

1 **RESPONSE TO REVIEWER 1:**

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3 The manuscript titled “TROPOMI NO₂ for urban and polluted areas globally from 2019 to 2024”
4 presents a comprehensive analysis on NO₂ VCD changes in cities worldwide. It details the
5 contrasts in NO₂ trends across cities, and potential drivers the embedded anthropogenic
6 emissions, including environmental regulation, local economic growth and regional conflicts.
7 Although the study illustrates the latest evolution of global air pollution, and offers a valuable
8 reference for future research, the manuscript, in its current form, contains several critical issues
9 that warrant major revisions. Therefore, I recommend reconsideration for its publication after the
10 authors adequately address the concerns outlined below.

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

12 The current manuscript lacks a discussion of the uncertainty of NO₂ VCDs and its potential
13 impacts on the conclusions. This information is crucial for distinguishing trends from interannual
14 fluctuations, and for separating meaningful emission changes from the noise inherent in satellite
15 retrievals. However, uncertainty considerations are absent from the main text and figures. In
16 addition, further validation of the NO₂ background values is necessary, along with sensitivity tests
17 (e.g. evaluating the results using different percentile thresholds in the background selection). The
18 interannual variability of the background should also be evaluated (e.g. in Fig. 12), as this could
19 influence the interpretation of relative changes in VCD enhancements. Moreover, the spatial
20 consistency of the background should be examined, particularly in regions where adjacent cities
21 are expected to share similar background levels.

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

- 24 1. We have now added statistical significance testing to all trend plots. Significance is
25 determined through linear regression on monthly de-seasonalized TROPOMI time series
26 (see revised Sec. 2.3). We successfully show that statistically significant trends can be
27 separated from insignificant trends from the monthly TROPOMI data from 2019 to 2024,
28 (e.g. in Figures 2-5), and that this time period is not too short.
- 29 2. To address general uncertainty within TROPOMI retrievals, we added text to note the 15-
30 20% relative uncertainty related to monthly and annual averages, as highlighted in a
31 recent previous study (Glissenaar et al., 2025). We also added text to mention the
32 systemic biases of operational TROPOMI NO₂ retrievals, that can lead to underestimated
33 NO₂ VCDs over highly polluted regions (-31.4% bias for values $>15 \times 10^{15}$) and
34 overestimated (+26.5% bias) NO₂ VCDs in less-polluted regions with VCDs $<2 \times 10^{15}$
35 (Lambert et al., 2025). To address uncertainty associated with estimates at the city and
36 country level, we have included uncertainty estimates when reporting relevant values.
- 37 3. We thank the reviewer for the suggestion to re-evaluate the method used to quantify the
38 background NO₂ concentration. We have added a section to the supplementary document
39 (Section S1 Sensitivity of Urban Background NO₂ VCDs) in which we conduct a sensitivity
40 test to evaluate the impact of using different percentile thresholds on the results related to
41 the VCD enhancement (previously Figure 12, which has now been split up into multiple
42 figures). In that supplementary section, we test using different percentile thresholds as the
43 background concentration and find that changing the used percentile does not
44 meaningfully impact our results nor change the directionality of the trends. Following this
45 analysis, we find that the 50th percentile as the threshold is an adequate choice for most

46 cities (See Supplementary information). In that same section we highlight test case of
47 large adjacent cities and how the background concentrations for those cities vary.
48 Although more sophisticated methods of background quantification exist (Fioletov et al.,
49 2025) we find that using a percentile as an assumed background concentration is an
50 acceptable choice given the large number of cities being evaluated ($N > 11,000$).

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

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

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

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

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

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

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89 Specific Comments

90 Page 2, Line 56-67: I would suggest to include a brief overview about NO₂ VCD changes in India,
91 Oceania and Africa here, since these regions also play important roles in this study.

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

94 “In contrast, urban regions of India have shown NO₂ increases over the past few decades, linked
95 to urbanization and energy demand growth (Hilboll et al., 2013; Ghude et al., 2020). Over Africa
96 and South America, NO₂ VCD trends through the mid-2010s have been less pronounced,
97 reflecting limited industrialization and more dominant contributions from biomass burning and
98 natural sources (Geddes et al., 2016; Castellanos et al., 2014).”

