

Dear Reviewer,

We really appreciate your constructive comments, which helped a lot to improve the analysis and writing of the manuscript. The point-by-point response to your comments is listed below and the revisions/additions/edits are shown in the tracked-change file.

Review of the manuscript: “Estimating the variability of NO_x emissions from Wuhan with TROPOMI NO₂ data during 2018 to 2023”

General comments

The manuscript employs the superposition column model (previously published in literature) in combination with TROPOMI tropospheric NO₂ column data to estimate city-scale NO_x emissions and lifetimes and their variabilities. The paper is an extension of a previous work from the same (almost) authors covering a longer period, which allows for the study of the seasonal, weekly and interannual variability. Overall, the manuscript is well written, but in my opinion, there are some parts of the methodology that requires clarification. I suggest publications if the following issues are properly addressed:

Specific comments

Methodology (Sect 2.3): The explicit definition of lifetime and of the final emission E appears to be missing.

Response: Thank you for the comment. In this study, NO_x ‘lifetime’ is the ‘chemical lifetime’, and we have clarified this in the revised manuscript Sect. 2.4: ‘In Eq. (1), k (s⁻¹) represents the loss rate of NO₂ at the TROPOMI overpass time, and the relationship between k and NO₂ chemical lifetime $\tau_{[NO_2]}$ (h) is $k = \frac{1}{\tau_{[NO_2]} * 3600}$ ’, and the NO_x chemical life is determined by $\tau_{[NO_x]} = \tau_{[NO_2]} \cdot \frac{[NO_x]}{[NO_2]}$. We have also changed the term ‘lifetime’ to ‘chemical lifetime’ throughout the revised manuscript.

The definition of the final emission E is defined as ‘the total NO_x emissions E (in the unit of mol s⁻¹) from the study domain can be calculated with $E = \sum_{i=1}^{15} E_i \times L$ ’.

Please refer to Page 6-7, Line 163-164 and Line 180-183 in the revised manuscript.

L164 “The terms E_i , k , and α are fitted” what about the background coefficient b ?

Response: b is also fitted. The sentence has been rephrased to: ‘The terms E_i , k , α and b are fitted through a least squares minimization to the TROPOMI observed NO₂ line densities ($N_{TROPOMI}(x)$) and the a priori ABACAS NO_x emissions ($E_{ABACAS,i}$) to determine $N(x)$.’ Please refer to Page 6, Line 173-174 in the revised manuscript.

L164- Concerning the OH concentrations, if I understand properly, you use that information to constrain the fitted k coefficient. Is this needed to obtain a “good fit”? Is this worth running a full CTM? What would happen if you let the fit run free (or set

a reasonable fixed range), so that you would be not dependent on CTM outputs? What is the variability of the monthly OH? I suppose that if it changes a lot, it makes sense to have a dynamic initial guess, but could you discuss more your choices in this regard? I ask this because, you are making a case for data-driven emission estimation methods, but you still need model data to make your method work. This should be mentioned, I think.

Response: Thank you for the insightful comments. Yes, at first we needed the CTM output OH concentration to constrain the fitted k coefficient, and we ran full CTM for the monthly OH. The OH concentration changes a lot from month to month, the summer value can be several times higher than the winter value. We agree that this contradicts with the ‘CTM-independent method’ that we have claimed in the introduction. In the revision of the work, the CTM output OH concentration is no longer used in the fitting of the NO₂ line density to make this method free from the CTMs.

Instead, we use an initial guess of the NO₂ chemical lifetime of 4 h for the cold months (October to March) and 2 h for the warm months (April to September), the k (s⁻¹) coefficient is derived through $k = \frac{1}{\tau_{[NO_2]} * 3600}$. During the fitting procedure, we let k changes between 1/4 and 4 times of the initial value. We could not let the fit run free with k, in which case the final NO_x emissions and chemical lifetimes would be determined by the k value, and might result in a very long chemical lifetime in summer or very short chemical lifetime in winter. On the other hand, we could not set the k range too narrow on the condition that k may change significantly from day to day and month to month.

Since that we set a wide range for k, the emission term is used in the cost function to reduce the dependence of the fitted NO_x chemical lifetimes and emissions on k, and we have evaluated that the uncertainty of the a priori emission (35%) can result in 10% and 30% influence on the final estimated emission and chemical lifetime, respectively.

L171 “We restrict the emissions to a gaussian shape” It is not clear how you do that, could you clarify?

Response: Yes. We assume that the NO_x emissions intensities in the city are distributed as a Gaussian shape along with the wind direction: $E_i = amp * \exp[-(x_i - cen)^2 / wid]$, the parameter *cen* is initially set as the location of the city center, and we let it shift along the wind during the fitting procedure, *amp* and *wid* are also fitted to obtain E_i .

