Responses in Red.

References to line/paragraph numbers are provided with respect to the previous preprint (available online) and not the revised version.

Text Changes in Blue.

The changed text includes references to figures and tables from the revised manuscript.

Dear Editor, Reviewers and Readers,

While checking our simulations during the preparation of our response to the reviews of our manuscript, we discovered an error in our emissions. Due to unit conversion errors in both the emissions preprocessing and emissions checking routines, our simulations had approximately 50% larger NOx emissions than they were supposed to. We have corrected the NOx emissions and performed our simulations again with the corrected emissions. We have updated our revised manuscript with the new results. We now discuss the changes from our previous model run where appropriate in our responses to the comments of each reviewer.

In general, the corrected simulation have ~33 % smaller anthropogenic NOx emissions than previously reported, but with similar sign of trends. This has resulted in smaller tropospheric O3 burden, surface ozone and CH4 oxidation rate, but a larger ozone production efficiency of NOx sources and CH4 lifetime. The sign of trends in contributions from each of the tagged sources of NOx have mostly remained similar to what was previously reported. We have therefore retained the major structure of our manuscript as previously submitted and corrected the results wherever applicable.

Apart from this, we also made several changes in the revised manuscript mainly related to the grammatical and technical correctness after proof-reading as recommended by the reviewers, not all of which are shown in this responses document. However, we tracked all the changes to the revised manuscript that acts as a good reference to all the changes incorporated into the manuscript.

Comments by Anonymous Referee 1:

This manuscript details an application of tagging technique to the attribution of tropospheric ozone changes, to identify the contributions of regional and sectoral NOx/RC emissions. The authors present a detailed calculation to demonstrate the influence of the equatorward shift of surface anthropogenic NOx emissions, and how the ozone burden and its trend are contributed by emissions from different regions and sources. The manuscript is well-organized and written. Overall, I think

this is a neat study and fits the scope of ACP. However, certain aspects deserve further discussion before publication.

We thank the referee for the positive comment and valuable feedback on our manuscript.

Major comments:

1) The equatorward shift of precursor emissions is a key point to make in this study but is mainly supported by references and inference in this study, for example, line 475. It is important to give more direct evidence that more precursors or O3 itself have been lifted into the free troposphere over past years (e.g. trends of tagged surface anthropogenic NO_x in the free troposphere), and it is worth further discussing whether the increase of tropospheric ozone burden (largely in the free troposphere) is mainly contributed by the lifted precursors or the lifted O3 based on your tagged simulations, if possible.

We agree that a more direct evidence of O3 itself or precursors being lifted into the free troposphere would be important to establish the predominant role of convection. Therefore, at line 291, we now add another paragraph:

"To illustrate the predominant role of convection in transporting ozone and it's precursors at the tropics into the free troposphere, we show the zonal mean vertical profiles of annual mean O3 and NOx (sum of NO and NO2) mixing ratios for the 2000-2018 climatological mean attributed to NOx emitted from a typical tropical region: Southeast Asia and a typical northern mid-latitude region: Europe (Fig. 6; see Fig. S1 for the location of defined regions).

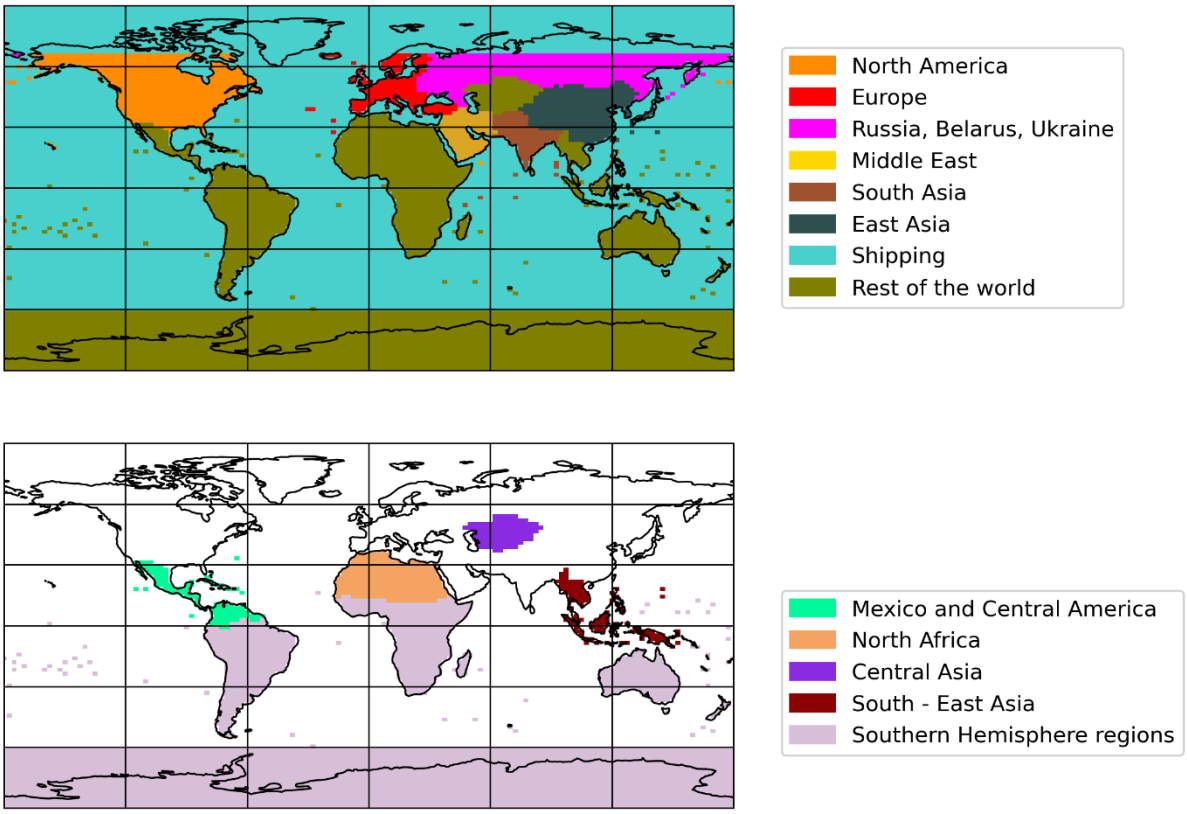


Fig. S1: a) Regions considered as regional anthropogenic tags in our simulations (NOx and RC tagged), b) Explicitly tagged regions within the "Rest of the World" tag in our NOx-tagged simulation.

The vertical gradient in the ozone attributed to European NOx is consistent with production of ozone primarily within the boundary layer with subsequent vertical transport into the free troposphere. While there is indeed some ozone attributed to Southeast Asian NOx emissions present in the boundary layer, the mixing ratio of this attributed ozone is much higher in the free troposphere, which is consistent with ozone production aloft. This is due to emitted NOx directly being transported aloft, eventually leading to free tropospheric ozone production. Further, we also note the increasing trend in NOx burden attributed to anthropogenic NOx emissions in the free troposphere (above 700 hPa; Figure S6)"

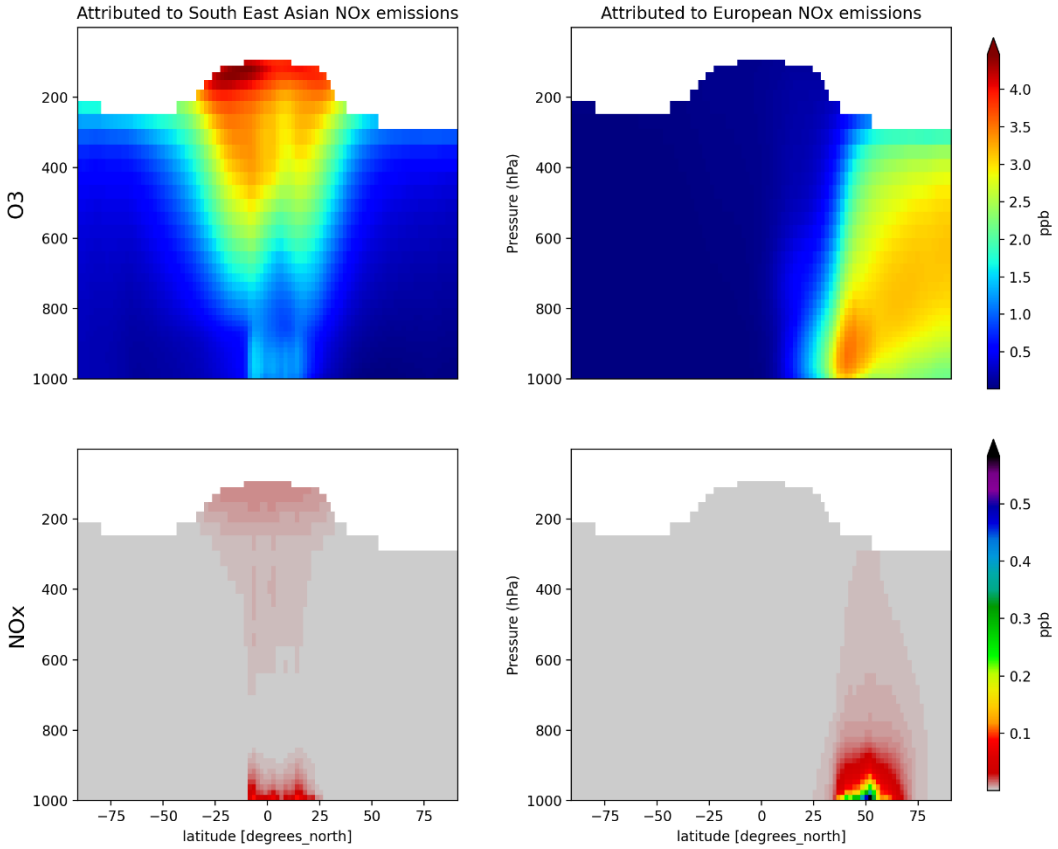


Fig. 6: Vertical profiles of zonal climatological (2000-2018) mean mixing ratio within the troposphere: O_3 (top panels) and NO_x (bottom panels) attributed to Southeast Asian (left panels) and European (right panels) anthropogenic NO_x emissions.

