Responses to Report#1

We thank the reviewer for his comments and recommendations. Our responses are in blue following each comment.

Re-review of Elshorbany et al.

Although several of the previously identified fatal points have been removed, there are still several important issues that remain unaddressed, as follows:

1. Section 2.2.1: I have to repeat that it is fundamentally important to describe satellite data usage criteria, e.g., cloud fraction and data quality flag.

Answer: We have added the data usage criteria for the used satellite products.

2. In Line 784 (ATC-1.pdf), the authors added a statement that variations and trends with respect to season or altitude are not considered. However, I strongly suggest conducting seasonally-separated trend analysis for NO2, CO, HCHO, and the HCHO/NO2 ratio. With data shown in Figure S6, I believe the authors could address this issue. The chemical regime analysis is only meaningful for the ozone production season.

Answer:

We have conducted a separate seasonal and latitudinal analysis of ozone photochemical regimes, including O3 and its precursors, NO_2, CO, and HCHO, and will be submitted within two weeks to the same special issue (Fadnavis et al., 2024).

In addition, we now show the seasonal variability of TrC-O_3 trends in Figure S7 as also outlined in section 3.4.1.

We think that this will address the reviewers' comments and also avoid confusing the readers with too many analyses.

”

I am also still concerned about the altitude dependence, about the applicability of the HCHO/NO2 COLUMN ratio to infer chemical regime and its change, beyond the strong ozone production region (i.e. non-urban region). The small trends in NO2 and HCHO might be from different altitudes, particularly in the global-scale analysis, about the background atmosphere (lines 796ff). In my opinion, the term "regime" should be only used for the urban or photochemically-active region and the relevant season, where near-ground ozone production is obvious and dominating the column quantities.

Answer:

Ozone regimes can usually be explicitly determined using ozone sensitivity studies (e.g., Fadnavis et al., 2024, this special issue). However, indicators, such as HCHO/NO_2 which uses remote sensing products of NO2 and HCHO can still provide valuable insights into the chemical regimes. We mentioned in section 3.4.7 that the HCHO/NO2 is not an optimal indicator of zone sensitivity, which would require a sensitivity analysis “Although imperfect (e.g. Souri et al., 2023), this indicator yet provides some qualitative information on the evolution of the O_3 regime over the last years (Nussbaumer et al., 2023). We note that this analysis does not consider variations in the ratios and their trends with respect to season or altitude.”. In addition, our results based on HCHO/NO2 are consistent with our ozone sensitivity analysis from our sensitivity paper (e.g., Fadnavis et al., 2024, this special issue) and support our findings.

The word “regime” indicates the prevailing conditions in a certain region(s) and, as the reviewer suggested, we didn’t use it on a global scale or to indicate general conditions.

3. When trends in TrC-NO2 or HCHO are analyzed in combination to TrC-O3, not only STE (newly stated in lines 588, 591, and 763 of ATC1.pdf), but also baseline trends as well as increased/decreased long-range transport of ozone are equally or even more important. These points have to be mentioned in the Abstract too. I do not find Li et al., 2024 in the references.

Answer: We thank the reviewer for this comment, and we have now included long-range transport (LRT) throughout the text and in the abstract.

4. Concerns about the analysis over Australia. Why more points appear in Figure 21 (HCHO/NO2 ratio) than Figure 18 (TrC-HCHO) over Australia? What is the NOx concentration levels over the central Australia, showing an increasing trend? What does the "shift toward VOC-sensitive conditions with ozone production" mean over such region?

Answer: This comment is comparing two different quantities. Figure 18 shows the HCHO trends only whereas Figure 21 shows the HCHO/NO2 ratio annual trends. The caption in both figures read “white areas correspond to regions where the trends remain statistically insignificant at a 95% confidence level.”. Therefore, the different number of points for the two different quantities is due to the different significance (confidence) of the different quantities.

As shown in Figure 4 “Mean (2005-2019) of TrC-O_3, TrC-NO_2, TrC-HCHO, and TC-CO”, TrC-NO2 levels over central Australia is 0.5 Pmolec/cm2, and mean (2005-2019) HCHO/NO2 ratio is very high (10) indicating mean (2005-2019) NO sensitive conditions. However, since the HCHO/NO2 ratio shows a small trend, the system is slowly moving toward VOC-sensitive conditions. As mentioned in the last sentence of the last paragraph of section 3.4.6, this is due to increasing NO2 trends but decreasing HCHO trends “Similarly, while NO_2 trends are slightly increasing over central and southern Australia, trends of TrC-O_3 and TrC-HCHO are decreasing, which indicates a trend toward VOC-limited conditions”

5. While the authors state that HCHO trends are inconsistent with that of O3 in regions including southeastern US (after revision, Lines 761-765), it is said consistent in line 1291-1299, in the Eastern US. Some clarification is needed.

Answer: The sentences in lines 761-765 refer to the trends in some regions, with an example over the southeastern US while in lines 1291-1299 in the conclusion we refer to the trends in the northeastern US. The reviewer is specifically referring to the following sentences in lines 761-765 "HCHO trends are inconsistent with that of O_3 (sec. 3.4.1) **in some regions** which might be due to several factors, such as their different sensitivity to NO_x and hydrocarbons (Luecken et al., 2018) but also possible STE contribution to tropospheric ozone levels, especially in midlatitudes (Willimas et al., 2019; Li et al., 2024). For example, while TrC-O_3 is increasing in the **southeastern US**, TrC-NO_2, TC-CO, and TrC-HCHO are decreasing, which, in addition to the local chemistry, might indicate a STE signal." and lines"1291-1299 "The decreasing trends of TrC-O_3 over parts of the **northeastern US** and Europe are likely due to the decreasing trend of TrC-NO_2, which is due to the effective measures applied over the last two decades to mitigate air pollution in these regions. TrC-HCHO trends are decreasing in the Eastern US, some parts of northern and western Africa, and western and northern Europe, and increasing in South Asia, central Africa, northern Australia, and Brazil. TrC-HCHO trends are consistent with that of TrC-O_3 over Eastern US and Europe."