L171-172 “a scale factor is applied to the emission term. It is found to be ~0.1 for all the days that lead to the best fit of the NO₂ line densities.” It is not clear where this number comes from: what do you mean with “best fit”? Also, does this mean that you are minimizing the difference between your estimates and the inventory? This sounds

strange if you then evaluate your estimates against the same inventory. Can you clarify?

Response: The cost function is expressed as:
$$\text{func} = \left(\frac{N(x) - N_{TROPOMI}(x)}{N_{TROPOMI}(x)} \right)^2 + fac * \left(\frac{E_i - E_{ABACAS,i}}{E_{ABACAS,i}} \right)^2$$
, it is to minimize the difference between the fitted and TROPOMI NO₂

line densities and between the estimated emission and the inventory. Here we set a scale factor for the emission term, and the factor is limited to be between 0.1 and 0.2 to make sure that the cost function is dominated by the minimization of the difference between the fitted and TROPOMI NO₂ line densities.

The reason we add the emission term in the cost function is because we set a very wide range for the NO_x loss rate k by allowing it change between 1/4 and 4 times of its initial value. Without the constraint from the a priori emission, the final results of the model would be determined by the k value. On the other hand, we could not narrow down the varying range for k, for we have no more accurate information of it.

We agree with the reviewer that it is not appropriate to validate the estimated NO_x emissions with the ABACAS NO_x inventory that is used to constrain the fitting. In the revision, we use the EDGAR v8.1 monthly NO_x emission from 2018 to 2022 and MEIC v1.4 monthly NO_x emission from 2018 to 2020 to validate our estimation. Please refer to the revised Figure 2 and section 3.1 in Page 8 Line 207-222 in the revised manuscript.

L183 “We also exclude the days with estimated NO_x emissions beyond 0.5-1.5 times the ABACAS bottom-up emissions.” Why do you exactly do that? I read your reasoning concerning the uncertainty and the seasonal variability, but I think you could include also “bad” results as well or at least provide some statistics about them. How many of such days are there? What are the possible reasons for disagreements?

Response: Thank you for the constructive comments. The day to day, monthly and year to year variation and the uncertainty of the NO_x emissions were all within the ±50% range, so the estimated data beyond this range were excluded. The extremely high and low estimation is mainly caused by the bias of the satellite NO₂ observation. Overall we obtained 24 days higher than 1.5 times of the a priori emission and 15 days lower than 50% of the a priori emission, the total of them made up about 10% of the total number of the estimations.

Most of the high emission days are in winter and low emission days are in summer, so the filter of them dampened the seasonal variability. For this reason, in the revision of the work, we choose to keep these results for analysis.

L254 “Their much lower summer-to-winter emission ratio may be caused by much lower estimated summertime NO_x emissions or much higher winter emissions or both.” This sentence is maybe a bit self-evident. Are there any specific difference to

be mentioned here?

Response: Thank you for the comment. We have added discussions on the estimated summer-to-winter NO_x emission ratio in Page 11, Line 276-280 in the revised manuscript:

Two possible factors may contribute to the large difference in the summer-to-winter emission ratio between this study and Lange et al. (2022). First is the different treatment to the NO_x -to- NO_2 ratio. We use a fixed NO_x -to- NO_2 ratio of 1.26, while Lange et al. (2022) calculated the ratio from day to day, and it was lower in summer than in winter, leading to a lower NO_x emission estimation in summer. Second is that we use the bottom-up emission inventory to constrain our estimation, the flat seasonality of the bottom-up emissions leads to a higher summer-to-winter ratio of this study.

L271-273 “In this work, the a priori NO_x emissions are used to restrict the computation of NO_x emissions. Thereby, we have partly avoided the possible underestimation of NO_x emissions.” This is again what might be problematic. If you restrict the computation of the emission to the a priori inventory-based information, is it right to verify your estimates against those same emission inventory values? And, in general, if you need a good bottom-up inventory for your method to perform well, what is the added value of the satellite-based estimates? What would happen without that emission term in the cost function?