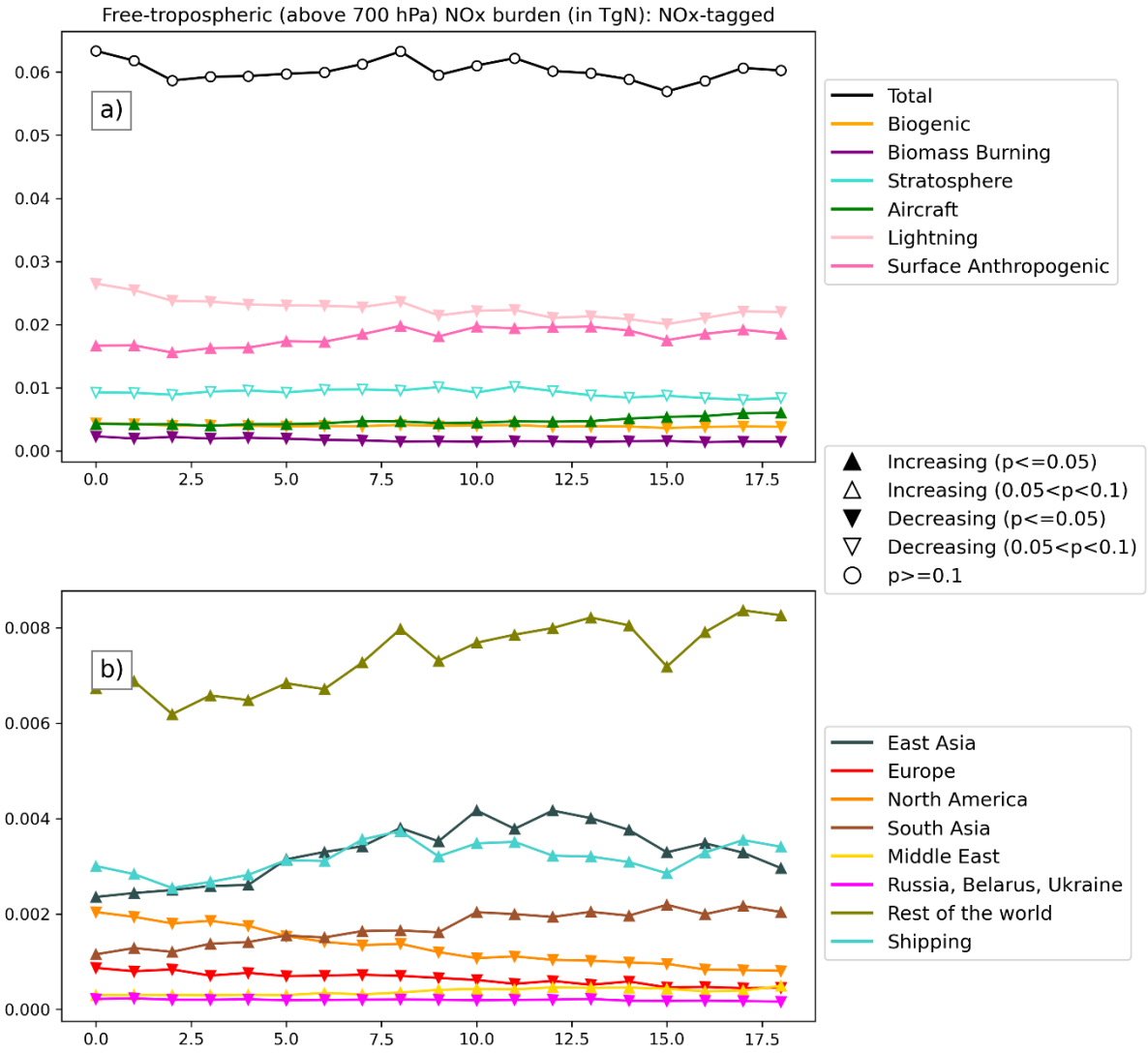


Fig. S5: Timeseries of free tropospheric NO_x burden (above 700 hPa and within the troposphere; in TgN) over the 2000-2018 period. Shown are the total simulated free tropospheric NOx burden and the contributions from the NOx-tagged simulation. Bottom panels are the regional contributions to the total anthropogenic component (dark pink line) shown in the top panels. The symbols indicate the sign and certainty of trend in the plotted quantity.

We now include in section 4 at line 520: "We show using vertical profiles of tagged O3 and NOx mixing ratio fields (Fig. 6) that it is the emitted NOx in the tropics which predominantly gets lofted into the free troposphere and eventually form ozone. Our tagging method only conveys information on the precursor source of the simulated ozone and associated gas molecules. It, however, does not convey information about where exactly the ozone molecule was formed: in the free troposphere, or within the boundary layer. While addressing this is beyond the scope of our study, further studies could employ ozone tagging method used in Sudo and Akimoto (2007) and Derwent et al., (2015) that tag/label ozone molecules based on the location at which the ozone molecules are formed."

2) For East Asia, when discussing the trend, it makes no sense to discuss the whole period, and the two periods (prior-2011 versus post-2011) should be separated.

We now replace the discussion on the 2000-2018 trends related to East Asian emissions and associated quantities with the corresponding trends for the split periods (2000-2011 and 2011-2018). These quantities have been included in Tables S2 (NOx-tagged) and S6 (RC-tagged).

In section 3.1, we replace line 199 with: "Among the anthropogenic NOx emissions, East Asian emissions are the largest (~20 %; Table 2), with an increasing trend with high certainty (~0.25 TgN/yr2;Table 3 and Fig. 3) over the 2000-2018 period. These emissions increase at 0.49 TgN/yr2, peak in 2011 and start declining after that at -0.44 TgN/yr2 (Zheng et al., 2018), as shown in Fig. 3c and Table S2, which largely explain the timeseries of anthropogenic and total NOx emissions timeseries shown in Fig. 3a.", and line 236 with "Among the anthropogenic NMRC emissions, East Asian emissions are the largest (~95.36 TgC/yr (6.78 %); Table 4), increasing at 3.54 TgC/yr2 peaking in 2011 and decreasing after that at -2.82 TgC/yr2(Fig. 3(d); Table S6) as for the NOx emissions."

In Section 3.2.1, we add the following line before the sentence at line 300: "Similarly, the magnitude of the percentage slope in tropospheric ozone burden attributed to East Asian NOx emissions for both the pre-2011 (2000-2011) and post-2011 (2011-2018) periods is smaller compared to the corresponding percentage slope in East Asian NOx emissions (Table S2)."

Further in section 3.3, we replace the lines starting from 369 until the end of the paragraph with: "Fig. 7c shows that the OPE of East Asian NOx has a trend of low certainty over the 2000-2018 period. However, when shorter periods are considered, it decreases with high certainty during the pre-2011 period when emissions increase (2000-2011 period) and increases with high certainty for the post-2011 period when emissions decrease (2011-2018 period; Table S1)."

3) It is confusing to me how the stratospheric influx is tagged that it could be attributed to NOx and RC. In line 255, the tropospheric ozone is attributed to oxidation of N2O, but if NOx-tagged means tagging the NOx emissions, how could it tag the NOx produced from N2O? It would be clearer if the authors clarify the tagging technique in more detail. Also, as changes of stratosphere-to-troposphere exchange are also important for the trends of tropospheric ozone burden (especially the free troposphere), please include the discussion of STE in the summary and conclusion, and also abstract.

We have added some text to the end of the paragraph beginning at line 255 to clarify the confusion that could possibly arise: "As described in Section 3.1.2 of

Butler et al. (2018), ozone production in the stratosphere is handled directly in the chemical mechanism with the addition of new reactions producing the relevant odd oxygen species. Similarly, production of NO in the stratosphere from the oxidation of N2O by O1D is also handled directly with an additional chemical reaction."

We also clarify this by adding a footnote in Table 1: "**NOx-tagged simulation attributes ozone to influx from the stratosphere (same as RC-tagged), and also to NO produced from oxidation of stratospheric N2O"

We also include in the abstract: "These tags include various natural (biogenic, biomass burning, lightning NOx and RC from methane oxidation), and regional anthropogenic (North American, European, East Asian, South Asian etc.) precursor emission sources, and influx from stratosphere." We do not further discuss results related to STE contribution in the abstract, mainly due to word limitation.

