

Dear Editor,

The authors thank you and the reviewers for the efforts you and the reviewers put into improving this manuscript. The point-by-point response to your comments is listed below and the revisions/additions/edits are shown in the tracked-change file.

I'm please to accept your manuscript "Estimating the variability of NO_x emissions from Wuhan with TROPOMI NO₂ data during 2018 to 2023" after minor revisions. Two of the reviewers have provided lists with minor comments - please carefully address them in your revised manuscript. In addition, please check my comments below.

* Abstract: I suggest to reformulate the sentence "We estimate a summer-to-winter emission ratio of 0.77, which is overestimated to some extent, though it is even higher provided by the bottom-up inventories" to "We estimate a summer-to-winter emission ratio of 0.77, which may be overestimated to some extent, but is still lower than suggested by bottom-up inventories"

Response: Done.

* Figure 1: Wrong units in emissions in right figure

Response: Corrected.

* Figure 4. Please add label to y-axis even if the quantity shown is the normalised emission

Response: Added.

Review #1:

Review of the manuscript: "Estimating the variability of NO_x emissions from Wuhan with TROPOMI NO₂ data during 2018 to 2023" by Zhang et al.

I would recommend accepting the manuscript for publication after minor revisions as follows (the line numbers refer to the revised manuscript):

L28 rephrase as: "though it is even higher in the bottom-up inventories"

Response: This sentence has been rephrased as 'We estimate a summer-to-winter emission ratio of 0.77, which may be overestimated to some extent, but is still lower than suggested by the bottom-up inventories.'

L33 " , the estimation for Wuhan is ~4% for the emissions and ~8% for the chemical lifetime." What is the estimation here? Do you mean the underestimation compared to something? Not sure what these numbers represents... Please clarify.

Response: It should be ‘underestimation’. This sentence is rephrased as ‘...in Wuhan’s case, the underestimation is ~4% for the emissions and ~8% for the chemical lifetime’.

L146 “114.4°E, 30.6°N,” put this in parenthesis maybe?

Response: added.

L147- “Compared to Zhang et al. (2023), our study domain is limited to the urban area (within the Fourth Ring Road) of Wuhan. For one reason, most (~ 60%) of the NO_x emissions are concentrated in the urban area (Zhang et al., 2023); for another, we use regional mean wind fields and NO_x chemical loss rate, the larger study domain would induce large uncertainty in the result.” This sentence is a bit strangely formulated. Maybe try to rephrase it for example as follows: “Compared to Zhang et al. (2023), our study domain is limited to the urban area (within the Fourth Ring Road) of Wuhan, as most (~ 60%) of the NO_x emissions are concentrated in this area (Zhang et al., 2023). In addition, since we use regional mean wind fields and NO_x chemical loss rate, the larger study domain would induce larger uncertainty in the result.” Or something similar.

Response: This sentence is rephrased as suggested.

L151 “One demission of the grid map is along with and the other perpendicular to the wind direction.” Maybe rephrase as “One dimension of the grid map is along the wind direction, and the other is perpendicular to it.”

Response: Rephrased.

L153 “rotated grid map” should you say here as well resampled instead of rotated? Same in the title of Fig. 1b.

Response: The grid map is rotated, and the TROPOMI observation is resampled into the rotated grid map. We have corrected the title of Fig. 1b.

L229-231 “Overall we calculate a mean NO_x chemical lifetime of 2.82 h, close to the 2.46 h estimated by Zhang et al. (2023), and is around 5% lower than Lange et al. (2022) reported 2.94±0.3 h for the NO_x effective lifetime.” Rephrase maybe as follows: “Overall we obtain a mean NO_x chemical lifetime of 2.82 h, which is close to the 2.46 h estimated by Zhang et al. (2023), and around 5% lower than the value (2.94±0.3 h) reported by Lange et al. (2022) for the NO_x effective lifetime.”

Response: Rephrased.

L231 “The fitting result for cold months NO_x chemical lifetime is 4.25 h, and for most of the days, the estimated NO_x chemical lifetime is between 1.5 h and 6 h.” replace with “For the cold months the estimated NO_x chemical lifetime is 4.25 h, and for most of the days, the estimated NO_x chemical lifetime is between 1.5 h and 6 h.”

Also what do you mean with “most of the days”? How many?

Response: The sentence is rephrased as: ‘For the cold months, the estimated NO_x chemical lifetime is 4.25 h, and for ~70% of the days, the estimated NO_x chemical lifetime is between 1.5 h and 6h. For the warm months, ~65% of the chemical lifetime estimation is within the 0.8 – 2.5 h range, and the mean value is 1.62 h.’

L313 here and elsewhere, the word “valley” could be replaced with “sudden decrease/reduction/drop” or something similar.

Response: Corrected.

L337 we compute -> we obtain

Response: Corrected.

L398 we discard, We should be with first capital letter

Response: Corrected.

L402 “Our analysis of the temporal variability of the estimated NO_x chemical lifetime and emission is also of uncertainty, though this part of uncertainty is difficult to quantify.” This sentence does not read right, please check and rephrase.

Response: This sentence is changed to ‘Uncertainties also exist in the analysis of the temporal variability of the estimated NO_x chemical lifetimes and emissions.’

L404 ... for this is also found... you mean: “since this..”?