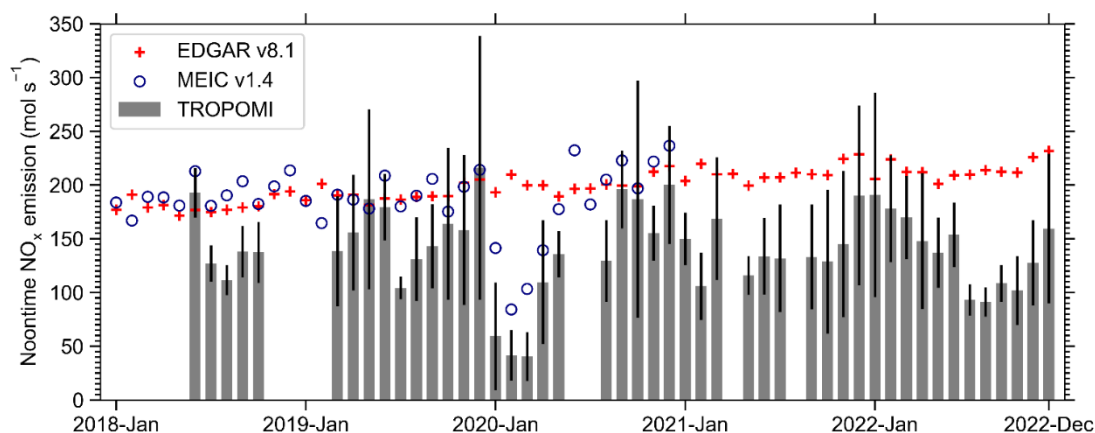
Response: We agree with the reviewer that it is not appropriate to validate the estimated NO_x emissions with the bottom-up emission inventory that is used to constrain the estimation, and in the revised manuscript, we have used the monthly EDGAR v8.1(2018-2022) and MEIC v1.4 (2018-2020) to validate the estimation, please refer to Figure 2, Page 8, Line 204-219 in the revised manuscript.

According to our cost function, the bottom-up emission plays only a small role in it since we give it a scale factor varying from 0.1 to 0.2 in the fitting, the final result is dominated by the minimization between the fitted and TROPOMI observed NO_2 line density.

Because of limited information on the NO_2 chemical loss rate, we gave a quite large changing range for it, without the constrain from the bottom-up emission inventory, the model will randomly settle down with a loss rate that lead to the minimum difference between fitted and TROPOMI observed NO_2 line density. The fitted chemical loss rate would be very high in winter or very low in summer, so we need the emission term in the cost function to keep the loss rate falls into a reasonable range. In the future, if we find a solution to narrow down the changing range of NO_2 chemical loss rate during our fitting, the emission term can be removed from the cost function.

L387 “the difference is only 4.7% compared to the ABACAS inventory.” Again, the satellite-based emissions are limited to remain close to the ABACAS inventory, so a smaller difference is expected.

Response: We agree with the reviewer that we should not validate our results with the ABACAS inventory, so we collected the monthly emissions of EDGAR v8.1 from 2018 to 2022 and MEIC v1.4 from 2018 to 2020 to validate the monthly NO_x emission of this work.



Overall, the TROPOMI estimation is close to the bottom-up emission inventories during cold months, while much lower during warm months. For the three years (2018 to 2020) when MEIC v1.4 data is available, the difference between TROPOMI and MEIC v1.4 is within 35%, and both of them capture the NO_x emission reduction in early 2020 due to COVID-19 lockdown. TROPOMI and EDGAR v8.1 are close to each other (within 30% difference) in 2018 and 2019, but the discrepancy is larger since 2020. EDGAR v8.1 is > 50% higher than TROPOMI from 2020 to 2022. Please refer to Figure 2 and Page 8, line 207-222 in the revised manuscript.

Conclusions: you could more thoroughly comment on the limitations of the method, such as the dependence on CTM data and on bottom-up emission inventory data.

Response: The reviewer's suggestion is well taken and we have thoroughly discussed the uncertainty and limitation of this study. Please refer to Sect. 4 in Page 15-16 and Line 436-446 in Page 17 in the revised manuscript.

Technical corrections

Abstract: TROPOMI should be defined

Response: Done.

L39 you should probably add a more general (maybe also older) references to this first statement.

Response: the references here are changed to:

Bassett, M. and Seinfeld, J. H.: Atmospheric equilibrium model of sulfate and nitrate aerosols, *Atmospheric Environment*, 17, 2237-2252, [https://doi.org/10.1016/0004-6981\(83\)90221-4](https://doi.org/10.1016/0004-6981(83)90221-4), 1983.

Penner, J. E., Atherton, C. S., Dignon, J., Chan, S. J., and Walton, J. J.: Tropospheric nitrogen: A three-dimensional study of sources, distributions, and deposition, *Journal of Geophysical Research*, 96, 959-990, <https://doi.org/10.1029/90JD02228>, 1991.

Jacob, D.: Introduction to Atmospheric Chemistry, Princeton Univ. Press, 1999.

L57 It should be noted that the superposition column model presented here is also dependent on CTM (via OH), so it does not solve the issue of running such complex models.