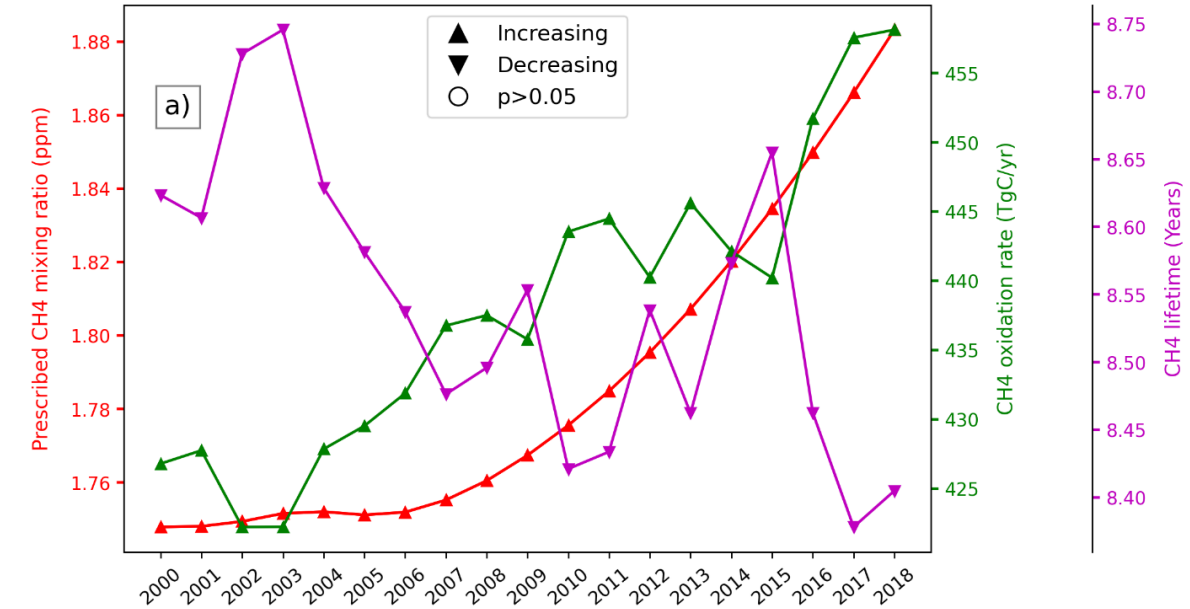
In section 4 we extend the sentence at line 512: "We simulate the largest contribution to tropospheric ozone burden from anthropogenic NOx emissions (in our NOx-tagged simulation) and reactive carbon from methane oxidation (in our RC-tagged simulation), both with significantly increasing trend, followed by contribution of ozone influx from stratosphere but with no significant trend in its contribution."

4) Line 231. The methane lifetime trend is inconsistent with some other studies that suggested a longer lifetime of methane due to the reduction of OH. The simulated OH trend should also be shown in Fig.4 and be compared to other model studies to give a better sense of the accuracy of the model mechanism in this study and give a better reasoning for the methane lifetime decrease.

We now recognize that our smaller values of CH4 lifetime compared to previous studies was due to the incorrectly calculated larger NOx emissions. We corrected the NOx emissions and performed the simulations again. We now also update our method of calculating the tropospheric methane lifetime (total atmospheric methane burden divided by the tropospheric methane oxidation rate). Therefore, our new simulations have a CH4 lifetime of ~8.54 years, which is closer to the range simulated by previous studies, as opposed to the previously simulated ~6.5 years. We now add the timeseries of OH in Fig. 3, and replace the last sentence in this paragraph with:

"CH4 oxidation with OH radical being the major loss pathway for CH4, we show the time-series of OH in the bottom part of Fig.3. Here we simulate an increasing trend in air-mass-weighted tropospheric OH concentration (a prominent indicator for tropospheric oxidizing capacity; e.g., Voulgarakis et al., 2013, Chua et al., 2023) but with low certainty. Nevertheless, the CH4-reaction-weighted tropospheric OH

concentration (Lawrence et al., 2001) shows an increasing trend with high sensitivity. This increasing trend in tropospheric OH concentration is consistent with the trend discussed in Chua et al., (2023) and explains the decreasing lifetime of CH_4 as these quantities are inversely proportional. The increasing OH availability despite increasing CH4 concentration could be due to increasing NOx emissions over the simulated period that recycle HO2 to OH (e.g. Lelieveld et al., 2008; Chua et al., 2023). The mean magnitude of our air-mass-weighted tropospheric OH concentration (~12.63 * 10^5 molec/cm^3) is slightly larger than previous studies (~10.75 * 10^5 molec/cm^3 in Chua et al., 2023 or ~11.7 * 10^5 molec/cm^3 in Voulgarakis et al., 2013) which is likely why we simulate a mean CH4 lifetime slightly smaller than the aforementioned studies (~9 years), but within the multi-model range (7.1-10.6 years) reported in Voulgarakis et al., (2013)."



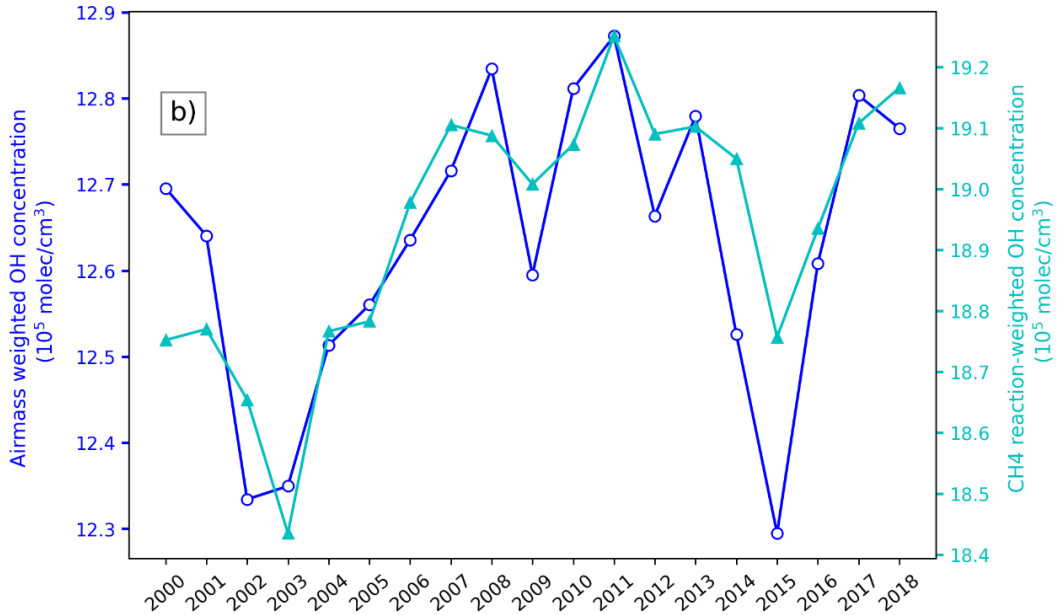


Fig. 4: a) Red line: Annually varying Methane concentration (in ppm) prescribed in our model. Green line: Methane oxidation rate (in TgC/yr) simulated by our model. Magenta line: Lifetime of Methane (in years). b) Airmass weighted and CH4-reaction weighted tropospheric OH concentration (in 10^5 molec/cm^3). The symbols on the plotted timeseries indicate the sign of trend in the plotted quantity. The mean values, slope of trend, 95 % confidence interval and p-value for the 2000-2018 period are provided in Table S5.

Minor comments:

1)Ozone enhancements in the free troposphere are linked to the monsoon-induced transport. The changes in monsoon and other meteorological factors (i.e. climate noise) may also affect the amount of lifted surface ozone/precursors, and the they may show a trend under global warming. This should be mentioned that the simulated trends are not only contributed by changes in precursor emissions.

We further add to the discussion in section 4, continuing from the addition mentioned in response to Major comment 1): "Our study highlights the dominant effect of equatorward shifting of O3 precursor emissions which contributes to the tropospheric O3 burden trend. Nevertheless, there remain other climatic factors that may play an amplifying or offsetting role to these contributions. These include, for example, changes in the general circulation of the atmosphere and monsoonal changes under a warming planet, as well as the natural variability of climate."

2) The authors showed the emissions from different regions and pointed out an equatorward shift. It would be better to give a quantified number of how the total emissions in Tropics versus Midlatitudes change over time, to quantify that the decrease of emissions in East Asia after 2011 won't alter this phenomenon.

We now conclude the paragraph at line 199 with: "We further illustrate this equatorward shift using zonal sum profiles of deviation in anthropogenic NOx emissions from year 2000 (Fig. S4a). The zonal NOx emissions relative to year 2000 show a positive (negative) deviation south (north) of ~35°N latitude, indicating an equatorward shift in the overall global NOx emissions."

Similarly, we conclude section 3.1.2 with the sentence:" We also see an equatorward shift in global anthropogenic NMRC emissions, similar to the anthropogenic NOx emissions in our zonal sum profiles of deviation in anthropogenic NMRC from year 2000 (Fig. S4b)."

The decrease in East Asian emissions after year 2011 won't alter this phenomenon as the East Asian region is largely situated north of 30°N, as shown in Fig. S1 above.