For further clarification, we have modified the sentences in line 761-765 to be "HCHO trends **varies** with that of O_3 (sec. 3.4.1) which might be due to several factors, such as their different sensitivity to NO_x and hydrocarbons (Luecken et al., 2018) but also possible STE contribution to tropospheric ozone levels, especially in midlatitudes (Willimas et al., 2019; Li et al., 2024). For example, while TrC-O_3 is increasing in the southeastern US, TrC-NO_2, TC-CO, and TrC-HCHO

are decreasing, which, in addition to the local chemistry, might indicate a STE signal. TrC-NO_2 trends are decreasing over the northern coast of Australia while those of TrC-O_3 and TrC-HCHO are increasing. While the increase of HCHO/NO_2 might indicate a trend toward NO-limited conditions (see below), the increase of TrC-O_3 trends in this region might also indicate increasing trends of STE contribution (Li et al., 2024). **However, TrC-HCHO trends are consistent with that of TrC-O_3 in other regions, e.g., over the northeastern US and Europe.** Similarly, while NO_2 trends are slightly increasing over central and southern Australia, trends of TrC-O_3 and TrC-HCHO are decreasing, which indicates a trend toward VOC-limited conditions (see below)."

Overall, the manuscript needs major revision in the areas listed above.

We thank the reviewer for his comments, which we have addressed as outlined above.

Responses to Report#2

We thank the reviewer for his comments and recommendations. Our responses are in blue following each comment.

Review of Elshorbany 2024
Tropospheric Ozone Precursors: Global and Regional Distributions, Trends and Variability

The revised paper is much improved, shorter, and more evidenced. On the whole the three reviewer's comments have been addressed. I would however recommend that the authors address the below outstanding comments before it can be recommended for publication.

We thank the reviewer for his comments, and we have addressed all the remaining issues.

Review 1
Mostly addressed. References are missing in the responses to the reviewer.
Answer: The mentioned reference is included in the body of the text.
Review 2
1) Addressed sufficiently
2) Needs a reference for the response to the comment around ocean emissions/regions. Issues with this statement were also highlighted within Reviewer 3's comments and this needs more substance and discussion in the text.
Answer: The mentioned reference is included in the body of the text. We have also clarified this sentence as per our response to the reviewer.
3) Addressed sufficiently
4) Addressed sufficiently
5) Addressed sufficiently
6) Addressed sufficiently
Review 3
1) Based on the importance of methane to future tropospheric ozone burden highlighted by Cooper's review (in particular with respect to the IPCC AR6 reference); this needs more attention before considering the impact of the reactive species. Discussion of methane distribution and trends and impact on ozone with reference to other studies should be included in the introduction and also the emphasis of the new paragraph should be the other way around. Suggest discussing methane importance first and then move the focus for the paper to higher reactivity precursors as this is then the new work.
Answer:
Per the reviewer's suggestions, we have added the following paragraph to the introduction: "Methane, with an assessed total atmospheric lifetime of 9.1 ± 0.9 years (Szopa et al., 2021), is also a crucial driver of tropospheric ozone(Fiore et al., 2002; Isaksen et al., 2014). Its accelerated growth rate of 7.6 ± 2.7 nmol mol^{-1} yr^{-1} between 2010 and 2019 (Canadell et al., 2021) is largely driven by anthropogenic activities (Szopa et al., 2021). NOAA GML observations of methane (NOAA, 2024) show that methane concentrations in the atmosphere have increased sharply since 2005 (an 8% increase from 2005 to 2023). Future scenarios show that emission control measures can influence future changes to air pollutants. Although the global increases in CH_4 abundance may offset benefits to surface O_3 from local emission reductions (Fiore et al., 2002; Shindell et

al., 2012; Wild et al., 2012; Szopa et al., 2021), recent reports (e.g., Itahashi et al., 2020; Zanis et al., 2022), showed the dominant role of precursor emission changes in projecting surface ozone concentrations under future climate change scenarios. In this study, we investigate the relation between ozone trends and the trends of its precursors, with a focus on NO_2, CO, and HCHO."

2) Addressed sufficiently
3) Addressed sufficiently
4) As this is a paper discussing trends for TOAR-II and contributing to the Community special issue then the guidance should be observed and as advised the authors should try and change the wording they have used from 'significant' to statements about 'confidence'.
Answer:
We have changed the significance levels to the corresponding confidence level.

5) Addressed sufficiently
6) Addressed sufficiently
7) Addressed sufficiently

8) Addressed sufficiently

Minor comments

Mostly addressed but some detail is missing in the responses, if changes have been made- and it doesn't look like it- they are not described in the response. Authors need to respond to the comments around Figure 6, line 326, line 343, Figure 14, lines 355-357.

Answer:
We have addressed all the comments related to the figures. The comment on Figure 6 was regarding the time period in the caption, which has been corrected. There was no comment on Figure 14 or the mentioned lines in the revies or track changes versions.