99 Page 2, Line 59: “x” --> “x”. Check throughout the manuscript.

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

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

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

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

112 Page 4, Line 111: According the latest ATBD (2.8.0, 2024-11-18– released) for TROPOMI NO₂,
113 the nadir ground pixel dimensions were 7.0 × 3.5 km² before 6 August 2019. The data description
114 here is inaccurate.

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

117 “These Level 2 data have a nadir spatial resolution of 3.5 x 7.0 km² before and 3.5 x 5.5 km² after
118 06 August 2019.”

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

122 We appreciate this suggestion. We have added a section to the supplement titled “S1 Sensitivity
123 of Urban Background NO₂ VCDs”, where we (1) describe the method used to quantify background
124 concentrations, (2) provide results of a sensitivity analysis evaluating the impact of different

125 percentiles on the general conclusions we've drawn within this manuscript, (3) an evaluation of
126 trends in urban background VCDs over time and (4) the impact and evaluation of background for
127 adjacent cities / clusters of cities. In short, following this thorough analysis, we have updated the
128 choice of background concentration to the 50th percentile. We test using different percentile
129 thresholds as the background concentration and generally find that changing the used percentile
130 does not meaningfully impact our results nor change the directionality of the trends. Please refer
131 to the revised methods section (Sec. 2.4) and the supplement document for more information.

132 Page 6, Line 157: The claimed acceleration in the decreasing trend requires statistical validation;
133 otherwise, such descriptions might be just removed. (Also, for the descriptions on Page 10, Line
134 239, Page 10, Line 221, and Page 14, Line 310)

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

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

141 We have added the description of the mining regions within the captions of each figure (Figures
142 6-11 in the revised manuscript)

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

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

146 Page 8, Figure 3: The information of NO₂ VCD uncertainty and significance tests on the
147 regression is missing. In addition, please ensure consistency of significant figures or decimal
148 precision for all numerical data throughout the manuscript.

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

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

155 Thank you for this suggestion. We have since removed this text for the sake of shortening the
156 manuscript. However, we calculated that 66 of the 71 urban clusters in Eastern Russia (or 93%)
157 exhibited larger 2024 NO₂ VCDs than in 2019.

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

160 We have double checked the manuscript to ensure that the units (e.g. 10¹⁵ Molecules cm⁻²) are
161 used consistently throughout the manuscript. If this is in reference to the range of plotted data on

162 the colorbars (e.g. -1 to 1 in Fig. 4 vs. -2 to 2 in Fig. 6), we find that different regions require
163 different values on the color bar for the sake of effective data visualization, as not all regions
164 experience a similar range of concentrations.

165 Page 10, Line 232: What is the term “largest” referring to or being compared against in this
166 description? (other cities or other land type? Also, for the descriptions on Page 11, Line 248, Page
167 11, Line 253-254, Page 12, Line 263-264 and Page 12, Line 270)

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

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

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

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

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

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

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

191 We have increased the size of the legend for readability.

192 Page 14, Line 311: Typo.

193 Thank you, we have fixed the typo.

194 Page 14, Line 311: The abnormally high NO₂ VCD values require further examination to exclude
195 artifacts, including applying data filters based on Level-2 QA flags. It should also be verified
196 whether any spurious outliers affect the averaging process.

197 The oversampled level 3 TROPOMI NO₂ product we use in this study are quality controlled, with
198 the recommended QA flag > 0.75 applied to the L2 data before oversampling. It removes cloud-
199 covered scenes (cloud radiance fraction > 0.5), partially snow/ice covered scenes, errors, and
200 problematic retrievals. The number of valid observations for the Moscow urban cluster during
201 March 2022, when a monthly average value of 59×10^{15} molec. cm⁻² was calculated, had more
202 observations (233) than the median March (159) in the six-year record. Despite the abnormally
203 high value, this appears to be a valid mean value for that month.

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

205 We have increased the size of the text for readability.

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

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

212 Page 18, Line 376: It is not immediately clear why population-weighted VCDs are preferred here
213 over direct NO₂ VCDs for me. Would directly showing NO₂ VCDs make major differences?

