Response to Reviewer 1:

The manuscript titled “TROPOMI NO_2 for urban and polluted areas globally from 2019 to 2024” presents a comprehensive analysis on NO_2 VCD changes in cities worldwide. It details the contrasts in NO_2 trends across cities, and potential drivers the embedded anthropogenic emissions, including environmental regulation, local economic growth and regional conflicts. Although the study illustrates the latest evolution of global air pollution, and offers a valuable reference for future research, the manuscript, in its current form, contains several critical issues that warrant major revisions. Therefore, I recommend reconsideration for its publication after the authors adequately address the concerns outlined below.

We sincerely thank the reviewer for the thoughtful review to improve the quality of this manuscript.

The current manuscript lacks a discussion of the uncertainty of NO_2 VCDs and its potential impacts on the conclusions. This information is crucial for distinguishing trends from interannual fluctuations, and for separating meaningful emission changes from the noise inherent in satellite retrievals. However, uncertainty considerations are absent from the main text and figures. In addition, further validation of the NO_2 background values is necessary, along with sensitivity tests (e.g. evaluating the results using different percentile thresholds in the background selection). The interannual variability of the background should also be evaluated (e.g. in Fig. 12), as this could influence the interpretation of relative changes in VCD enhancements. Moreover, the spatial consistency of the background should be examined, particularly in regions where adjacent cities are expected to share similar background levels.

We appreciate this feedback, and have made the following additions to the manuscript to address these concerns:

1. We have now added statistical significance testing to all trend plots. Significance is determined through linear regression on monthly de-seasonalized TROPOMI time series (see revised Sec. 2.3). We successfully show that statistically significant trends can be separated from insignificant trends from the monthly TROPOMI data from 2019 to 2024, (e.g. in Figures 2-5), and that this time period is not too short.
2. To address general uncertainty within TROPOMI retrievals, we added text to note the 15-20% relative uncertainty related to monthly and annual averages, as highlighted in a recent previous study (Glissenaar et al., 2025). We also added text to mention the systemic biases of operational TROPOMI NO_2 retrievals, that can lead to underestimated NO_2 VCDs over highly polluted regions (-31.4% bias for values $>15\times10^{15}$) and overestimated (+26.5% bias) NO_2 VCDs in less-polluted regions with VCDs $<2\ \times10^{15}$ (Lambert et al., 2025). To address uncertainty associated with estimates at the city and country level, we have included uncertainty estimates when reporting relevant values.
3. We thank the reviewer for the suggestion to re-evaluate the method used to quantify the background NO_2 concentration. We have added a section to the supplementary document (Section S1 Sensitivity of Urban Background NO2 VCDs) in which we conduct a sensitivity test to evaluate the impact of using different percentile thresholds on the results related to the VCD enhancement (previously Figure 12, which has now been split up into multiple figures). In that supplementary section, we test using different percentile thresholds as the background concentration and find that changing the used percentile does not meaningfully impact our results nor change the directionality of the trends. Following this analysis, we find that the 50th percentile as the threshold is an adequate choice for most

cities (See Supplementary information). In that same section we highlight test case of large adjacent cities and how the background concentrations for those cities vary. Although more sophisticated methods of background quantification exist (Fioletov et al., 2025) we find that using a percentile as an assumed background concentration is an acceptable choice given the large number of cities being evaluated (N >11,000).

4. Based on comments from this and the other reviewers, we have made substantial changes to the revised manuscript, by rearranging many of the figures and sections for clarity. We are confident these changes greatly improve the quality of this work.

The manuscript includes several qualitative descriptions that are not supported by sufficient validation or statistical testing. For instance, it states that there is an accelerated decreasing trend in NO_2 VCDs in both China and European countries. However, given that the dataset used in this study begins in 2019, the time range may be too short to detect or validate such trend acceleration. Similarly, the manuscript mentions an accelerated NO_2 increase over Moscow in early 2022. Yet, Fig. S9 appears to show only a brief, anomalous spike in NO_2 VCDs, followed by a return to typical levels. These interpretations, as currently presented, are questionable and require rigorous statistical validations.

We have greatly expanded the statistical testing within this iteration of the manuscript and have removed any claims of "accelerating" trends. Significance is determined through linear regression on de-seasonalized TROPOMI time series (see revised Sec. 2.3). We successfully show that statistically significant trends can be separated from insignificant trends from the monthly TROPOMI data from 2019 to 2024 (e.g. in Figures 2-5), and that this time period is not too short.