Response: Agree. In the revision of the work, we use an initial guess of the NO₂ chemical lifetime of 4 h for the cold months (October to March) and 2 h for the warm months (April to September), the k (s⁻¹) coefficient is derived through $k = \frac{1}{\tau_{[NO_2]} * 3600}$.

In this way we avoid the running of CTM and make this method computational efficient.

L58 Beirle et al. (2011) actually do not use plume rotation, but they separate the data in 8 classes based on wind direction and then fit the EMG function. Rotation and EMG together were used for example by Lu et al. (2015) among many others. <https://acp.copernicus.org/articles/15/10367/2015/>

Response: The reviewer's point is well taken and the sentence has been rephrased as 'Beirle et al. (2011) reduced the 2-dimensional NO₂ map surrounding a large point source (such as a megacity or a power plant, factory) to the 1D NO₂ line density by integrating the NO₂ column density across the wind direction.' Please refer to Page 2, Line 58-59 in the revised manuscript.

L60 Empirical Modified Gaussian model (EMG) -> this is actually Exponentially-Modified Gaussian model

Response: Corrected.

L62 applied (... -> this is not a complete reference list, add e.g. at the beginning of the references

Response: Added.

L91 10-15% there is tilde instead of a dash line here.

Response: We have changed this expression to '10%—15%'.

L144-145 "rotate the grid map toward the mean wind direction" I would avoid the word rotation here as plume rotation is often used to indicate another method (e.g. Fioletov et al. 2017). This is actually just a resampling to a grid aligned with the wind direction as you properly described in the caption of Fig. 1.

Response: The reviewer's comment is well taken and we have rephrased this sentence as 'We construct a 15 × 15 grid map centered at the city center with each grid size of 0.05 ° × 0.05 ° (6km × 6km) toward the mean wind direction. One demission of the grid map along with and the other perpendicular to the wind direction.' Please refer to

Page 5, Line 150-152 in the revised manuscript.

Fig. 1 panel a: in the title: original -> original

Response: Corrected.

L114-124 Does it mean that you only directly use GEOS-CHEM data for the initial value of [OH]? Maybe you could clarify this a bit.

Response: The GEOS-chem simulated OH concentration is no longer used in the work. Instead, we use an initial guess of the NO₂ chemical lifetime of 4 h for the cold months (October to March) and 2 h for the warm months (April to September) to

derive the k (s⁻¹) coefficient: $k = \frac{1}{\tau_{[NO_2]} * 3600}$.

L191-192 “There are least valid days in winter (December to February) after spring (March to May) for the cloudy and polluted conditions in winter.” not sure what you mean here, could be “There are least valid days in winter (December to February) due to the cloudy and polluted conditions.”

Response: Thank you for the comment, this sentence has been changed to: “There are least valid days in winter (December to February) due to the cloudy and polluted conditions.”.

L240 To verify this, it would be useful to check some traffic data in the city, if publicly available.

Response: The reviewer’s suggestion is well taken and we’ve found evidence from the ‘Annual Report on Wuhan Transportation Development (2023)’ (https://jtj.wuhan.gov.cn/znjt/zxdt/202409/t20240904_2450210.shtml, last access: 25 November 2024, in Chinese) that the traffic flow passed through the Outer Ring Road and the Fourth Ring Road of Wuhan was highest on Friday and lowest on Tuesday and Sunday, but the difference is only less than 2%.

This can confirm our finding that there’s no significant weekday/weekend difference in NO_x emissions from Wuhan. Please refer to Page 10, Line 252-255 in the revised manuscript.

L242 Add references here.

Response: Added.

L299 “under 2022” you mean as compared to or lower than 2022?

Response: Yes, it should be ‘compared to 2022’ and it is corrected.

L344 “It has a small influence (less than 1% in Wuhan’s case) on the overall estimation of city NO_x emissions, for the days with fast wind make up only less than

10% of the total number of days.” The grammar here is a bit off, please rephrase.

Response: Thank you for the comment. This sentence is removed in the revised manuscript.

L415 “The Wind fields” the world wind should not start with capital letter.

Response: Corrected.

Reference:

Beirle, S., Boersma, K. F., Platt, U., Lawrence, M. G., and Wagner, T.: Megacity emissions and lifetimes of nitrogen oxides probed from space, *Science*, 333, 1737-1739, [10.1126/science.1207824](https://doi.org/10.1126/science.1207824), 2011.

Lange, K., Richter, A., and Burrows, J. P.: Variability of nitrogen oxide emission fluxes and lifetimes estimated from Sentinel-5P TROPOMI observations, *Atmospheric Chemistry and Physics*, 22, 2745-2767, [doi: 10.5194/acp-22-2745-2022](https://doi.org/10.5194/acp-22-2745-2022), 2022.