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

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

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

225 "Evaluating trends in NO_x emissions inventories in African cities, we find a mean difference of -
226 8.0% (EDARv8.1) and -6.7% (CEDs) between inventory NO_x emission trends and VCD_{ENH}
227 trends, indicating a potential underestimate in both emissions inventories in African cities for this
228 period."

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

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

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

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

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

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

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

245 Thank you for bringing up this point. The sharp increase observed in winter 2022 in Europe is
246 indeed due to increases in Russian cities. We have created a version of this figure with Russian
247 cities removed and added to the supplement as Figure S18, and added the following text to line
248 373 of the revised/tracked changes manuscript:

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

251 The noted difference in the magnitude for Europe in 2022 between previous Figure 13 and Figure
252 9 is due to the fewer data points during winter. When averaged over the colder months, the 2022
253 increase is stark, as seen in Figure 13. When averaging to an annual value, the higher number of
254 observations in the warmer months, when an increase was not observed, dilutes the impact of
255 the winter values.

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

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

260 “Additionally, while many of the trends presented here reflect changes in anthropogenic NO_x
261 emissions, it is important to recognize that atmospheric chemistry also influences the observed
262 NO₂ variability. Seasonal differences in photochemical lifetimes (i.e., longest in winter), boundary
263 layer mixing (i.e., more vertical mixing in summer), chemical partitioning between NO and NO₂
264 (i.e., the fraction of NO₂ is largest in winter) and meteorological variability can all modulate the
265 magnitude and timing of observed trends. These processes likely contribute to some of the
266 regional and seasonal differences highlighted in this study.”

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

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

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311 **RESPONSE TO REVIEWER 2:**

312

313 **General comments:**

314 This paper presents a comprehensive assessment of urban NO₂ changes worldwide from 2019
315 to 2024 using TROPOMI NO₂ VCD observations. Differences in NO₂ VCD changes over
316 populous cities and broader areas are disclosed, probably driven by anthropogenically induced
317 factors such as urbanization, industrial activities, government interventions, and societal
318 disruptions. The paper also attempts to quantify the influence of background NO₂ and
319 NO₂ seasonal variability on the trend analysis. The research topic fits in the scope of ACP, and
320 the manuscript is already in good shape. I recommend its publication after the authors address
321 the following comments.

322 *We thank the reviewer for this thoughtful review.*

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

326 **Specific comments:**

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

330 *We appreciate this suggestion. We have changed the title of the manuscript to "Global NO₂*
331 *Trends from TROPOMI (2019–2024): Urban Changes and Emerging Hotspots".*

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

335 *Thank you for this suggestion. We have modified the text on line 44 of the revised manuscript to*
336 *reflect the different types of remote sensing methods that exist for NO₂:*

337 *"NO₂ can also be remotely-sensed from ground-based instruments capable of inferring vertical*
338 *profiles of NO₂, such as using multi-axis differential optical absorption spectroscopy, MAX-*
339 *DOAS (Vlemmix et al., 2010)"*

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

343 *Thank you for this suggestion. We have added the following text to the methods section (Line*
344 *102) to highlight the value of GHS-SMOD:*

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

350 Section 2.2: Please briefly describe the uncertainty of the NO₂ VCD product used here.

351 Thank you for this suggestion. To address general uncertainty within TROPOMI retrievals, we
352 added text to note the 15-20% relative uncertainty related to monthly and annual averages, as
353 highlighted in a recent previous study (Glissenaar et al., 2025). We also added text to mention
354 the systemic biases of TROPOMI NO₂ retrievals, that can lead to underestimated NO₂ VCDs
355 over highly polluted regions, and overestimated NO₂ VCDs in less-polluted regions (Lambert et
356 al., 2025). To address uncertainty associated with estimates at the city and country level, we
357 have added error bars to relevant figures and include uncertainty estimates when reporting
358 relevant values.

359 We have added text to line 129 of the revised manuscript, highlighting the most common areas
360 of uncertainty related to NO₂ retrievals.

