, M. G., Wind, P., Wojcik, G., Wu, S., and Zuber, A.: Multimodel estimates of intercontinental sourcereceptor relationships for ozone pollution, J. Geophys. Res., 114, D04301, https://doi.org/10.1029/2008JD010816, 2009.

Grewe, V., Tsati, E., and Hoor, P.: On the attribution of contributions of atmospheric trace gases to emissions in atmospheric model applications, Geosci. Model Dev., 3, 487–499, https://doi.org/10.5194/gmd-3-487-2010, 2010.

McDuffie, E. E., Smith, S. J., O'Rourke, P., Tibrewal, K., Venkataraman, C., Marais, E. A., Zheng, B., Crippa, M., Brauer, M., and Martin, R. V.: A global anthropogenic emission inventory of atmospheric pollutants from sector- and fuel-specific sources (1970–2017): an application of the Community

Emissions Data System (CEDS), Earth Syst. Sci. Data, 12, 3413–3442, https://doi.org/10.5194/essd-12-3413-2020, 2020.

Mertens, M., Kerkweg, A., Grewe, V., Jöckel, P., and Sausen, R.: Attributing ozone and its precursors to land transport emissions in Europe and Germany, Atmos. Chem. Phys., 20, 7843–7873, https://doi.org/10.5194/acp-20-7843-2020, 2020.

Krotkov, N. A., McLinden, C. A., Li, C., Lamsal, L. N., Celarier, E. A., Marchenko, S. V ., Swartz, W. H., Bucsela, E. J., Joiner, J., Duncan, B. N., Boersma, K. F., V eefkind, J. P ., Levelt, P . F., Fioletov, V . E., Dickerson, R. R., He, H., Lu, Z., and Streets, D. G.: Aura OMI observations of regional SO2 and NO2 pollution changes from 2005 to 2015, Atmos. Chem. Phys., 16, 4605–4629, https://doi.org/10.5194/acp-16-4605-2016, 2016.

Kwok, R. H. F., Baker, K. R., Napelenok, S. L., and Tonnesen, G. S.: Photochemical grid model implementation and application of VOC, NO_x, and O_3 source apportionment, Geosci. Model Dev., 8, 99–114, https://doi.org/10.5194/gmd-8-99-2015, 2015.

Lin, M., Fiore, A. M., Horowitz, L. W., Langford, A. O., Oltmans, S. J., Tarasick, D., and Rieder, H. E.: Climate variability modulates western U.S. ozone air quality in spring via deep stratospheric intrusions, Nat. Commun., 6, 7105, https://doi.org/10.1038/ncomms8105, 2015.

Lupaşcu, A. and Butler, T.: Source attribution of European surface O3 using a tagged O3 mechanism, Atmos. Chem. Phys., 19, 14535–14558, https://doi.org/10.5194/acp-19-14535-2019, 2019.

Thor, R. N., Mertens, M., Matthes, S., Righi, M., Hendricks, J., Brinkop, S., Graf, P., Grewe, V., Jöckel, P., and Smith, S.: An inconsistency in aviation emissions between CMIP5 and CMIP6 and the implications for short-lived species and their radiative forcing, Geosci. Model Dev., 16, 1459–1466, https://doi.org/10.5194/gmd-16-1459-2023, 2023.

Thunis, P., Clappier, A., Tarrason, L., Cuvelier, C., Monteiro, A., Pisoni, E., Wesseling, J., Belis, C., Pirovano, G., Janssen, S., Guerreiro, C., and Peduzzi, E.: Source apportionment to support air quality planning: Strengths and weaknesses of existing approaches, Environ. Int., 130, 104825, https://doi.org/10.1016/j.envint.2019.05.019, 2019.

Wesely, M. L.: Parameterizations for surface resistance to gaseous dry deposition in regional-scale numerical models, Atmos. Environ., 23, 1293–1304, https://doi.org/10.1016/0004-6981(89)90153-4, 1989.