Author's Response

Dear Editor,

We are delighted to have the opportunity to resubmit a revised manuscript and would like to thank you very much for your help throughout the process. We would also like to sincerely thank the reviewer for the valuable suggestions, which we believe helped to significantly improve the quality of the manuscript. Detailed responses to the reviewer's comments are listed below, and the corresponding changes have been made in the revised manuscript based on these comments. All changes are marked in blue.

Response to Referee #1:

The authors made substantial revisions that address many of my concerns in the first round of review. I will outline several remaining comments below for the authors and the editor to consider, and I support the publication of the paper if they can be addressed.

Response:

We thank the reviewer for the comments and suggestions, which have been very helpful in improving the manuscript. The responses and revisions are listed below.

1) *Use of two satellite instruments. First, more details should be included to describe the "merging" process, either in the main text or in the supplement. To what extent did the OMI and TROPOMI data differ during 2018-2020, and how this differences were minimized during the "merging"?*

Secondly, does the NASA official OMI retrievals contain data up to 2021 to verify the trends in this paper from an "independent" perspective? The retrieval from the NASA official products are different although the same OMI radiance is used, but the consistent OMI instrument throughout the period will provide valuable verification, at least for the relative/qualitative trends. Such rigor is needed for trends from two instruments.

Finally, the TROPOMI resolution (0.01 deg) as described by the authors looks unusual, and should be checked to confirm.

Response: Accepted.

To avoid differences between the two satellite instruments, we replaced the official NASA OMI retrieval data covering 2013-2021 in the revised manuscript to study the long-term changes in NO_2 and HCHO. Tropospheric NO_2 vertical columns were obtained from OMI/Aura Level-3 NO_2 products (OMNO2d 003) with a grid resolution of 0.25°×0.25°, while HCHO total columns were obtained from OMI/Aura Level-3 HCHO products (OMHCHOd 003) with a grid resolution of 0.1°×0.1°. We recalculated the trends. Although the values have changed compared to the previous manuscript, the conclusions remain consistent.

The high spatial resolution TROPOMI data is still used to derive the HCHO/NO_2 threshold values marking transitions in O_3 formation regimes and to diagnose the ozone-NO_x-VOC sensitivity in China. After verification, the TROPOMI NO2 and HCHO resolution from Earth Engine is 1113.2 meters, which is about 0.009° in the China region. We have made the corresponding changes.

We revised the relevant description in Lines 101-109:

“The TROPOMI data from the Earth Engine Data Catalog are based on the algorithm developments for the QA4ECV reprocessed dataset for OMI and have been further optimized. The TROPOMI data, available for 2019-2021, are processed with a spatial resolution of 1113.2 meters (about 0.009∘ within China). The same chemistry transport model for HCHO and NO_2 is better suited for analyzing their ratio than products developed with different prior profiles.

The OMI data with longer time horizons (2013-2021) are used to study the long-term changes in NO_2 and HCHO and track changes in emissions of NO_x and VOCs. Tropospheric NO_2 vertical columns are obtained from OMI/Aura Level-3 NO_2 products (OMNO2d 003) with a grid resolution of 0.25°×0.25°, while HCHO total columns were obtained from OMI/Aura Level-3 HCHO products (OMHCHOd 003) with a grid resolution of 0.1°×0.1°. Since HCHO mainly presides in the troposphere, its total column can be regarded as the tropospheric column (Duncan et al., 2010).”

We revised Figures 5 and 6, as well as the data results in the manuscript.