361 “TROPOMI NO₂ retrievals are subject to measurement and retrieval uncertainties that
362 propagate into the oversampled Level 3 products. Typical uncertainties in monthly or annually
363 averaged tropospheric NO₂ vertical column densities are on the order of 15–20 %. Systematic
364 biases have also been reported, with overestimation in less polluted regions (+26.5% bias) and
365 underestimation in areas with high NO₂ concentrations (-31.4% bias), reflecting limitations in the
366 retrieval process (Glissenaar et al., 2025; Lambert et al., 2025).”

367 Additionally, we have now added statistical significance testing to all trend plots. Significance is
368 determined through linear regression on monthly de-seasonalized TROPOMI time series (see
369 revised Sec. 2.3). We successfully show that statistically significant trends can be separated
370 from insignificant trends from the monthly TROPOMI data from 2019 to 2024, (e.g. in Figures 2-
371 5).

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

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

376

377 Line 141-142: Please justify the definition of the background NO₂ concentration here, and
378 provide the sensitivity of the results in Section 6 to the choice of the percentile.

379 Thank you for this suggestion. We have added a section to the supplementary document
380 (Section S1 Sensitivity of Urban Background NO₂ VCDs) in which we conduct a sensitivity test
381 to evaluate the impact of using different percentile thresholds on the results related to the VCD
382 enhancement (previously Figure 12, which has now been split up into multiple figures). In that
383 supplementary section, we test using different percentile thresholds as the background
384 concentration and find that changing the used percentile does not meaningfully impact our
385 results nor change the directionality of the trends. Following this analysis, we find that the 50th
386 percentile as the threshold is an adequate choice for most cities (See Supplementary
387 information). In that same section we highlight test case of large adjacent cities and how the
388 background concentrations for those cities vary. Although more sophisticated methods of
389 background quantification exist (Fioletov et al., 2025), we find that using a percentile as an
390 assumed background concentration is an acceptable choice given the large number of cities
391 being evaluated (N >11,000).

392

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

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

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

400

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

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

407

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

410 Thank you for this suggestion. For each of these monthly level examples, we have now added
411 statistical significance testing to all trend plots. Significance is determined through linear
412 regression on monthly de-seasonalized TROPOMI time series (see revised Sec. 2.3). We
413 successfully show that statistically significant trends can be separated from insignificant trends
414 from the monthly TROPOMI data from 2019 to 2024, (e.g. in Figures 2-5).

415

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

419 We have removed this sentence from the revised manuscript.

420

421 Line 310: It is difficult to see that the increasing trend in Moscow accelerated in early 2022 from
422 Figure S9, except that NO₂ VCDs in winter time jumped to a higher level. Please provide a
423 quantitative discussion to demonstrate the acceleration.

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

426 “This increase was accompanied by anomalously high monthly mean concentrations in early
427 2022 (Fig. S11), following the onset of the Russia-Ukraine war in Ukraine, when monthly mean
428 NO₂ VCDs for March reached 59×10^{15} molecules cm⁻² (see Sec. 3.3)”

429

430 Section 6: please provide a summary of this section, i.e., to what extent the influence of
431 background NO₂ and seasonal variability can be on the analysis of urban NO₂ trends presented
432 above?

433 Thank you for this suggestion. We have added a section to the supplementary document
434 (Section S1 Sensitivity of Urban Background NO₂ VCDs) in which we conduct a sensitivity test
435 to evaluate the impact of using different percentile thresholds on the results related to the VCD
436 enhancement (previously Figure 12, which has now been split up into multiple figures). In that
437 supplementary section, we test using different percentile thresholds as the background
438 concentration and find that changing the used percentile does not meaningfully impact our
439 results nor change the directionality of the trends. Following this analysis, we find that the 50th
440 percentile as the threshold is an adequate choice for most cities (See Supplementary
441 information). In that same section we highlight test case of large adjacent cities and how the
442 background concentrations for those cities vary. Although more sophisticated methods of
443 background quantification exist (Fioletov et al., 2025), we find that using a percentile as an
444 assumed background concentration is an acceptable choice given the large number of cities
445 being evaluated (N >11,000).

