General comments:

This paper presents a comprehensive assessment of urban NO_2 changes worldwide from 2019 to 2024 using TROPOMI NO_2 VCD observations. Differences in NO_2 VCD changes over populous cities and broader areas are disclosed, probably driven by anthropogenically induced factors such as urbanization, industrial activities, government interventions, and societal disruptions. The paper also attempts to quantify the influence of background NO_2 and NO_2 seasonal variability on the trend analysis. The research topic fits in the scope of ACP, and the manuscript is already in good shape. I recommend its publication after the authors address the following comments.

We thank the reviewer for this thoughtful review.

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 that these changes greatly improve the quality of this work.

Specific comments:

Line 1-2: The current title is a little general and can not convey the key point of this research. I would suggest to improve the title by using the key conclusion of this study, which can better draw readers' attention.

We appreciate this suggestion. We have changed the title of the manuscript to "Global NO_2 Trends from TROPOMI (2019–2024): Urban Changes and Emerging Hotspots".

Line 39-40: The remote sensing method not only relies on spectrometers aboard satellites to infer vertical columns, but also can infer vertical profiles using ground-based instruments. The statement should incorporate the profile retrieval to ensure a more comprehensive description.

Thank you for this suggestion. We have modified the text on line 44 of the revised manuscript to reflect the different types of remote sensing methods that exist for NO_2:

"NO_2 can also be remotely-sensed from ground-based instruments capable of inferring vertical profiles of NO_2, such as using multi-axis differential optical absorption spectroscopy, MAX-DOAS (Vlemmix et al., 2010)"

Line 68-75: This paragraph only describes the different methods used to characterize the urban extent. The authors should add a brief discussion about the pros and cons of these methods, and highlight the advantage(s) of the GHS-SMOD, which is used in this study.

Thank you for this suggestion. We have added the following text to the methods section (Line 102) to highlight the value of 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."

Section 2.2: Please briefly describe the uncertainty of the NO_2 VCD product used here.

Thank you for this suggestion. 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 TROPOMI NO_2 retrievals, that can lead to underestimated NO_2 VCDs over highly polluted regions, and overestimated NO_2 VCDs in less-polluted regions (Lambert et al., 2025). To address uncertainty associated with estimates at the city and country level, we have added error bars to relevant figures and include uncertainty estimates when reporting relevant values.

We have added text to line 129 of the revised manuscript, highlighting the most common areas of uncertainty related to NO_2 retrievals.

"TROPOMI NO_2 retrievals are subject to measurement and retrieval uncertainties that propagate into the oversampled Level 3 products. Typical uncertainties in monthly or annually averaged tropospheric NO_2 vertical column densities are on the order of 15–20 %. Systematic biases have also been reported, with overestimation in less polluted regions (+26.5% bias) and underestimation in areas with high NO_2 concentrations (-31.4% bias), reflecting limitations in the retrieval process (Glissenaar et al., 2025; Lambert et al., 2025)."

Additionally, 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).

Section 2.2.1: The structure here is a little inappropriate because there is only one sub-section. I would suggest to merge Section 2.2.1 and Section 2.2 to one section.

We have removed subsection 2.2.1 and turned it into a separate section 2.3 and changed existing sections 2.3 and 2.4 to 2.4 and 2.5, respectively.

Line 141-142: Please justify the definition of the background NO_2 concentration here, and provide the sensitivity of the results in Section 6 to the choice of the percentile.

Thank you for this suggestion. 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).

Section 2.4: Please briefly describe the uncertainty of the EDGARv8.1 NO_x emissions.

At the suggestion of a separate reviewer, we have included an evaluation against CEDS as well as EDGAR. We have added the following text to line 176 of the revised manuscript (now in Section 2.5) describing general uncertainties in these bottom-up inventories:

“Uncertainties are inherent in such emissions inventories, with a roughly 10-50% uncertainty when aggregating emissions to the country level, and even larger uncertainty for individual grid points (Crippa et al., 2018).”.

Line 157-158: Is the statement “the decrease accelerated after the onset of the COVID-19 pandemic” one of the findings of this study, or a knowledge cited from other papers? If the former is true, please provide a quantitative discussion to support this point; if the latter is true, please provide supporting references.

Thank you for allowing us to clarify this point. We have modified the text to note “a continued decreasing trend”, as opposed to an accelerated trend.

Figure 3, Figure 8 and Figure S6: please provide the confidence level of each regression to clarify the statistical significance of the characterized trends.