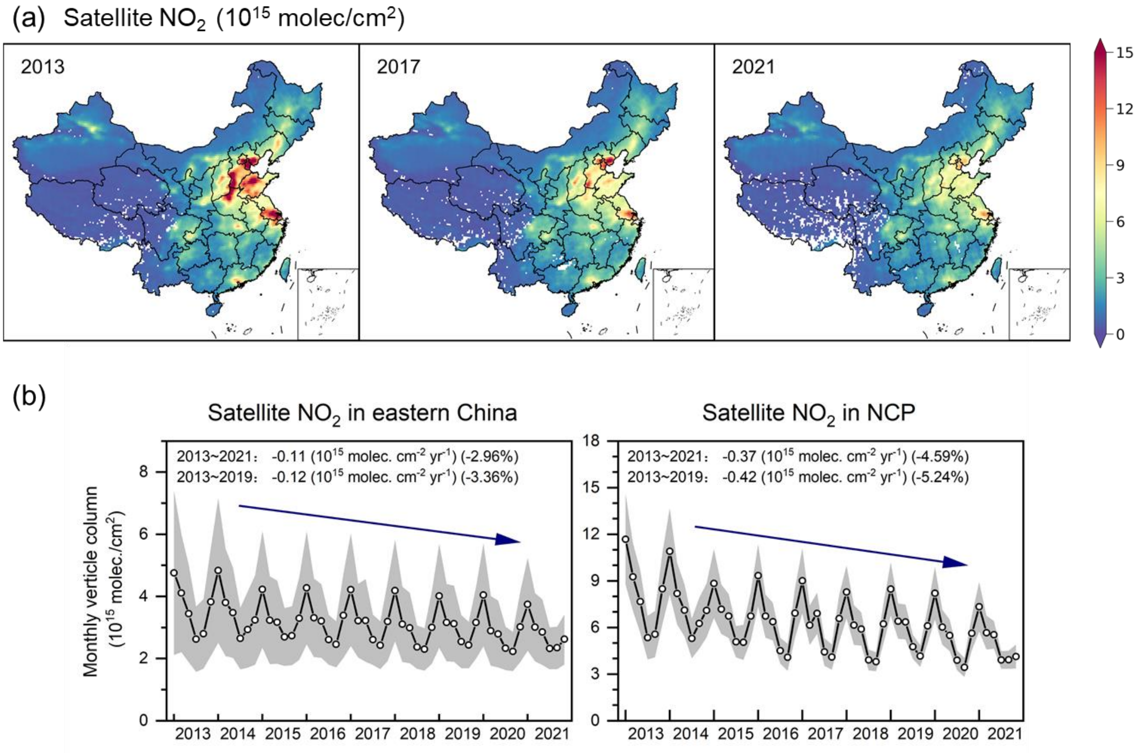

Figure 5: (a) Maps of average satellite-based NO_2 columns over China from April to September, and (b) monthly mean NO_2 columns averaged over eastern China and North China Plain in April-September. Gray shading: mean value ± 50% standard deviation across all grids for each month. Inset: absolute annual linear trend and percentage of annual trend (% per year, the linear trend divided by the 2013 mean values).

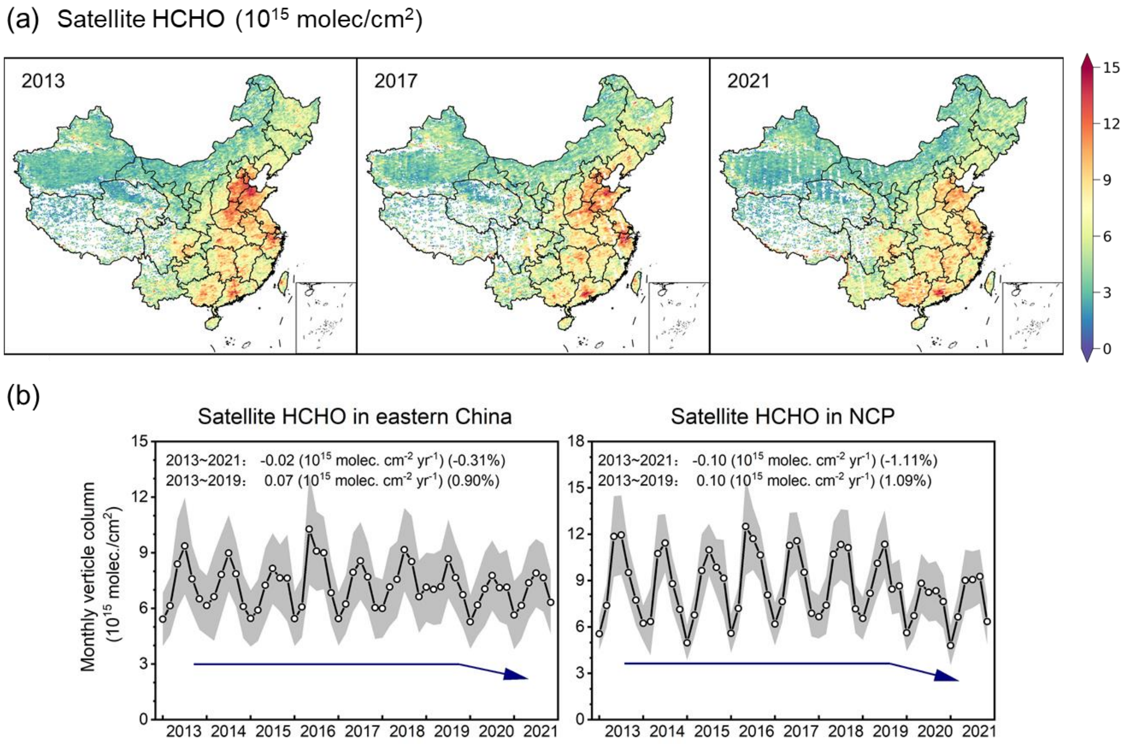

Figure 6: Same as Figure 5 but for satellite-based HCHO columns.

