

## Reviewer #2

This is a very well-done manuscript investigating NO<sub>x</sub> emissions and background NO<sub>2</sub> globally. Overall, this was an easy paper to review, so I appreciate the authors producing a well-written manuscript.

We would like to thank the reviewer for a very thorough and detailed review.

“My largest concern is regarding a mixing of two TROPOMI NO<sub>2</sub> algorithms v2.3.1 and v.2.4. I realize this does affect any major conclusions of the manuscript, but the current phrasing that “the impact of the change of the version should be smaller than 5-10% step change” is a hypothesis. A change to the surface reflectivity and a priori profiles may not have a large net effect globally, but for some individual cities, this change may be very large, exceeding a 10% difference. An easy, but also appropriate way to deal with this is simply to remove Fall 2022 – Fall 2023 data from the Section 6 analysis. I don’t think the paper would be any meaningfully different by doing this. I think it’s appropriate to be including the v2.4 2023 data in multi-year averages such as Sections 4 & 5 since those are not investigating interannual trends. If you would like to still include Fall 2022 – Fall 2023 data in Section 6, then I think it’s appropriate for you to do a case study... In Page 13 Line 16, you mention that Spring 2023 Europe-2 urban NO<sub>2</sub> is lower than Spring 2020. This could be a good test case... see if this still holds true if v.2.4 is used for Spring 2020.

The revised manuscript is based on versions v2.4 and v2.5 for the entire period. Old version v2.3 was not used anymore. However, the results are nearly the same as in the original version of the manuscript..

### Minor suggestions:

Page 3 Line 1 and throughout. Change NO<sub>2</sub> emissions to NO<sub>x</sub> emissions. If necessary (such as on figure captions), rephrase to “NO<sub>x</sub> emissions reported at NO<sub>2</sub>”.

We changed “NO<sub>2</sub> emissions” to “NO<sub>x</sub> emissions” where appropriate. However, we need to distinguish between emission estimates based on NO<sub>2</sub> data (e.g., Fig. 3a, 4, 10) from estimates of NO<sub>x</sub> emissions (Fig. C1). We also added the clarification “NO<sub>x</sub> emissions reported at NO<sub>2</sub>” to the figure captions.

Page 3 (anywhere): It may be good to state explicitly that you are using a variation of an exponentially modified Gaussian function fit, as compared to a variation of the flux divergence method. With one or two sentences, it may be helpful for future readers for you to compare/contrast to the flux divergence methods used by Beirle et al. 2023

(<https://essd.copernicus.org/articles/15/3051/2023/>) and Lonsdale and Sun 2023 (<https://acp.copernicus.org/articles/23/8727/2023/>)? I realize your method is discussed in-depth in Section 3, but it could be good to give a quick preview/summary here too.

We added a sentence that “*Unlike other similar studies that studied plumes from emission point sources (e.g., Lange et al. (2022); Beirle et al. 2023), this study included the background component in the analysis and separated urban emissions from emissions from industrial point sources.*”

Page 8 Line 29. The NO<sub>2</sub> lifetime is also very strongly a function of ambient VOC and NO<sub>2</sub> (Figure 1 Laughner and Cohen), as well as other meteorological factors that are hard to capture such as the amount of vertical mixing / plume width / coastal dynamics. For example, NO<sub>2</sub> lifetime in the center of a plume/city is different than at the edges of a plume/city. Liu et al., 2024 gives a list of effective NO<sub>2</sub> lifetimes by city (<https://acp.copernicus.org/articles/24/3717/2024/acp-24-3717-2024-supplement.pdf>) and I don't see a strong dependence on latitude. As a result, I am wary of the Lange et al 2022 parametrization, but also realize it is a step in the right direction. In any case, more discussion of NO<sub>2</sub> lifetime uncertainties is needed here.

Yes, NO<sub>2</sub> lifetime depends on various factors that could be different from city to city. This is why we also show average results for large regions to establish a “benchmark”, so individual cities can be compared to their benchmarks to determine if NO<sub>2</sub> levels for a particular city are different from average conditions of the region.