The manuscript appears to insufficiently account for the effects of seasonality on NO_2 VCDs. Given the strong seasonal variation in NOx lifetime, particularly the longer lifetime during winter, NO_2 VCDs in colder months can disproportionately influence interannual trends if seasonality is not properly addressed. However, the manuscript lacks adequate discussion or correction for these seasonal effects. Moreover, there appears to be a mischaracterization of seasons between the Northern and Southern Hemispheres. For instance, the manuscript uses data from the same calendar months to represent winter conditions in both Asia and Oceania. This approach is problematic, as most cities in Oceania are located in the Southern Hemisphere, where the seasonal cycle is inverted. As a result, the analysis may misrepresent seasonal trends in these regions, and further clarification or adjustment is necessary.

We thank the reviewer for addressing the question of seasonality in our work. To address these points, we have:

1. Replaced any existing monthly time series within this study with de-seasonalized trends. This was done for Figure 12 (previously Figure 3) and Figure 3 (previously Figure 8).
2. We have now included a more robust analysis of statistical significance throughout the manuscript. We use the de-seasonalized data to quantify significance of any trends.
3. We have addressed the discrepancy between Northern and Southern hemispheric cities by removing and reference to "warmer months" or "colder months" or season names, particularly when discussing South America, Africa or the lumped Asia and Oceania section. We instead refer to the months of observations, e.g. May-September.

Specific Comments

Page 2, Line 56-67: I would suggest to include a brief overview about NO_2 VCD changes in India, Oceania and Africa here, since these regions also play important roles in this study.

We appreciate this suggestion. We have added the following text at line 67 of the revised manuscript:

"In contrast, urban regions of India have shown NO_2 increases over the past few decades, linked to urbanization and energy demand growth (Hilboll et al., 2013; Ghude et al., 2020). Over Africa and South America, NO_2 VCD trends through the mid-2010s have been less pronounced, reflecting limited industrialization and more dominant contributions from biomass burning and natural sources (Geddes et al., 2016; Castellanos et al., 2014)."

Page 2, Line 59: "x" --> "×". Check throughout the manuscript.

Thank you for pointing this out. We have made the change to all mentions within the manuscript as well as any figures using the notation.

Page 4, Line 97: It should be explained why GHS-SMOD boundaries are used rather than administrative city boundaries, and clarify whether this choice affects the results.

Thank you for this question. To our knowledge, a dataset of true administrative / legislatively determined boundaries for all urban regions globally does not exist. We have added the following text at line 102 of the revised manuscript to emphasize the value of using GHS-SMOD:

"GHS-SMOD has the benefit of providing a globally consistent, satellite-derived definition of built-up areas, whereas administrative boundaries vary widely in definition and availability. Using physical built-up area boundaries from GHS-SMOD instead of administrative ones may shift the absolute spatial extent of some cities, but it does not materially alter the concentrations calculated in this study."

Page 4, Line 111: According the latest ATBD (2.8.0, 2024-11-18– released) for TROPOMI NO_2, the nadir ground pixel dimensions were 7.0×3.5 km^2 before 6 August 2019. The data description here is inaccurate.

Thank you for pointing this out. We have modified the text at line 121 of the revised manuscript to reflect the difference in spatial resolution before 06 August, 2019:

"These Level 2 data have a nadir spatial resolution of 3.5 x 7.0 km^2 before and 3.5 x 5.5 km^2 after 06 August 2019."

Page 6, line 142: Sensitivity tests should be conducted to assess the impact of using different percentile values in background selection. In addition, validation is needed. For example, by examining whether background values are consistent across adjacent cities.

We appreciate this suggestion. We have added a section to the supplement titled "S1 Sensitivity of Urban Background NO_2 VCDs", where we (1) describe the method used to quantify background concentrations, (2) provide results of a sensitivity analysis evaluating the impact of different percentiles on the general conclusions we've drawn within this manuscript, (3) an evaluation of trends in urban background VCDs over time and (4) the impact and evaluation of background for adjacent cities / clusters of cities. In short, following this thorough analysis, we have updated the choice of background concentration to the 50th percentile. We test using different percentile thresholds as the background concentration and generally find that changing the used percentile does not meaningfully impact our results nor change the directionality of the trends. Please refer to the revised methods section (Sec. 2.4) and the supplement document for more information.

Page 6, Line 157: The claimed acceleration in the decreasing trend requires statistical validation; otherwise, such descriptions might be just removed. (Also, for the descriptions on Page 10, Line 239, Page 10, Line 221, and Page 14, Line 310)

Based on this recommendation, we have changed the phrasing of this specific text to no longer claim an accelerated trend, but rather a continued decreasing trend. We note that the statistical analysis in this revised submission has been greatly expanded, and there are now many references to statistical significance of trends throughout this work.