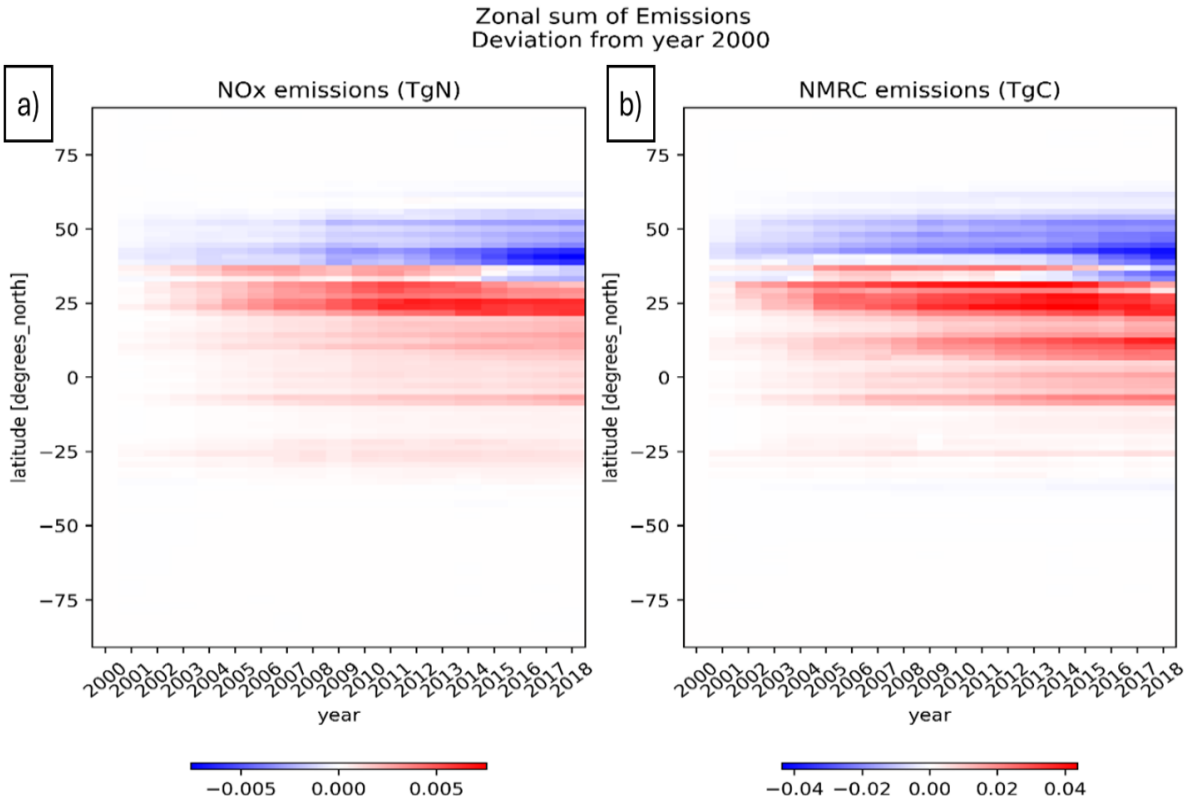


Fig. S4: Deviation from year 2000 in the zonal sum of annual anthropogenic emissions of a) NOx and b) NMRC emissions.

3) Line 40. “or” is a typo.

Corrected

4) Line 172. The comparison of simulations to ozonesonde uses different periods. The authors should also show the results of the overlapping period (2000-2010) to give a better sense of how the model accuracy is.

We now show the comparison for only the overlapping period (2000-2010) rather than the whole period (2000-2018). We also include the vertical profiles of comparison in climatology in the supplementary information as a response to a comment by Referee 2. We now conclude section 2.2 as: “The model captures the vertical distribution of ozone derived from ozone sonde climatology very well, although slightly biased high particularly in the upper troposphere at most sites (Fig. S2). Our free troposphere evaluation results are therefore largely consistent with previously evaluated versions of CAM4-Chem (Tilmes et al., 2012, Zhang et al., 2016, Emmons et al., 2020).”

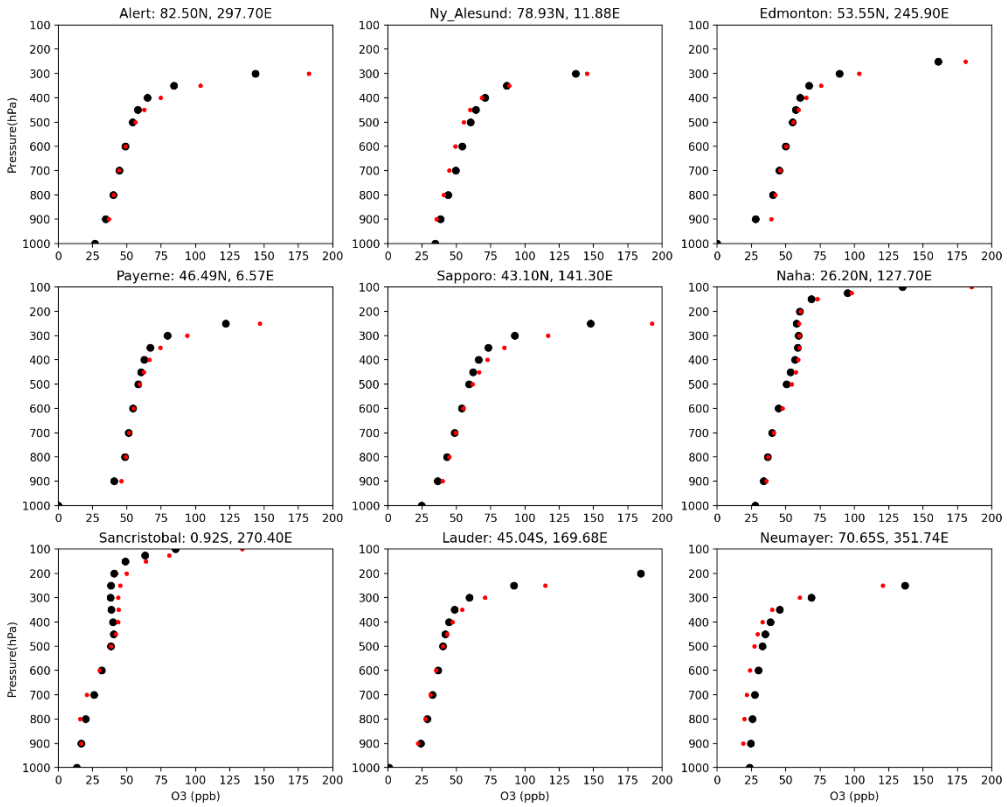


Fig. S2: Vertical profile comparison of annual mean tropospheric O3 mixing ratio at nine ozonesonde stations (coordinates stated as the title) from the model (red dots) for the 2000-2010 period and ozonesonde climatology (black dots) for the 1995-2010 period. The nine selected ozonesonde stations are latitudinally representative across the northern and southern hemispheres, as evaluated in Zhang et al., 2016.

5) Line 224, is not a good reasoning to me. What is the "inter-annual variability"?

We have now elaborated: "Our simulated methane oxidation rate also increases significantly but does not strictly follow this plateau followed by steep increase pattern and has large inter-annual variability (Fig. 4a). This could possibly be due to variability in the prescribed meteorology and OH concentration (Fig. 4b) resulting in variations in the oxidation rate of methane." We now include the air-mass and CH4-reaction weighted OH concentration time series in Fig.4b based on the recommendation in Major Comment #4.

6) Line 370. Isn't it actually a reasonable OPE result in East Asia? Seeing the emissions experienced an increase and then decreased after 2011.

We agree. We have now included two tables in Supplementary (S2 and S6), that summarize the quantities related to East Asia (emissions, TOB, OPE, surface ozone, population weighted ozone) for the split periods 2000-2011 and 2011-2018. Please see our response to Major comment #2.

7) All four-subplot figures (3, 5-8) need to add y-axis labels and add titles for the first row (not just the second row).

Corrected.

8) Format inconsistency in Tables 2-5.

Corrected.

Comments by Anonymous Referee 2:

General Comment:

As a follow-up study to Butler et al., (2020), this study aims to provide new knowledge such as long-term trends and contributions to population weighted O3 from each precursor source, and it certainly contains new findings. However, these new findings are not properly presented, and the results of the study are unsatisfactory in terms of creating useful new knowledge. The text is not well organized and is redundant in places. There are some unnecessary repetitions of the similar phrases (e.g. quotes from Edwards and Evans., 2017). English must be proofread. There are some grammar mistakes. It's better to reconsider the structure of the paper to get it published. Below are my main comments followed by specific comments, which I would like the author to use as a reference for revisions.

We thank the anonymous referee #2 for the well scrutinized review of our manuscript.

We have now taken more care in proofreading our manuscript and corrected all the known erroneous text, grammar, and repetitions in phrases (e.g. repeatedly citing Edwards and Evans 2017).

We have addressed all the comments in detail and included the recommendations in this document. Our response regarding restructuring our manuscript is provided in response to the major comment.

Major Comment:

Considering the theme of this paper, the flow of the paper would be better to first point out the main driver of long-term changes in TOB and surface O3 from the contributions from each precursor source, and then to interpret the changes in these sources from the changes in precursor emissions and/or OPE. However, the current paper presents the analysis of emissions, TOB, OPE, and surface ozone rather independently. The TOB sections (3.2.1 and 3.2.2) are focusing on the description of averaged quantities and clearly lacking detailed trend analysis. On the other hand, in the OPE section (3.3), there is an analysis of OPE trends, but it is not well discussed in relation to the trends in TOB and/or surface ozone. As a result, the new findings are presented here and there in fragments and do not form a coherent story. This is my main concern.

We do recognize that discussing the simulated TOB and surface ozone, followed by explaining this using precursor emissions could form a good coherent story. However, we would like to clarify that the theme and the intended flow of our manuscript has already been laid out in the last paragraph of our Introduction section, where we summarize the questions addressed in our manuscript.

(i) In Sections 3.1 and 3.2: What is the contribution of precursor emissions from various regions/sectors to the global tropospheric O3 burden and its trend?
(ii) Section 3.3: How does the ozone production efficiency of ozone precursor emissions respond to the changes in these emissions during the 2000-2018 period?
(iii) section 3.4: How do contributions of different ozone precursors to the tropospheric O3 burden contrast with their contribution to global mean surface O3 and population weighted O3?

We would therefore like to retain our sequence of first discussing the precursor emissions, followed by discussing the simulated fields such as tropospheric ozone burden, surface ozone etc. Similar structure is also seen in several previous studies (e.g., Gaudel et al., 2020, Zhang et al., 2021, Wang et al., 2022 etc.). This is because it is known that it is the change in emissions that cause the changes in TOB and surface ozone. We discuss the known drivers first (emissions), followed by simulated quantities. Referee #1 also agrees that our manuscript is "well-organized and written".