We agree that lifetime in the center of a city could be different than at the edges of a city. However, our isolation of background NO<sub>2</sub> should reduce the difference in lifetimes within the plume between the center and the edges of a city.

Liu et al., 2024 analyzed US data from a relatively narrow latitudinal belt, from 25 to 48 N, that may not be enough to see a substantial latitudinal dependence. The absolute values in that study are between 1.8 and 6.8 hours, i.e. in range of Lange et al., (2022) results (their Figure 6). The lifetime estimates by Lonsdale and Sun (2023) also show the latitudinal dependence with lifetime for southern subregion been shorter than for northern subregions. Lonsdale and Sun (2023) also noted that their estimated lifetimes are much longer (from 10 to 100 hours, according to their Figure 1b) than the values from other studies for all regions except southern East Asia. The main problem is that the lifetime estimates based on statistical analysis of satellite data are large for various reasons, not to mention the variability of the lifetime itself.

We added a paragraph on lifetime uncertainty and its impact on emission estimates to section 4.3 and discussion section.

Page 9 Line 11. Can you provide a bit more detail about what the fitting results are actually showing? Essentially this is a statistical re-creation of the TROPOMI NO<sub>2</sub>? Is that a correct interpretation? Is it simply a summation of background+pop-density sources+industrial sources?

We added a sentence that *“The fitting results (Fig. 1, column b) is a sum of the background, population density-related and industrial source-related components (Fig. 1, columns d, e, f, respectively). The residuals (Fig. 1, column c) are the difference between the mean TROPOMI NO<sub>2</sub> and the fitting results.”* We also added Maps of elevation and population density (Fig. D1).

Figure 1. It might be good to annotate percentages on to the panels of these plots comparing the Spring/Summer/Fall values to the winter. For example, annotate that Spring NO<sub>2</sub> is 20% (or whatever the exact value) lower than Winter NO<sub>2</sub>. I think that would strengthen the argument that population density NO<sub>2</sub> changes more dramatically than background.

We added such estimates: *“The amplitude of this annual cycle is the smallest for the background component: average summer values are 63% and 76% of average winter values for Houston and Guangzhou, respectively. For the industrial component, these values are 42% and 39%. The seasonal changes in the urban component are the largest among all the three components (summer values are 20% and 32% of wintertime values Houston and Guangzhou, respectively), so they may be caused by a difference in emissions themselves.”*

Page 9 Line 21. I think it'd be OK to further hypothesize why NO<sub>x</sub> emissions are lower in summer... Evidence suggests vehicle NO<sub>x</sub> emissions are larger in colder weather than warmer weather and that you may be observing this:

<https://www.sciencedirect.com/science/article/abs/pii/S0048969720369333>

<https://www.sciencedirect.com/science/article/abs/pii/S0269749121016341>

Thank you for these references. We added such discussion.

Section 5 (anywhere). It's important to re-emphasize here that TROPOMI is taking an early afternoon snapshot. I imagine NO<sub>2</sub> drops in the morning on “rest days” are more dramatic. You could allude to TEMPO / GEMS as next steps to investigate this here or in the Discussion.

We added the following sentence to section 5: *“Please note that TROPOMI takes measurements the early afternoon, and the weekend effect estimates presented in this section may not reflect the difference between workdays and weekends for mornings or evenings.”*

Page 13 Line 16 (also page 14 Line 15). This could be driven by changing algorithm. I recommend doing a case study for this to see if the result holds when using v2.4 for Spring 2023. If so, feel free to keep this sentence. If not, I suggest removal of this sentence.

The revised manuscript is based on v2.4 and v2.5 for the entire period and the Europe-2 springtime 2023 values are still lower than the 2020 values.

Page 15. I was surprised TEMPO / GEMS / Sentinel-4 were not mentioned in the Discussion section. It's important to note that changes in the urban/industrial emissions & background in the morning and late afternoon could be different. This is applicable to all Results sections, but perhaps most applicable to the weekday/weekend section. What new knowledge do you expect TEMPO/GEMS/Sentinel-4 to bring in the context of this work?

This is a good point. We added some discussion about potential impacts of our results on TEMPO / GEMS / Sentinel-4 data analysis.