Page 7, Line 164: Please clarify the definition of the mining regions (including A, C in Fig. 2; B in Fig. 4; D, E in Fig. S4; and G, F, H, I in Fig. 6).

We have added the description of the mining regions within the captions of each figure (Figures 6-11 in the revised manuscript)

Page 7, Line 166: The texts in Fig. S3 are not clear.

We have increased the text size in the legends in the top left of each panel. This is now Figure S20.

Page 8, Figure 3: The information of NO_2 VCD uncertainty and significance tests on the regression is missing. In addition, please ensure consistency of significant figures or decimal precision for all numerical data throughout the manuscript.

Thank you for this suggestion. We have now changed this figure to show de-seasonalized trends as a monthly % anomaly, from which trends can more clearly be quantified and visualized. For each panel, we have added the statistical significance of the trend by reporting a percent change per year, as well as a p-value. We have also updated the manuscript to ensure the use of consistent significant figures. See Sec. 2.3.

Page 9, Line 197: Please provide the specific number and proportion ("Nearly all").

Thank you for this suggestion. We have since removed this text for the sake of shortening the manuscript. However, we calculated that 66 of the 71 urban clusters in Eastern Russia (or 93%) exhibited larger 2024 NO_2 VCDs than in 2019.

Page 9, Figure 4: I would suggest standardizing the formatting of units throughout the manuscript for consistency.

We have double checked the manuscript to ensure that the units (e.g. 10^{15} Molecules cm^{-2}) are used consistently throughout the manuscript. If this is in reference to the range of plotted data on the colorbars (e.g. -1 to1 in Fig. 4 vs. -2 to 2 in Fig. 6), we find that different regions require different values on the color bar for the sake of effective data visualization, as not all regions experience a similar range of concentrations.

Page 10, Line 232: What is the term “largest” referring to or being compared against in this description? (other cities or other land type? Also, for the descriptions on Page 11, Line 248, Page 11, Line 253-254, Page 12, Line 263-264 and Page 12, Line 270)

These are directly in reference to the largest observed annual mean concentrations or differences in the spatial figures (Now Figures 6-11 in Sections 5.1 – 5.5; largest concentration refers to data plotted in panel a, while largest increase/decrease refers to data plotted in panel b). In the case of Europe as is referenced here, we are claiming that the largest values occur in urban settings, as opposed to non-urban settings (See Fig. 8a). In reference to Moscow, we are stating that the largest concentrations in Europe were observed over Moscow. Hypothetically, a power plant, or coal mine or some other source outside of the urban environment could in theory be the largest signature for a region. We are simply noting that the largest signatures observed here are in urban environments. We have made sure that these figures are properly referred to in the revised manuscript.

Page 10, Line 236: Please provide the specific number.

We have added the modified the text on line 354 of the revised manuscript, which now reads:

“Of the 1257 urban clusters in Europe, 1007 (80%) exhibited larger VCDs in 2024 than in 2019. Of the 53 European urban clusters with a population greater than 1,000,000, 2024 VCDs were lower than 2019 VCDs in 48 (91%), with the exception of Moscow and other cities of western Russia, which experienced increases (Fig. 9b)”

Page 12, Section 4: I would suggest to integrate Section 3 and Section 4.

Thank you for this suggestion. We have largely restructured the manuscript by combining multiple sections, although the content generally remains the same. Section 3 now highlights trends in major urban areas, Section 4 quantifies country-level, population-weighted trends, Section 5 highlights continental signatures and seasonality, and Section 6 highlights trends in major oil, gas and metal mining regions.

Page 13, Figure 7: The figure legend could be further improved to enhance readability.

We have increased the size of the legend for readability.

Page 14, Line 311: Typo.

Thank you, we have fixed the typo.

Page 14, Line 311: The abnormally high NO_2 VCD values require further examination to exclude artifacts, including applying data filters based on Level-2 QA flags. It should also be verified whether any spurious outliers affect the averaging process.

The oversampled level 3 TROPOMI NO_2 product we use in this study are quality controlled, with the recommended QA flag > 0.75 applied to the L2 data before oversampling. It removes cloud-covered scenes (cloud radiance fraction> 0.5), partially snow/ice covered scenes, errors, and problematic retrievals. The number of valid observations for the Moscow urban cluster during March 2022, when a monthly average value of 59 × 10^{15} molec. cm^{-2} was calculated, had more observations (233) than the median March (159) in the six-year record. Despite the abnormally high value, this appears to be a valid mean value for that month.