We did however slightly change the first question in the last paragraph of our Introduction section to avoid possible confusion:

(i) In Sections 3.1 and 3.2: What is the magnitude of precursor emissions from various regions/sectors and their contribution to the global tropospheric O3 burden? How do these emissions change and affect their contribution to the tropospheric ozone burden and its trend?

Even with our chosen sequence (emissions, TOB, OPE and surface ozone), the interconnectedness of these quantities has been discussed. Each subsection refers to the previous subsection(s) in its discussion. We are not sure what exactly makes the referee think that we present these sections independent of each other.

The TOB sections (3.2.1 and 3.2.2) are focusing on the description of averaged quantities and clearly lacking detailed trend analysis. On the other hand, in the OPE section (3.3), there is an analysis of OPE trends, but it is not well discussed in relation to the trends in TOB and/or surface ozone.

The referee points out that the sections discussing TOB lacks discussion of trend analysis and focusses on the mean. We generally agree with this, however we do note that the trends in the NOx (Section 3.2.1) and reactive carbon (Section 3.2.2) contributions to TOB are both discussed in comparison to the trends in their respective emissions. Section 3.2.1 in particular includes two paragraphs discussing trends in the NOx contribution to TOB (lines 296-306). We further include an additional paragraph in Section 3.2.1 that discusses the role of convection in transporting ozone and precursors into the free-troposphere and contributing to the trends in TOB (Please refer to our response to the major comment #1 by referee #1). For RC-tagged TOB trends, we now conclude section 3.2.2 with: "We also note that the percentage trend in contribution to tropospheric ozone burden from anthropogenic NMRC (1.03 %/yr) is more than that of the trend in anthropogenic NMRC emissions (0.61 %/yr), whereas the trend in contribution from methane oxidation (0.33 %/yr) is less than that of trend in methane oxidation rate (0.42 %/yr). While the biogenic NMRC emissions show an increasing trend with medium certainty, the trend in its absolute contribution to tropospheric ozone burden shows a small trend with low certainty."

Throughout Section 3.2 (TOB), we frequently point to the readers to refer to the OPE section (3.3) in order to get a sense of completeness in our explanation (e.g. Lines 295 and 300). We also discuss the trends in OPE in relation to the trends in both the emissions of precursors and the TOB in paragraph from lines 364 to 374 of section 3.3.1.

Explaining surface ozone trends is not related to trends in OPE. We only intend to contrast the contribution of emission sectors/regions to surface ozone compared to their contribution to TOB, as summarized in our 3rd question at the end of Introduction section.

In some places the numbers are not accurate. Authors should take care to use a consistent number of significant digits throughout the text. This seriously impairs the readability of the paper.

We appreciate careful assessment of our manuscript in making specific comments thereby making our manuscript more accurate and readable. We have now rewritten the quoted numbers in both text and tables consistently and with a fixed number of significant digits.

SPECIFIC COMMENTS:

- Mixing the use of "RC-tagged" and "VOC-tagged" should be eliminated , if there is no particular intension.

We now remove "VOC-tagged" and the usage of VOC/NMVOC in results and summary sections. We now consistently use RC-tagged. We also use the term NMRC (Non-methane reactive carbon; NMVOC and CO combined) to denote the RC from non-methane sources (biogenic, biomass burning, anthropogenic). For example at L123, we now define: "Anthropogenic emissions of NOx, and non-methane reactive carbon (NMRC: CO, and volatile organic compounds (VOCs) collectively)"... Also, please see our response to L237 below.

- L11: Mid-latitude regions include Asia, so this is not an accurate statement.

We avoid discussing East Asia in the abstract, given the word limitation and the added complexity in results associated with East Asia. We now write instead: "Over the past few decades, the tropospheric ozone precursor anthropogenic emissions: nitrogen oxides (NOx) and reactive carbon (RC) from northern mid/high-latitude regions (North America (NAM), Europe (EUR) etc.), have been decreasing, and those from (sub)-tropical regions (South Asia (SAS), Middle-East (MDE), etc.)"

- L16: Why is "methane" included in natural emission sources?

We now change this to "RC from methane oxidation" to avoid any confusion related to the inclusion of direct methane emissions, as we prescribe methane concentrations, and the resulting methane oxidation rate is considered as the emission rate of methane.

-L33: "CAM-Chem" is too technical term to be used in Short Summary.

We now re-write the first sentence (also considering the strict limitation on the number of characters to 500 including spaces) in short summary as: "Tropospheric O3 molecules are labelled with the identity of their precursor source to simulate the contribution from various emission sources to the global tropospheric O3 burden (TOB) and its trends." Here we avoid mentioning CAM-Chem, and just write "simulate" instead of "quantify", making it clear to the readers that it is a modeling study. We also write "global" TOB, indicating that we use a global CTM.

- L40: or -> of

Corrected

- L43-44: The definition of NOx and RC should be stated here.

Included now:"(NOx: NO and NO2) and reactive carbon (RC: CH4, CO and Volatile Organic Compounds (VOCs); Levy, 1972; Chameides and Walker, 1973; Crutzen, 1974)."

- L93: CAM4-Chem should be spelled out as here is its first mention.

We expand this now

- L116 : What surface fluxes?

We now write "sensible and latent heat" instead of "surface" fluxes

- Figure2: Why don't you show the comparison of ozone climatology? I don't think Taylor's diagram is the best choice.

We chose using a Taylor diagram, as it was also used in previous studies such as Tilmes et al. 2015, Zhang et al. 2016, Emmons et al., 2020 etc. for the evaluation of simulated vertical profile of O_3. We now make the comparison between the observed climatological period: 1995-2010 and the modeled period: 2000-2010, as suggested by Referee #1 (see our response to Minor comment #4). We also include the comparison of vertical climatology in the supplementary information (Fig. S2).

- L191: "being" -> typo?

Corrected

- L200: Table 3 -> Figure 3?

Corrected

- L200: remove duplicate "period"

Corrected

- L216: I don't think the contribution of Biomass burning (12%) is "very small"

We now rephrase the sentence:" Total anthropogenic NMRC emissions contribute 300 TgC/yr (~22%; Table 4) and Biomass burning emissions contribute 171 TgC/yr (~12.77 %). Aircraft emissions contribute very small amounts (Table 4)."

- L231-233: Are there any influences of fixed surface methane concentration for the decrease of CH4 lifetime?

We are not sure exactly what the reviewer means by fixed methane concentration influencing methane lifetime. Fixing the surface concentration of methane is standard practice in studies using Chemical Transport Models such as CAM-chem.

- L237: Only NMVOC emission? RC emission here should be the sum of NMVOC and CO.

Please see our response to the first specific comment. We now refer to the sum of NMVOC and CO as NMRC (Non-methane reactive carbon).

- L240: NOx emission -> RC emission

Corrected

- L244-245: I'm not sure if this definition of TOB is correct. In this definition, if surface O3 exceeds 150 ppb, will that grid not be included in TOB calculations?

We now clarify this as "The tropospheric ozone burden is calculated as the mass of ozone in the model grid cells below the ozone-tropopause, defined as highest layer in the upper troposphere with an ozone mixing ratio less than 150 ppb (E.g., Bak et al., 2022, Liu et al., 2022).".

- L247-249: Are the years of these TOB estimates the same as the years covered by this study? If not, the time periods covered in the different estimates should be provided here.

We now elaborate this paragraph by specifying the time period of 2005-2014 used in these previous studies: "Our simulated tropospheric ozone burden is towards the lower end of the values simulated by several multi-model studies for the 2005-2014 period (values from Griffiths et al., 2021): the CMIP 6 ensemble 356±31 Tg O3; ACCENT: 336 ± 27 Tg O3; Atmospheric Chemistry and Climate Model Intercomparison Project, ACCMIP: 337 ± 23 Tg O3; Tropospheric Ozone Assessment Report, TOAR: 340 ± 34 Tg O3; and IPCC: 347 ± 28 Tg O3 (Szopa et al., 2021)."

- Sections 3.2.1 and 3.2.2 : As stated in Major Comments, several values in these sections are not consistent with the corresponding values in Table 2 to 5. If the values in the tables are cited in the text, they should be quoted accurately.

We have now cross-checked all the quoted values in the text with those in the tables and figures and corrected wherever inconsistent.

- L298: Fig4 -> Fig3

Corrected

- L298-299: The smaller trend of TOB in % than that of NOx emission is apparent for regional contribution, but not in the case for the other contributions.

We clarify this now:" The percentage slope (ignoring the sign) of the trend in the contribution to tropospheric ozone burden by regional anthropogenic NOx sources, however, is generally smaller than that of NOx emission trend (Table 3)."

Similarly at line 367, we write: "The decreasing (increasing) NOx emissions from regional anthropogenic sources becoming more (less) efficient at producing ozone, and this leads to a dampening effect where there is a smaller percentage slope (ignoring the sign) in tropospheric ozone burden compared to the slope in NOx emissions"

- L319 and the other places such as Table 4 and 5: In this study methane "emission" is not explicitly considered, so the description about methane "emission" in the text (and also in the Table) should be more scientifically accurate.

In our revised figures, tables and discussion in the text, we now refer to this as "RC from methane oxidation" to avoid such possible confusions.

- L355-356: These OPE values are inconsistent with Fig 6c. Is the value of East Asia correct here?