446

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

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

456

457 **Technical comments:**

458 Line 50: Please check through the manuscript and replace the “x” with a times symbol at
459 corresponding places.

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

461 Line 57: “SCHIAMACY” should be “SCIAMACHY”.

462 We have fixed the error (now line 49).

463 Line 63: The statement “NO₂ concentrations increased through roughly 2005” is a little
464 confusing. I would suggest to rephrase this sentence to make it clearer.

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

466 “In the United States, NO₂ concentrations generally exhibited a decreasing trend from 2005
467 through the mid-2010s...”.

468 Line 94: It is better to be $1 \times 1 \text{ km}^2$.

469 We have made the change (now line 100).

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

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

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

473 Thank you, we have made that change.

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

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

476 Line 183: “select” to “selected”.

477 We have made the change to the Figure 3 caption.

478 Line 306: There is no Figure 7d.

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

481

482

483

484

485 References

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494 G., Lambert, J.-C., Van Roozendaal, M., and Eskes, H.: TROPOMI Level 3 tropospheric NO
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500 2010, 2010.

501

502 **RESPONSE TO REVIEWER 3:**

503

504 Huber and co-workers present a global scale analysis of NO₂ trends over large cities based on
505 satellite TROPOMI column data. The study covers 6 years of observations, from 2019 to 2024,
506 and includes the NO₂ sudden drop and subsequent rebound in many cities due to the COVID-19
507 disruptions, as well as air pollution changes in response to policy regulations, economic growth,
508 and armed conflicts. Based on population-weighted annual columns, cities of Asia, Europe and
509 North America show a NO₂ decline, whereas African cities show a moderate increase. Although
510 not strictly comparable, the observed trends and the anthropogenic NO_x emission trends from
511 the EDGARv8.1 bottom-up inventory give an indication that emission trends are underestimated
512 in inventories, in particular in the Southern Hemisphere. The manuscript is well written but too
513 lengthy, and the figures seem to support the drawn conclusions.

514 Thank you for this synopsis, and for taking the time to review our manuscript. We have made
515 substantial changes to both the content and structure of the revised manuscript, and we feel that
516 these changes greatly improve the clarity and quality of our work.

517 Although I appreciate the importance of the topic, there are several important issues with the
518 paper. First, the trend analysis lacks an estimate of the statistical significance. I have serious
519 doubts that the calculated trends in most cities are different from zero given the uncertainty in the
520 satellite measurement and the limited number of available observations per city. The findings are
521 therefore not sound. It is crucial to include a robust uncertainty analysis, and I argue that part of
522 the conclusions will considerably change.

523 Second, a major limitation is that the period (6 years) of the analysis is short for attributing the
524 observed trends to anthropogenic changes only, as short-term meteorological variability (not
525 accounted here) might partly explain the observed trends. Furthermore, the fact that the study
526 period includes the COVID-19 lockdowns makes the derivation of trends even more uncertain. I
527 recommend extending the analysis to a longer period, by combining OMI and TROPOMI datasets,
528 both having the same overpass (e.g. Glissenaar et al. <https://doi.org/10.5194/essd-2024-616>).

529 Third, the seasonal variation of NO₂ data is not adequately discussed. It is unclear whether a
530 threshold of available observations per month was used and why the May-to-September trends
531 are not compatible with the November-to-March trends in some regions, e.g. Europe. Overall, the
532 discussion is poor, and the scientific content is limited. To my opinion, the main problem of the
533 manuscript is the lack of innovation and inadequate analysis. Therefore, I cannot accept this
534 manuscript for publication in ACP.

535

536 Thank you for providing this thoughtful feedback. Below, we list the ways in which the above
537 concerns have been addressed. We are confident that the value and quality added to the
538 manuscript in including these substantial changes will be apparent.

- 539 1. In this revision, we have performed an uncertainty analysis, being sure to provide
540 statistical significance where relevant. We have made sure to note clearly if a trend is or

541 is not statistically significant. After performing this analysis, our original findings have been
542 strengthened.