2) *Since this paper extends previous investigations, it will be valuable to include one paragraph in the discussion section to summarize comparison of the findings in this paper vs. the previous ones, and highlight new findings and implications from this study.*

Response: Accepted.

To emphasize the new findings and significance of this study, we made additions and changes in Lines 313-324 of the revised manuscript,

"Based on the evaluation of the nonlinearity of O_3-NO_x-VOC chemistry captured by space-based HCHO/NO_2, this study derives thresholds marking the transitions between chemical regimes in different regions of China and diagnoses the current spatial distribution of O_3 production regimes. To reveal the causes of O_3 increases, O_3 responses to precursors changes are evaluated by tracking VOCs and NO_x with satellite HCHO and NO_2.

Results showed that the HCHO/NO_2 ranges of transition from VOC-limited to NO_x-limited regimes vary apparently among Chinese regions, which is inconsistent with previous studies (Wang et al., 2021; Li et al., 2021). The higher resolution TROPOMI satellite data enables a better match between the location of ground-based O_3 monitoring stations and the grid of satellite data, thus allowing a more accurate derivation of HCHO/NO_2 thresholds and reflection of ozone-NO_x-VOC sensitivity. For April-September 2021, VOC-limited regimes are widely found over megacity clusters (NCP, YRD, and PRD) and concentrated in developed cities (such as Chengdu, Chongqing, Xi'an, and Wuhan). NO_x-limited regimes dominate most of the remaining areas. Moreover, the high-resolution TROPOMI data can more accurately resolve strongly VOC-limited conditions in urban cores."

and in Lines 333-348:

"Identifying the causes of ozone increases since 2013 is crucial for effectively controlling the ozone pollution in China. From 2013 to 2021, satellite NO_2 and HCHO columns showed an annual decrease of 3.0% and 0.3%, respectively, indicating an effective reduction in NO_x emissions alone without effective VOC control. …

In summary, the significant reduction in NO_x alone without effective control of VOC, combined with the effect of the decrease in $PM_{2.5}$ mentioned in previous studies (Li et al., 2019; Li et al., 2022), has led to an increase in O_3 in major regions in China."

3) *Section 3.4 still reads loosely connected with the previous results to me. The selection of locations and time (e.g. April in Beijing and May in Chengdu) reads random, and unrepresentative of COVID. My suggestion is this section can be cut. The authors can develop this idea into an independent paper. I leave this comment to the editor to consider.*

Response: Accepted.

Considering the consistency of the topic related to long-term changes, we have removed the section on cases during COVID-19 in the revised manuscript.

4) *Line 368-373: In the Li et al. (10.5194/acp-20-11423-2020, 2020) paper, the ozone trends over NCP (2013-2019) is 3.3 ppb/yr, in which 1.4 ppb/yr is attributed to meteorology. So I do not support the authors description here by simply comparing numbers. 1 ppb/yr is a significant component of 3.3 ppb/yr. I suggest to revise these words to state that "PM, VOC, and NOx are all very important anthropogenic drivers of ozone trends in China", which is also supported by a recent paper (Li et al., 10.1021/acs.est.2c03315, 2022).*

Response: Accepted.

Our study highlights that the increase in ozone in major areas is due to significant reductions in NO_x only without effective VOC control. In the revised manuscript, we modified the relevant expressions as follows in Lines 346-348: "In summary, the significant reduction in NO_x alone without effective control of VOC, combined with the effect of the decrease in

$PM_{2.5}$ mentioned in previous studies (Li et al., 2019; Li et al., 2022), has led to an increase in O_3 in major regions in China."

We also revised the relevant expressions in the Introduction (Lines 59-61): "Recent O_3 trends in China have been driven by a variety of anthropogenic factors. Some model simulations revealed a strong influence of the $PM_{2.5}$ decrease on the O_3 increase and attributed that response to the aerosol sink of hydroperoxy (HO_2) radicals (Li et al., 2019)."