Page 15, Figure 8: The figure labels/text are not clear.

We have increased the size of the text for readability.

Page 17, Line 349-350: Such causal relationships require careful validation. I recommend revising the statement here.

Thank you for pointing this out. Numerous studies have identified the impact of the shutdowns in China on VCDs within the country, which we cite on line 274 of the revised manuscript (Zheng et al., 2021; Cooper et al., 2022; Levelt et al., 2022; Ma et al., 2023; Zhao et al., 2024), to make clear this is a reference to previous studies and not determined from our work alone.

Page 18, Line 376: It is not immediately clear why population-weighted VCDs are preferred here over direct NO_2 VCDs for me. Would directly showing NO_2 VCDs make major differences?

We choose to use population-weighted VCDs when aggregating by region (continent or country), as it more accurately reflects the column concentration that people in a region's urban areas are exposed to. An equitable spatial average would give unnecessary weight to concentrations in less populous cities where fewer people live and the relative satellite uncertainties are larger.

Page 21, Line 433: Since the comparison here is based on the relative changes of VCDs and emissions with respect to 2019, it is hard to conclude that emissions are underestimated. At most, it may suggest a possible underestimation in the emission trend. (Also for Page 24, Line 481)

We apologize for the unclear language. We never meant to say that the emissions are underestimated in totality but instead underestimated trends. We have modified the text to reflect the year-to-year variability (i.e. trend) in the emissions inventory is likely underestimated based on discrepancies between its trends and TROPOMI's trends now on line 340:

"Evaluating trends in NOx emissions inventories in African cities, we find a mean difference of -8.0% (EDARv8.1) and -6.7% (CEDS) between inventory NOx emission trends and VCD_{ENH} trends, indicating a potential underestimate in both emissions inventories in African cities for this period."

Page 21, Line 435: Impacts of uncertainty in VCD background need to be quantified.

We thank the reviewer for this suggestion. We have added a section to the revised supplement document that provides an analysis of the urban background concentrations. Please refer to the revised supplement document Section S1.

Page 21, Line 435: The mean difference is likely underestimated due to the inclusion of 2019.

Thank you for pointing this out. As 2019 has a value of 0, we have ensured that data from that year are not included in the average, and we have updated the relevant values.

Page 22, Line 451: There appears to be a mischaracterization of seasons between the Northern and Southern Hemispheres, since Asia and Oceania are shown together in Fig. 13.

Thank you for noting this. We have modified the text throughout the manuscript to refer to the months explicitly (e.g. May-September), as opposed to “warmer months” or “summer”, particularly when referring to South America, Africa and Oceania.

Page 23, Figure 13: Is the sharp increase during the winter of 2022 primarily driven by anomalously high values over Russia? If so, the authors should consider presenting additional results with Russia excluded. Intuitively, I find that this sharp increase appears inconsistent with Fig. 9c, where most cities do not show a similar increase in 2022.

Thank you for bringing up this point. The sharp increase observed in winter 2022 in Europe is indeed due to increases in Russian cities. We have created a version of this figure with Russian cities removed and added to the supplement as Figure S18, and added the following text to line 373 of the revised/tracked changes manuscript:

“We note that the seasonal trends in Europe show notable winter and summer decrease if evaluating trends with Russian cities removed (Fig. S18).”

The noted difference in the magnitude for Europe in 2022 between previous Figure 13 and Figure 9 is due to the fewer data points during winter. When averaged over the colder months, the 2022 increase is stark, as seen in Figure 13. When averaging to an annual value, the higher number of observations in the warmer months, when an increase was not observed, dilutes the impact of the winter values.

Page 24: Line 481: Discussion about the impacts of NOx chemistry and its seasonality should be included.

Thank you for this suggestion. We have added the following text to line 510 of the revised manuscript:

“Additionally, while many of the trends presented here reflect changes in anthropogenic NOx emissions, it is important to recognize that atmospheric chemistry also influences the observed NO_2 variability. Seasonal differences in photochemical lifetimes (i.e., longest in winter), boundary layer mixing (i.e., more vertical mixing in summer), chemical partitioning between NO and NO_2 (i.e., the fraction of NO_2 is largest in winter) and meteorological variability can all modulate the magnitude and timing of observed trends. These processes likely contribute to some of the regional and seasonal differences highlighted in this study.”

Page 24, Line 491-492 (“tall-stack sources”): Could the authors provide supporting references for this statement?

We have added the citation to line 506 (Brett et al., 2025).

References

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