It has now been corrected. We also make sure that all our quoted values in the revised text from the new simulation results are consistent. We have now updated the discussion and quote the result correctly. We discuss the OPE of East Asian precursor emissions and its trend for the split periods (2000-2011 and 2011-2018), as recommended by Referee #1.

- L379: NOy should be defined as here is its first mention.

Defined now: "NOy (NOx and reservoir species for NOx; e.g., NO3, N2O5 etc.)"

- L446-448, L488: Why are you quoting relative trends (in %/yr) here? How can you state that these contributions are the main contributors? Are there any qualitative criteria to pick up main contributors? (and those criteria is for relative trend?)

Main contributors: In line 446-448, we write that increasing stratospheric contribution and contribution from anthropogenic NOx mainly drive the increasing trend in total global mean surface ozone. While these contributions certainly are major contributions to the climatological mean, we recognize that these contributors are not the only main drivers of trend in total surface ozone. Every contribution (increasing, decreasing, or trends with low-mid certainty) definitely has

some role to play in influencing the trend in total ozone. However, since stratosphere and anthropogenic contributions are already main contributors to the climatological mean and are showing increasing contributions, they easily stand out as the major drivers of increasing trend in total ozone. For L488, the above explanation holds. We replace "mainly contributed by" with "mainly driven by" at L486, to keep the wording consistent and avoid confusion among readers.

Criteria to pick main contributors: In our study, we just recognize that increasing trends are mainly driven by increasing trends in major contributors to the climatological mean. We do not use any specific criteria to demarcate and emphasize the role of a single contributor. Further studies could use methods such as fixing the contribution of a component instead of allowing it to vary with time and see the resulting trend in total ozone. Large changes in total ozone trends due to absence of a trend in contribution from a given component could indicate the importance of that component in driving the trend in total ozone.

The units to express trends: We agree that expressing trends in only %/yr isn't any criterion to determine whether any single tagged component is the major contributor. We also recognize that this could lead to confusion among readers. We have therefore now also included the trends in the original units (ppbv/yr) along with trends in %/yr. The main application of trends in %/yr is to compare the rate of change in quantities whose units aren't compatible (E.g.: Comparing TOB (in Tg O3) trends with trends in global mean surface ozone (in ppbv))

- L454-456, L459-460: Why is the contribution of ship emissions to population-weighted surface O3 large? Could you spend more words for this?

We now add a map plot comparing the spread of surface ozone and population weighted ozone attributed to ship NOx emissions (Fig. 10). We now add at L459:

"Despite NOx emissions from international shipping happening only over ocean grid cells, the ozone attributed to ship NOx spreads across land areas and contributes to the total surface ozone by ~3-6 ppb (Fig. 10a). Although there is no population over the regions where international ship NO_x is emitted, the global population weighted mean ozone attributed to ship NOx is comparable to that of global mean surface ozone (Table 2) as large populations at several coastal areas and densely populated inland regions in East Asia and South Asia are exposed to ozone attributed to ship NOx emissions (Fig. 10b)."

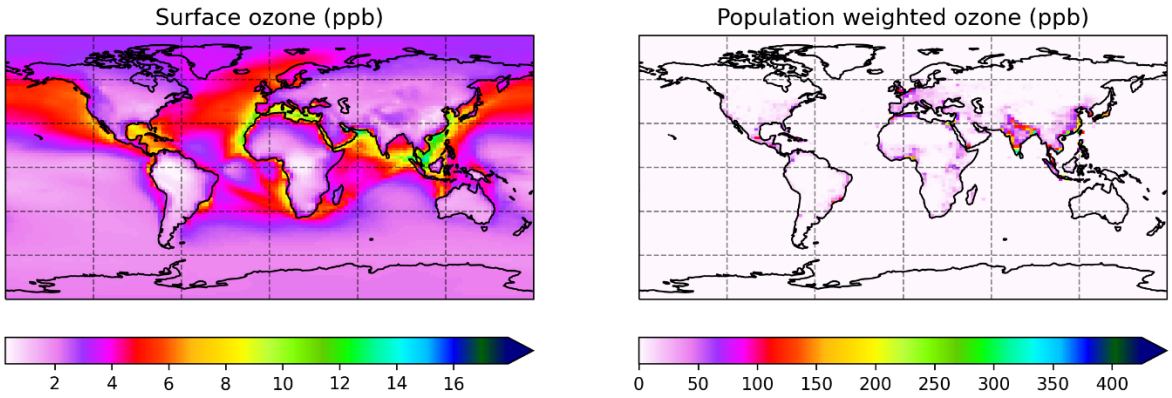


Fig. 10: Spatial distribution of climatological (2000-2018) annual mean O_3 attributed to NO_x emissions from international shipping (in ppbv): a) Surface ozone and b) Population weighted surface ozone.

Comments by Referee 3 (Michael Prather):

This manuscript is the second within a month that I have been asked to review by ACP editors, which is centered on the use of tagged O3 tracers as way of attributing the impact of different emission sources of NOx and VOCs (called reactive carbon, RC, here). For the first ms, see (1). As for (1), I find that the concept of tagged O3 tracers is inherently incorrect and leads to false attributions. Given my questioning the fundamental validity of this work, it is necessary to sign this review as I did for (1). If wiser heads believe my interpretation unfounded then let the Editor decide.

We thank Michael Prather for his detailed review of our manuscript, his opinion on the perceived limitation of tagging method being applied in our study, and for providing references and specific comments that enhance the quality of our manuscript.

Since Michael Prather has brought up another paper using an ozone tagging methodology as a part of his review of our manuscript, we feel it is appropriate to refer to the open discussion of that other paper (egusphere-2024-324). The authors of egusphere-2024-324 provide a very good discussion of the differences between model perturbation and tagging approaches in their reply to Michael Prather's review. Like the authors of egusphere-2024-324, we also feel that the key to responding to the review of our manuscript by this referee hinges on this distinction. Their response can be accessed here: (https://editor.copernicus.org/index.php?_mdl=msover_md&_jrl=778&_lcm=oc108lcm109w&_acm=get_comm_sup_file&_ms=117926&c=271228&salt=106681673156584561). We further provide our own response to the review by Michael Prather below.

We agree that the tagging methodology cannot be used to explain the *impact* of emissions perturbations or changes in stratospheric influx on tropospheric ozone. It

is used to determine the *contribution* from the tagged source. In our work, we do not attribute the *impacts* of emission changes, but rather calculate their absolute and relative *contributions* of various sources to the total tropospheric ozone. The differences in the derived attributions from these two methods are apparent and very much expected in a non-linear system. Here is a quote from Grewe et al., (2012) that stresses on the non-interchangeability of the two methods (perturbation and tagging):

"It has to be stressed that both methods were previously applied in many investigations: The tagging method to identify contributions from individual sources (Horowitz and Jacob, 1999; Leliveld and Dentener, 2000; Meijer et al., 2000; Grewe, 2004) and perturbation approach to investigate atmospheric sensitivities to individual sources (Horowitz et al., 1998; Stevenson et al., 2006). Both methods are useful and target different objectives, but they are not interchangeable (Wang et al., 2009; Grewe et al., 2010)"

Further, this quote from Butler et al., (2020) can help explain the differences more vividly, and in terms of their usefulness/policy relevance:

"In the absence of non-linear chemical interactions, these two different approaches (perturbation and tagging) ultimately yield the same results; however, for tropospheric ozone, which can show highly non-linear interactions between its NOx and reactive carbon precursors under some circumstances, these approaches can sometimes yield very different results (Grewe et al., 2010; Mertens et al., 2018). As air pollution mitigation strategies must involve some change in emissions, perturbation studies will always be necessary for policy-relevant modelling of atmospheric chemistry. However, tagging studies on their own can play a role in helping to identify which emissions to mitigate (Grewe et al., 2010). When combined with perturbation studies, tagging can reveal how the contribution of unmitigated sources to ozone changes in response to mitigation measures (Mertens et al., 2018)."

Tagged tracers for key species with major chemical feedbacks give false results. I hope we can agree that using tagged tracers for CH4 sources fails to include the well established feedbacks and thus underestimates the attributable CH4 perturbation by 40%. This feedback has been standard since the 1995 IPCC SAR and a later heuristic figure is given in (2). For O3 we have lived since Roelofs & Lelieveld with the use of O3S as a tagged tracer of stratospheric ozone that has been implemented in all the MIPs and continues to show that O3S/O3 = 30%-40%, thus attributing 30%+ of tropospheric O3 to the stratosphere.

We have long known that O3 also has feedbacks on it chemical production and loss, but have not pursued it. Last year, (3) used chemical modeling of the ATom profiling

of the Atlantic and Pacific Ocean basins to demonstrate that positive O3 perturbations reduced the production of odd-oxygen and thus accelerated its own destruction. That feedback, i.e., dln(P-Ox)/dlnO3 = -0.3 to -0.4, for the ATM data underestimates the sensitivity because it was based on a 24-hour calculation. Over the lifetime of the O3 perturbation (>10 days) the added O3 will accelerate loss of NOx to HNO3 (increased NO2:NO, more OH) and reduce P-Ox further. This year, (4) pursued this effect with direct additions of 'stratospheric' O3 and quantified the perturbation lifetime (~25 days) and watched the decay (also ~25 days). This implies that the impact of stratospheric O3 on tropospheric is about 8%, not 30%. The tagged tracer fails to alter the baseline O3 as it should. Simply, tropospheric O3 is highly buffered.