Response: Corrected.

L405 “Second, we have overestimated the summer-to-winter emission ratio...”
Overestimated compared to what?

Response: During the fitting procedure, we use a fixed NO₂-to-NO_x ratio and the bottom-up emissions to constrain the NO_x emissions, we may have obtained a dampened seasonality of NO_x emissions, thus a higher summer-to-winter emission ratio. Therefore, we say that ‘we have overestimated the summer-to-winter emission ratio’.

L422 align -> aligned

Response: Corrected.

L446 “dominated with fast winds.” -> dominated by fast winds

Response: Corrected.

Overall, I would recommend a review by an English mother-tongue, because I might not be able to catch imprecisions in English grammar.

Response: Thank you for the recommendation. A mother-tongue of English was invited to review the grammar.

Review #2:

Dear Zhang et al.,

Thank you for the updated version of the manuscript and the detailed answers to the review comments. Most of the suggestions from the reviewers are considered, which improves the quality of the manuscript and understanding of the study.

However, I still have some concerns about certain parts of the methodology and the resulting seasonality of the estimated emissions. The fixed NO_x/NO₂ ratio, the fixed bias correction factor, and constraining the fitting based on bottom-up emissions with a flat seasonality all lead to a dampened seasonality in the estimated emissions. These points are more discussed in the new version of the manuscript but must be kept in mind when analyzing variability.

The authors followed the reviewer's suggestions to remove the filter, which filtered days with estimated emissions beyond 0.5-1.5 times the ABACAS bottom-up emissions; another point in the method that dampened the seasonality. This results in a lower summer-to-winter emission ratio of 0.77 compared to 0.87 in the original manuscript.

The authors followed the suggestion to compare their estimated emissions to the monthly emissions of the EDGAR emission inventory. This provides new/more insights into the seasonal deviations between bottom-up and top-down emission estimates. With, in general, lower emissions from the top-down approach but much closer agreements in winter and larger discrepancies during the summer months.

In the revision, the term lifetime was changed to chemical lifetime. Can your estimated lifetimes represent the true chemical lifetime? Since it is usually influenced by downwind changes (influencing additional sources, plume meandering, wind), it is often called mixed or effective lifetime (e.g., Valin et al. 2015, Beirle et al. 2011, supplement).

Response: Thank you for the comments. The NO_x lifetime estimated through the superposition column model is derived from the NO_x chemical loss rate ($\tau = \frac{1}{k}$), and this is why it is referred to as the 'chemical lifetime'. The chemical lifetime is a part of the 'effective lifetime', which is composed of the chemical loss, the plume meandering, and other loss pathways.

Minor comments:

Line 99: Change "Compared to the previous versions v1.x, the version 2.3.1 includes a different treatment of the surface albedo to avoid negative and > 1 cloud fractions, and updates the FRESCO-wide cloud retrieval that leads to a lowering cloud pressure. " to "Compared to the previous versions v1.x, version 2.3.1 includes a different treatment of the surface albedo to avoid negative and > 1 cloud fractions, as well as an updated

FRESCO-wide cloud retrieval resulting in lower cloud pressures.”

Response: Changed.

Line 167: You don’t say what your initial guess of 2h and 4h lifetime for summer and winter is based on. Later, in line 226, you say it is based on Zhang et al. 2023 but don’t mention the details anymore. Please add information in Line 167 and maybe again in line 226.

Response: Added.

Line 178: I think the second part of the sentence “for too large a varying range is applied for it.” doesn’t make sense like this. For too large what? Instead of “a varying range is applied for it” maybe “a scale factor (fac) is applied to it”

Response: This sentence is rephrased as ‘The emission term is used in the cost function to reduce the dependence of fitted NO_x chemical lifetimes and emissions on the $\tau_{[NO_2]}$.’

Line 205: Change “, and they ...” to “. However, they ...”

Response: Done.

Line 218: I think comparing the monthly emission inventory data and the different deviations in summer and winter is interesting. Can you comment on possible reasons?

Response: When we use a top-down method to estimate NO_x emissions, we assume that the atmosphere is in a state of equilibrium: Emission – Loss = Atmospheric Content. In cold months, because of the slow loss rate, it is easy to reach equilibrium. However, in warm months, especially in summer, the fast loss of NO_x may make it difficult to achieve the equivalent state. Thus it might lead to underestimation of NO_x emissions of the top-down method in warm months.

Line 226: is determined instead of are determined

Response: Corrected.

Line 321: Delete significant. “A decrease (13.6%) is seen in 2023 compared to 2022.”

Response: Deleted.

Line 349: Replace “our” with “the”

Response: Done.

Line 359: “...the superposition column underestimate...”, the word model is missing here

Response: Added.

Line 405-410: Please add the point you mentioned in Sect. 3.2.2 here again; you are constraining your fit with emission inventory data, which shows a flat seasonality, probably leading to a dampened seasonality in your estimates.

Response: Added.

Line 429: Is the 2018-2019 comparison for the MEIC emission inventory? I think this information is missing here.

Response: The sentence is rephased as ‘TROPOMI estimation is lower than MEIC by less than 30% for 2018 – 2020. When compared with EDGAR v8.1, TROPOMI estimation is lower by ~20% for 2018 and 2019, but is 30~40% lower in 2020 – 2022.’