C2: <u>'Comment on egusphere-2024-720'</u>, Anonymous Referee #2, 22 Apr 2024, Citation:

https://doi.org/10.5194/egusphere-2024-720-RC2

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

This manuscript describes global and regional trends of tropospheric ozone and its precursors (NO2, CO, and HCHO) and aims to investigate the spatio-temporal characteristics of the ozone formation regime. Satellite observations of the tropospheric column have been used as the main data source, while ozone sonde data and GEOS-GMI model simulations have also been analyzed to study the sub-columns. The topic is central to the scope of the journal, and the trend analysis from OMI/MLS seems sufficiently valid.

However, particularly for the part analyzing ozone formation regimes, I find the authors' discussion rough and even flawed: my main argument is that without considering 1) the trends of "long-range transported" ozone, 2) the seasonality, or 3) the major components from lower/middle/upper tropospheric sub-columns that drive the trends (although treated in the model for Figure 11), the assessment of the regimes is not correct. For example, in lines 652-653, it is doubtful to conclude that most regions in the southern hemisphere are VOC sensitive regions (lines 365 and 652), simply from the fact that the trends of O3 and HCHO are decreasing while the trend with NO2 is increasing. In the atmospheric chemistry theory, VOC-limited conditions must occur where NOx is abundant (and thus OH loss is controlled by its reaction with NO2), which is unlikely for "most regions in the southern hemisphere".

Answer: We agree with the reviewer that other factors contribute to the burdens of the tropospheric ozone column such as stratosphere-troposphere exchange (STE), regional and long-

range transport of ozone precursors, reactivity, and seasonality. We have now included a discussion about possible contributions from STE as it has been recently highlighted as an important contributor to tropospheric ozone column especially in the middle and upper troposphere, and particularly in midlatitudes. We also noted in section 3.4.7 that this analysis does not consider variations of the ratios and their trends with respect to season or altitude. Regarding the chemical regimes, we have clarified that the trends of the HCHO/NO<sub>2</sub> ratio indicate moving towards a VOC or NOx sensitivity regime rather than already in a VOC or NOx-sensitive regime.

Another crude statement is made in lines 343-344, that the positive trends in the 30-60degS band are mainly driven by oceanic emissions, without any supporting results. All other parts discussing regimes need to be reviewed and reconsidered.

The statement has been further clarified (... and the positive trends in this band are contributed mainly by oceanic regions (see Error! Reference source not found.).).

The discussion on the TrC-HCHO/TrC-NO ratio (section 3.4.7) seems to be the opposite. If the ratio decreases, the chemical status must be becoming more VOC sensitive (rather than NOx sensitive, line 675). The increasing trend should indicate more NOx sensitive conditions (rather than ROx sensitive, line 678).

As explained in section 3.4.7, there are two important aspects to differentiate between, 1) the HCHO/NO ratio, 2) the trend in HCHO/NO2. The mean HCHO/NO2 ratio is shown in Figure 19. We explain in lines 657 – 665 that higher HCHO/NO2 ratio is related to NO sensitive condition as the reviewer pointed out. There is no discrepancy here.

We also added the following sentence to clarify the different interpretations of the HCHO/NO2 trends. "For example, over the eastern US and Europe, the HCHO/NO2 ratio (Figure 19) is low but the HCHO/NO2 trend is showing a slightly increasing trend indicating a direction towards the opposite, NO sensitive conditions.

However, there is a typo mistake in lines 675 and 678 that was made in the final stage of the article and was not detected before the submission. We thank the reviewer for mentioning this typo, which we have corrected.

I also did not understand why the positive emission trends with soil HONO in the southern hemisphere lead to a decrease in O3 (line 1074).

This is a typo and the sentence has been removed.

When presenting the satellite data used in Section 2.2.1, the authors need to describe what kind of data screening was applied, in particular with respect to cloud fraction and solar zenith angle. It is also necessary to describe which emission inventory was used for the GEOS-GMI model simulations has to be described (leading to an erroneous positive CO trend over East Asia).

The Satellite data products used in this article are already published and their references are listed in Table 1 and cited as appropriate in the text. The same applies to the GEOS-GMI setup and simulation results are described. In addition, we include now in section 2.2.3. a discussion and references for the emissions used in GEOS-GMI simulations.

In section 3.2, only the first authors papers are cited. It is needed to provide a more balanced citation.

More references are now cited.

Considering the importance of understanding the chemical regimes and providing valid information for the abatement strategy (line 141), I do not recommend publication of this manuscript in its current form.

The article has been revised including the reviewer's comments.