The O3S/O3 of 30-40 % (quoted from previous studies; e.g., Roelofs and Lelieveld, 1997) is the percentage of the burden of O3S (110 Tg O3) in the troposphere *contributing* to the total tropospheric ozone burden (TOB; 271 Tg O3). In Prather and Zhu 2024 (4), the contribution from STE (possibly in the order of 30-40 %; not reported) was most likely already included in the 345 Tg O3 of TOB simulated in the CTRL run. This O3S is not a quantity that conveys the *impact* of changes in TOB due to introduction of STE flux changes, but rather a *share* of tropospheric ozone that is of stratospheric origin.

The 8% (~26 Tg O3) however, is the *impact* of perturbation in STE calculated by multiplying the derived perturbation lifetime (~24-25 days) with the STE flux (400 Tg O3/yr; observation based). This perturbation lifetime is calculated as the change in TOB in the eO3ste runs (~6.64 Tg O3) divided by the *imposed additional* flux of ozone from STE to the troposphere (100 Tg O3/yr).

Therefore, deriving the tropospheric burden by multiplying perturbation lifetime with the emitted ozone flux, is certainly a valid approach to assess the *impact* of additional fluxes of ozone into the troposphere, but does not communicate the *contribution* of a source to the total TOB. Since O3 is highly buffered and non-linear, large differences between *contribution* derived from tagged tracers and *impact* derived from perturbation are very much expected.

I believe that we need to recognize that major gases, like CH4, O3, and N2O, have chemical feedbacks that alter their perturbation lifetimes (change in burden / change in emissions); and we should stop pursuing simple linear models to attribute their impacts.

Michael Prather

UC Irvine

We agree that several chemical-feedbacks are at play here. But would also like to clarify again that in our study we do not attribute the *impacts* of changes in emissions (which generally requires CTRL and PERTURBED simulations). We only attribute the *contribution* from emissions, within the CTRL simulation itself, as with studies that simulate O3S. Therefore, any perturbation lifetime changes shouldn't matter, as we aren't introducing any emission changes that are resulting in these lifetime changes.

Regarding the accounting of chemical feedbacks: We do not use "simple linear models" where the decay rate of the tagged tracer such as O3S is either prescribed based on a previous standard simulation (e.g., Liu et al., 2020) or calculated online (e.g., Williams et al., 2019). In our approach, we modify the chemical mechanism to label the participating chemical species with their source identities (tags). The reactions of the tagged species follow the same reaction rate calculation of the default untagged species. Thus, a change in production/loss rates (related to the reaction rates) in the untagged species due to perturbation or any other factors is also reflected in the reaction rates of the tagged species. This makes our tagging method valid, reliable and especially useful in assessing the compensating feedbacks i.e. the changes in contributions from various sources when perturbations are introduced in the simulations. For example, in Table 4 of Butler et al., 2020, a perturbation simulation with 25% increase in prescribed CH4 concentration results in ~13% increase in CH4 contribution to TOB. There is also a compensating reduction in contributions from other tagged reactive-carbon sources leading to an overall increase in TOB by only ~3%.

A comparative study by introducing additional STE fluxes, as in Prather and Zhu 2024, and quantifying the changes in TOB and contribution from STE using the O3S tagged tracer method and our TOAST tagging method, intending to assess the chemical feedbacks, could be an interesting topic for future studies.

Given my questioning the fundamental validity of this work, it is necessary to sign this review as I did for (1). If wiser heads believe my interpretation unfounded then let the Editor decide.

In summary: Chemical tagging is a well-established approach used in several previous studies (cited in the paragraph at Line 75), thereby making this method fundamentally valid for the quantification of the *contribution* from various sources, and not the *impacts* of perturbation.

In addition to the problem of tagged O3 tracers, there are other issues that make this manuscript not ready for publication. Some specific detailed comments.

Thanks again for the detailed specific comments, helping us improve the quality of our manuscript. We have addressed all these comments below.

L11: I think this is only northern mid-latitudes?

We now write: "from northern mid/high-latitude regions (North America (NAM), Europe (EUR) etc.)"

L22: 'consistent with previous work' Yes, indeed, this was shown 20+ years ago in (5), it is old-hat

Thanks for the reference. We now include this additional citation in our Introduction section (see our response to L56 below)

L23: this throwaway causal statement about convection and lifetime is probably incorrect. more likely it is just sunlight (lower latitude)

We agree that there are many factors influencing the production of ozone. In our revised manuscript we now also acknowledge the role of larger availability of sunlight and warmer temperatures which in general leads to larger reaction rates at the tropics.

Recent studies (Zhang et al., 2016, 2021) have established the predominant role of strong convection in the tropical regions on the increase in tropospheric ozone burden, through sensitivity simulations. Zhang et al., 2016 note: "Increases in O3 precursor emissions south of 35◦ N are transported efficiently to the middle and upper troposphere, from strong convection in the Hadley cell, whereas emission decreases north of 35◦ N stay at high latitudes and low elevation in Ferrell cell circulation (Fig. 4). When global emissions shift equatorward, strong convective mixing over the tropics and subtropics lifts O3 and its precursor NOy to higher altitudes (Figs 3b and 4b and Supplementary Figs 4 and 5), where the O3 lifetime is longer, favouring O3 accumulation. When emission increases occur in NH mid-latitudes, less NOy is lofted to high altitudes (Fig. 4c)." Also, please see our response to major comment #1 by Referee #1.

We therefore modify this sentence in the abstract as "Tropical regions, despite smaller emissions, contribute more to TOB compared to emissions from higher latitudes, consistent with previous work, predominantly due to large convection (in addition to strong sunlight and larger reaction rates) at the tropics thereby lifting O3 and its precursor molecules into the free troposphere where ozone's lifetime is longer."

L56: ibid, I doubt that convection in the subtropics is that much more than mid-latitudes over the continents in summer. I really feel that you have no justification for these attributive statements.

Please see our response to your previous comment (L23), where we quote from Zhang et al., 2016 that establishes the predominant role of tropical convection. In our revised manuscript, we make sure that we do not portray the convection as the only reason for efficient ozone production by precursor emissions in the tropical regions. We now also modify the sentence at L56 to: "This is mainly due to larger convection of polluted air masses from the boundary layer into the free troposphere, in addition to larger reaction rates and NOx sensitivity over the tropical regions (Wild et al., 2001) compared to extra-tropical regions."

We agree that it is quite possible for subtropical and mid-latitude convection over continental regions during the summertime to be similar. However, we analyze the annual quantities and not the seasonal behavior in our study. Further, we now demonstrate the predominant role of convection in the efficient production of ozone by NOx from tropical regions through vertical profiles of simulated ozone and NOx fields attributed to anthropogenic emissions from tropical regions (e.g. Southeast Asia) and compare them with those from mid-latitude regions (e.g. Europe). Please see our response to Referee #1's comment 1).

L70: again, see (5).

We include this reference now. See our response to L56.

L77: here we greatly disagree about what tagging methods do.

Please see our response to the main comment (above). We don't disagree, we rather agree that the tagging methods don't convey the *impact* of emissions. We clarify that our method conveys the *contribution* from various tagged emission sources.

L96: I do not see how this is quantitative.

As explained in the sentence, we separately attribute the simulated contribution to either NOx or RC source. We further elaborate now by adding to the paragraph and explaining: "To illustrate: O3 attributed to biogenic emissions in NOx-tagged simulation would clearly mean that the contribution is from emitted biogenic NOx only, irrespective of where the RC comes from to produce the O3 molecule. Contribution from biogenic RC emissions can be simulated in the RC-tagged simulation. In this way, the roles of NOx and RC emissions from a given sector are exclusively simulated in separate simulations."

L112: the extension tagging to surface O3 is even more questionable

Please see our response to the main comment. The tagging method can be applied to quantify *contribution* from various sources to surface ozone concentration just as well as for tropospheric ozone in general (E.g., Butler et al., 2020, Li et al., 2023a)

L118ff: The model resolution is a bit coarse, especially for modeling pollution NOx, but for that even 1x1 is not enough. What is worrisome is the nudging since that changes the residual circulation and the convection. What is done for convection, since the authors invoke that often in explaining low-latitude O3 production.

We agree. We also state the limitation of model resolution especially in the context of the OPE of ship NOx emissions at lines 359-363. We also agree that the nudging does change the residual circulation and convection. We therefore set a 10 % relaxation of meteorology towards analysis fields (see Lines 120-121), consistent with previous studies that use this nudging approach within CESM for MERRA/MERRA2 meteorology reanalysis dataset (e.g., Tilmes et al., 2015, Emmons et al., 2020).

L125: Aviation NOx has only 3 altitude? I would think it important to include the full spread of cruise altitudes.

We clarify that the aviation emissions are provided for three distinct phases of flight (landing/takeoff, climbing/descent, and cruising). The vertical distribution of aviation emissions for each of these flight phases is taken from HTAPv3 emission inventory We agree that the wording in our manuscript was unclear, and replace this with the following text in our revised manuscript:

"We specify aircraft emissions at various altitudes effectively representing three different flight phases (landing/take-off, ascent/descent, and cruising)."

L129: you say "interpolate" but I hope you mean "integrate" as you would want to ensure that the totals are conserved.

We now replace "interpolate" with "re-grid (using a standard conservative re-gridding algorithm)" to clarify this possible confusion among readers.

L134: This is truly odd, why not just use the observed zonal mean CH4 surface data from NOAA? The idea of using a flux inversion is potentially dangerous and certainly at odds with typical AerChemMIP protocols. (Thus, comparisons become more difficult).