543 2. We do not claim that the observed changes are only anthropogenic in nature; we are
544 simply reporting observed concentrations. Previous work has shown that meteorological
545 variability has lesser influence when aggregating to the country or continental level as
546 opposed to at the local level (Petetin et al., 2020), and our presented evaluation of NO₂
547 VCD urban enhancements against anthropogenic emissions inventories is only presented
548 at the urban continental level. Despite this, even if hypothetical changes in NO₂ over a six-
549 year period were to be driven by meteorological as opposed to anthropogenic factors, we
550 still find that such a trend would be worth reporting. We have added the following text to
551 line 510 in the conclusions of the revised manuscript to acknowledge these limitations:

552 “Additionally, while many of the trends presented here reflect changes in anthropogenic
553 NO_x emissions, it is important to recognize that atmospheric chemistry also influences the
554 observed NO₂ variability. Seasonal differences in photochemical lifetimes (i.e., longest in
555 winter), boundary layer mixing (i.e., more vertical mixing in summer), chemical partitioning
556 between NO and NO₂ (i.e., the fraction of NO₂ is largest in winter) and meteorological
557 variability can all modulate the magnitude and timing of observed trends. These processes
558 likely contribute to some of the regional and seasonal differences highlighted in this study”.

559 3. We appreciate the suggestion to include a longer time series by including an evaluation
560 of OMI trends, however, there are instrument (i.e., coarser resolution) and algorithm
561 differences that make a stitching of these two datasets complex. The inherent advantage
562 of this work is that we are using a single instrument with a consistent algorithm throughout,
563 to calculate the trends. There has been prior work documenting NO₂ trends between 2005
564 – 2019 using OMI and it would be repetitive to repeat that work. We instead cite the OMI
565 NO₂ trends papers to give the reader references to refer to.
566

567 4. The novelty in this paper is the timeframe and global expansiveness (11,000 cities) of the
568 analysis that we are conducting. To our knowledge, no published paper reports satellite-
569 based NO₂ trends through the year 2024. We are documenting urban and country-level
570 NO₂ trends that, before this paper, have not been documented in the literature. For
571 example, it has not been documented that Seoul has the largest NO₂ drop between 2019
572 and 2024 and that Tehran has the largest NO₂ burden in 2024; this scratches the surface
573 of newly documented NO₂ trends that we report herein. This is an important scientific
574 advancement because we are documenting which urban areas appear to be implementing
575 effective control strategies and which are not. This has implications for future control
576 strategies.
577

578 5. Thank you for the questions regarding seasonality. We have expanded our discussion in
579 the revised manuscript to more clearly discuss those results. The noted difference
580 between May-September and November-March in Europe is driven by Western Russian
581 cities, which caused the initial spike in Winter 2022. We have added the same figure to
582 the supplement with Russian cities removed (Fig. S17), to show that seasonal trends
583 match more closely when excluding Russian cities.

584 Specific comments:

585

586 - L.112-116: Any data filtering used?

587 Thank you for allowing us to clarify. We are using a level 3 data product (Goldberg, 2024), which
588 applies a filter to remove all pixels with a qa_value filter < 0.75. We have added the following text
589 to line 122 of the revised manuscript to make this clear:

590 "Data were quality controlled to remove Level 2 pixels with a qa_value < 0.75 before
591 oversampling, which removes data with quality issues related to clouds, surface reflectivity (e.g.
592 snow and ice) or other retrieval errors".