Thank you for this suggestion. For each of these monthly level examples, 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).

Line 305-306: Please provide a quantitative discussion to support the statement “The observed annual decreases in these East Asian cities were primarily driven by decreases during the winter months”.

We have removed this sentence from the revised manuscript.

Line 310: It is difficult to see that the increasing trend in Moscow accelerated in early 2022 from Figure S9, except that NO_2 VCDs in winter time jumped to a higher level. Please provide a quantitative discussion to demonstrate the acceleration.

That is a good point. We have changed the text to no longer claim that the trend is accelerating, now on line 215 of the revised manuscript.

“This increase was accompanied by anomalously high monthly mean concentrations in early 2022 (Fig. S11), following the onset of the Russia-Ukraine war in Ukraine, when monthly mean NO_2 VCDs for March reached 59×10^{15} molecules cm^{-2} (see Sec. 3.3)”

Section 6: please provide a summary of this section, i.e., to what extent the influence of background NO_2 and seasonal variability can be on the analysis of urban NO_2 trends presented above?

Thank you for this suggestion. 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).

Line 446-456 and line 482: It should be careful to define May – September and November – March as either "warm" or "cold" months, given the different hemispheres in which the continents are located. The interpretation of the results for Asia and Oceania is problematic, because May – September is summer time for Asia but is winter time for Oceania, while November – March is winter time for Asia but is summer time for Oceania. Please revise the discussions here and in Section 7.

Thank you for this suggestion. We have modified the text related to time of year by only referring to the months used (e.g. May-September) as opposed to referring to a time of year as warmer or colder.

Technical comments:

Line 50: Please check through the manuscript and replace the "x" with a times symbol at corresponding places.

We have replaced all "x" with an "×" symbol both within the text and in each relevant figure.

Line 57: "SCHIAMACY" should be "SCIAMACHY".

We have fixed the error (now line 49).

Line 63: The statement "NO_2 concentrations increased through roughly 2005" is a little confusing. I would suggest to rephrase this sentence to make it clearer.

We have modified the text on line 65 of the revised manuscript, which now reads:

"In the United States, NO_2 concentrations generally exhibited a decreasing trend from 2005 through the mid-2010s…".

Line 94: It is better to be 1 × 1 km^2.

We have made the change (now line 100).

Line 127: Please add a period after “approach”.

Thank you for catching this. We have added the period.

Line 142: It is clearer to extend “UC” to “urban cluster”.

Thank you, we have made that change.

Line 148: Do you mean “Sec. 2.2.1” here?

That is correct, thank you. We have since modified this to read “Sec. 2.3”.

Line 183: “select” to “selected”.

We have made the change to the Figure 3 caption.

Line 306: There is no Figure 7d.

Thank you for catching this. This was supposed to read Figure 8d, not 7d. We have changed this in the text.

References

Crippa, M., Guizzardi, D., Muntean, M., Schaaf, E., Dentener, F., Van Aardenne, J. A., Monni, S., Doering, U., Olivier, J. G. J., Pagliari, V., and Janssens-Maenhout, G.: Gridded emissions of air pollutants for the period 1970-2012 within EDGAR v4.3.2, Earth Syst Sci Data, 10, 1987–2013, https://doi.org/10.5194/ESSD-10-1987-2018, 2018.

Fioletov, V., Mclinden, C. A., Griffin, D., Zhao, X., and Eskes, H.: Global seasonal urban, industrial, and background NO2 estimated from TROPOMI satellite observations, Atmos. Chem. Phys, 25, 575–596, https://doi.org/10.5194/acp-25-575-2025, 2025.

Glissenaar, I., Boersma, K. F., Anglou, I., Rijsdijk, P., Verhoelst, T., Compernolle, S., Pinardi, G., Lambert, J.-C., Van Roozendael, M., and Eskes, H.: TROPOMI Level 3 tropospheric NO 2 dataset with advanced uncertainty analysis from the ESA CCI+ ECV precursor project , Earth Syst Sci Data, 17, 4627–4650, https://doi.org/10.5194/ESSD-17-4627-2025, 2025.

Vlemmix, T., Piters, A. J. M., Stammes, P., Wang, P., and Levelt, P. F.: Retrieval of tropospheric NO2 using the MAX-DOAS method combined with relative intensity measurements for aerosol correction, Atmos Meas Tech, 3, 1287–1305, https://doi.org/10.5194/AMT-3-1287-2010, 2010.