We agree that data from NOAA could be used as well, which can further be used for comparison with MIPs. Nevertheless, our study was not performed within the framework of AerChemMIP. As far as we can tell, the NOAA data archive only provides measurement timeseries from individual stations, and global mean mole fractions. CAMS is also a standard product used in previous studies (e.g., Shaw et al., 2022). In our case we don't use the full CAMS flux inversion, but rather just the mole fraction fields from the CAMS inversion product v18r1. Furthermore, we do not use the full latitude/longitude gridded surface mole fractions from CAMS, but rather derive zonal and monthly mean averages from these fields, which we believe give a

good representation of the zonal and monthly mean trends and variability in the underlying measurement data due to the use of data assimilation in the CAMS inversion.

L154: why are you truncating the computer output to get ~1% errors? Is that necessary?

We don't truncate the computer output. Our intended message was that the differences between our tagged and untagged simulations due to numerical precision effects are very small. We remove this sentence to avoid possible misunderstanding from readers.

L184: why exclude aviation NOx from surface ozone?

This is only while referring to anthropogenic component in our discussion. We slightly modify this sentence now: "When referring to anthropogenic emissions and their contributions to tropospheric ozone in our discussion, we only refer to surface-based anthropogenic emissions and exclude aircraft emissions."

L231: the methane lifetime is very 'hot' here, about 6.5 yr, at the bootm end of any of the CMIP models. It is not clear whether this includes stratospheric loss and soil loss. Given the extreme value here, it would be useful to explain how you calculate it. Also, Figure 4 is hard to understand, did you give the correct legend here?

We agree. We have also recognized that the anthropogenic NOx emissions were previously ~1.5 times more than intended, leading to a larger CH4 oxidation rate and smaller CH4 lifetime than expected. Therefore, we conducted our simulations again with corrected anthropogenic NOx emissions and our new estimate of CH4 lifetime is ~8.54 years. Please see our response to Referee #1's major comment 4 for the modified figure.

We now specify how we calculate methane lifetime in Line 321: "The tropospheric methane lifetime (~8.54 years), calculated as the total atmospheric methane burden divided by tropospheric methane oxidation rate, decreases with high certainty (-0.01 years/yr; Table S5) in our simulations (Fig. 4a: Magenta line)."

Stratospheric and soil losses haven't been included, as we only consider the tropospheric burden and oxidation rate of CH4.

L255: Here is where the tagging shows a clear difference from O3 perturbation experiments.

We agree. This difference is very much expected in non-linear chemical regimes, as explained in our response to the major comment.

L330ff: The Ozone Production Efficiency is calculated incorrectly and obviously has the wrong units. As the authors note, most researchers have move to calculating the net production of O3 (Tg-O3/y) per emissions of NOx (Tg-N/y), for a units of Tg-O3/Tg-N, or better in moles. The units given in line 340 are missing the 'time' unit.

Thanks. We now change this from “the annual mean ozone precursor emissions” to “the amount of precursors emitted in a given year”. This fixes the seemingly missing time unit in the denominator of OPE.

L356: The original reference here for more tropically oriented production per NOx emission is Wild 2001 (5). And the reason is not due to the smaller "availability" of NOx, but simply to sunlight, temepratures, and water vapor.

As specified in the previous responses, we now do not exclusively mention the tropical convection to be the only driver of free tropospheric ozone increases. We also specify the other factors such as larger reaction rates at the tropical regions due to stronger availability of sunlight and warmer temperatures. We now include Wild et al., 2001 reference at L356.

References

(1) https://egusphere.copernicus.org/preprints/2024/egusphere-2024-324/#discussion

(2) Prather, M.J. (2007) Lifetimes and time-scales in atmospheric chemistry, Phil. Trans. R. Soc., A 365: 1705–1726, https://doi.org/10.1098/rsta.2007.2040. FIGURE 2

(3) Prather, M. J., Guo, Hao and Zhu, Xin (2023) Deconstruction of tropospheric chemical reactivity using aircraft measurements: the ATom data, Earth Syst. Sci. Data, 15, 3299–3349, doi: 10.5194/essd-15-3299-2023. TABLE 2 & SECTION 6.1

(4) Prather, M.J. and Xin Zhu (2024) Lifetimes and timescales of tropospheric ozone, Elementa: Science of the Anthropocene, 12 (1): 00112, doi: 10.1525/elementa.2023.00112

(5) Wild, O., M. J. Prather, H. Akimoto (2001) Indirect long-term global cooling from NOx emissions, Geophys. Res. Lett., 28, 1719-1722, doi: 10.1029/2000GL012573. FIGURE 3

Comments by Owen Cooper:

Line 51: When mentioning the equatorward shift of ozone precursors you could also cite a new paper recently published in the TOAR-II Community Special Issue (Li et al., 2024), which shows that the equatorward shift is continuing in Asia, according to the latest bottom-up emissions inventories.

Thanks for the reference. We now include in the paragraph of line 51: "A recent study (Li et al., 2024) confirms an equatorward shift in Asian NOx emissions between 2010 and 2017 according to bottom-up estimates."

Another TOAR-II paper under review is Mertens et al. (2024) and there are some similarities between this paper and yours. It would be helpful to briefly mention this other TOAR-II submission and comment on similarities or any differing conclusions.

Mertens et al., (2024) is an extension to the previously published Mertens et al., (2018) which we have already discussed at several places in our manuscript. There aren't many directly relatable results in Mertens et al., (2024) that we can readily discuss in our current manuscript, as they use the combinatorial approach to tagging (as opposed to the separate NOx and RC tagging in our study) and their focus is mainly on the contributions from land transport emissions on tropospheric O3 and OH in future scenarios. However, we discuss the result related to OPE as being consistent with that discussed in Mertens et al., (2024).

At the end of paragraph at Line 331, we write: "A similar metric: Ozone Burden Efficiency (OBE) has been defined by Mertens et al., (2024), as the ratio of ozone burden in the troposphere attributed to precursor emissions (both NOx and RC) from a given source using the combinatorial tagging approach (in Tg O3) to the emitted NOx during the year (Tg NO) for various tagged sectors."

We split the paragraph at line 364, with the first paragraph ending with: "The decreasing (increasing) NOx emissions from regional anthropogenic sources becoming more (less) efficient at producing ozone, and this leads to a dampening effect where there is a smaller percentage slope (ignoring the sign) in tropospheric ozone burden compared to the slope in NOx emissions (Table 3). This result is also consistent with Mertens et al., (2024) where an increase in NOx emission from various tagged sectors is associated with a decrease in OBE in both present-day and future scenarios."

Line 40: When referencing the estimated number of deaths due to ozone exposure, the 95% confidence interval (as reported by Malashock et al., 2022) should also be provided.

We now include the interval: "(~423100 Ozone attributable deaths [95 % confidence interval: 223200,659400] in 2019; Malashock et al. 2022)"

Tables 3-5: As described in the Guidance note on best statistical for TOAR analyses, all trends should be reported with 95% confidence intervals and p-values. The expression "insignificant" should not be used. The reader can tell if a trend value is meaningless based on 95% confidence interval and the p-value.

Thanks. We now include the 95% confidence interval and p-values in tables 3-5 and in supplementary tables.

Additionally, we now have a sub-section (Section 2.3: Trend Analysis):

"We express the trend for the 2000-2018 period in various quantities discussed in our study: Precursor emissions, tropospheric ozone burden, global surface mean and population-weighted mean ozone concentration as a slope of the timeseries in respective units/yr, and in %/yr relative to the first absolute value (for year 2000). This slope is calculated using the Theil-Sen estimator method available as a python module (https://docs.scipy.org/doc/scipy/reference/generated/scipy.stats.theilslopes.html), which also calculates the 95 % confidence interval of the slope. We further use the Mann-Kendall test to determine the extent to which the detected trend is monotonic (Hussain and Mahmud, 2019), expressed by the p-value. In the TOAR Guidelines for best statistical practices (Chang et al., 2023), it is recommended not to use a dichotomized expression such as significant/insignificant trend. Hence, we also provide the p-value and the 95 % confidence interval in the supplementary information for the interested readers to assess the meaningfulness of the estimated trend in our study. For the sake of discussion in sections 3 and 4, we categorize the trends with p-values less than 0.05 to be of high certainly, between 0.05 and 0.1 to be of medium certainty and larger than 0.1 to be of low certainty (Table 3 in Chang et al., 2023)."

For all our time-series figures, we specify p-value rather than the term "No-trend". See the figures above (responses to Referee #1's major comments 1 and 4) in this document as examples.

References (in addition to the ones provided in the main manuscript)

Bak, J., Song, E.-J., Lee, H.-J., Liu, X., Koo, J.-H., Kim, J., Jeon, W., Kim, J.-H., and Kim, C.-H.: Temporal variability of tropospheric ozone and ozone profiles in the Korean Peninsula during the East Asian summer monsoon: insights from multiple measurements and reanalysis datasets, Atmos. Chem. Phys., 22, 14177–14187, https://doi.org/10.5194/acp-22-14177-2022, 2022.

Chang, K.-L., Schultz, M. G., Koren, G., and Selke, N.: Guidance note on best statistical practices for TOAR analyses, arXiv preprint arXiv:2304.14236, 2023b.We also avoid the expression "insignificant" at several places in our manuscript. Instead, we use the terms "low/middle certainty" based on Chang et al., (2023).

Chua, G., Naik, V., and Horowitz, L. W.: Exploring the drivers of tropospheric hydroxyl radical trends in the Geophysical Fluid Dynamics Laboratory AM4.1 atmospheric chemistry–climate model, Atmos. Chem. Phys., 23, 4955–4975, https://doi.org/10.5194/acp-23-4955-2023, 2023.

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Lawrence, M. G., P. Jöckel, and R. Von Kuhlmann. "What does the global mean OH concentration tell us?." *Atmospheric Chemistry and Physics* 1.1 (2001): 37-49.

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