593

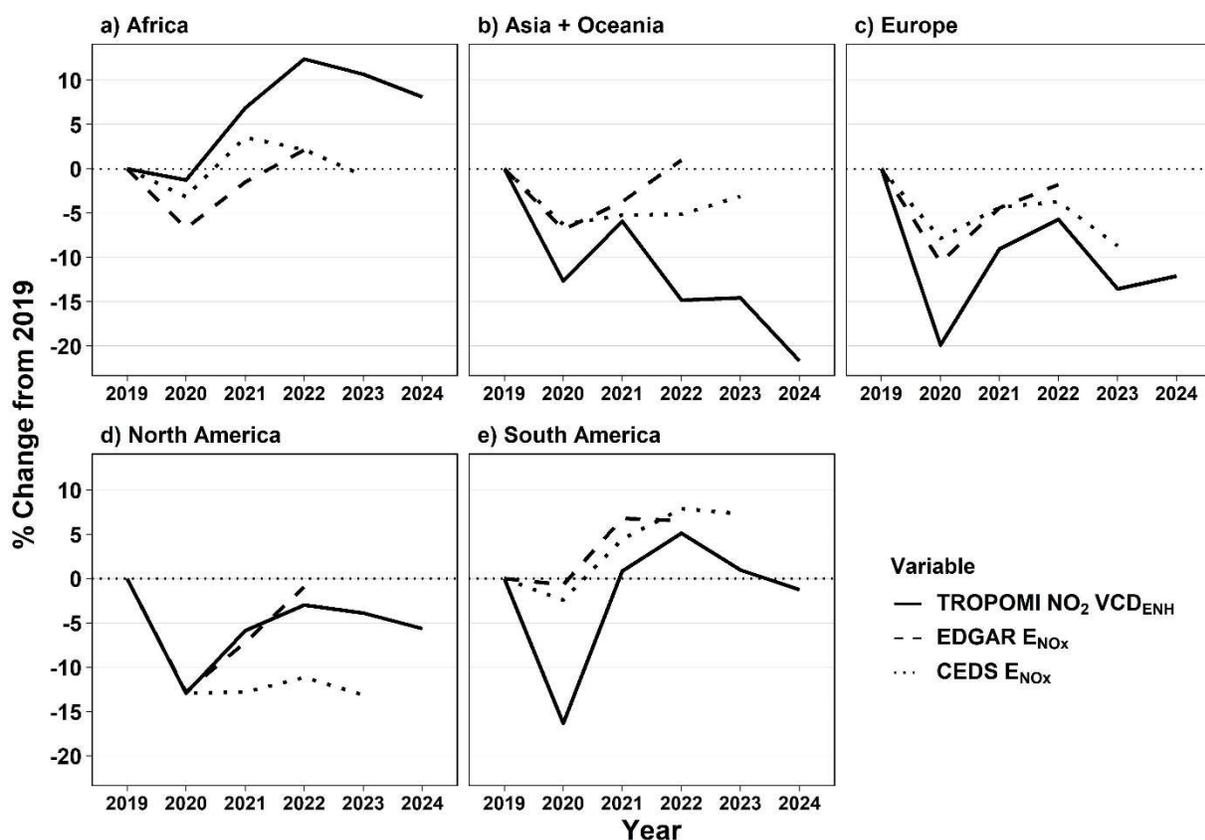
594 - L. 57: Read SCIAMACHY

595 Thank you for pointing this out. We have corrected the error.

596

597 - Why not using CEDS (instead of EDGAR)? The latest CEDS version covers 2023.

598 Thank you for this suggestion. We have now expanded the analysis to include an evaluation of
599 EDGAR as well as CEDS NOx emissions. Results can be found in Section 5 of the revised
600 manuscript.



601

602

603 - A table summarizing the percentage changes would be useful

604 We appreciate this suggestion, however given the large number of cities (11,000+), we find
605 including a table of all percent changes would not be realistic.

606

607 - Figs 2, 4, 5, 6 are similar and could be grouped in 2

608 Based on this suggestion, we re-grouped some of the figures from the original submission to have
609 one main figure per continent, which more effectively groups results based on geographical
610 location.

611

612 - Unclear whether the strong column changes in Quito and Santos are due to changes in
613 anthropogenic activities, or to data issues.

614 The level 2 TROPOMI data used to produce the level 3 product used in this paper (Goldberg,
615 2024) are quality controlled to remove all pixels with a QA_flag value < 0.75, which remove pixels
616 with data issues. There is no evidence for the changes in these cities being attributed to “data
617 issues”. We do not claim that these changes are a direct result of anthropogenic activities; we are
618 simply highlighting cities that exhibited notable changes in NO₂ VCD.

619

620 - For which among the cities of Fig.7 are the trends significant?

621 Thank you for this question. For the labeled cities in each panel, we have noted which represent
622 a statistically significant trend with an asterisk. These results are now spread across Figures 6-
623 11.

624

625

626

627

628 References

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634 Pando, C.: Meteorology-normalized impact of the COVID-19 lockdown upon NO₂ pollution in
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