

Author comments on the manuscript “TANGO CO₂ and NO₂ Observations: Synergistic Usage to Improve Emission Quantification and Characterize Atmospheric Chemistry”, Reviewer 1

We thank the reviewer for the careful and constructive reading of the manuscript. The comments have helped us to improve the presentation and physical justification of the model. In this document we provide our replies to all comments. The original comments are typeset in italic and bold face font, followed by our reply. Line numbers refer to the original version of the manuscript unless stated otherwise.

1 Major Comment

1. *My main concern of the manuscript is related to the model presented in Equation (1). As a reference, the paper by Meier et al. (2024) is given. This is indeed the same equation used in that paper; however, Meier et al. (2024) use this model to analyze the NO₂-to-NO_x ratio and not the CO₂-to-NO₂ ratio. In the context of Meier et al. (2024), I think it is clearly justified. The idea is that, after the initial period, NO and NO₂ go to a steady state where the ratio is constant (or m_0). See Figure 1b in that paper. Figure 1a shows how NO, NO₂, and NO_x behave. Note that they analyze this model to about 100 km from the emission source.*

The manuscript here, however, uses the same model to analyze the CO₂-to-NO₂ ratio at distances of less than 16 km from the emission source. At first glance, I thought the equation was written the other way around. For example, NO_x-to-CO₂ would behave in this way, with NO_x decaying exponentially and CO₂ being a constant tracer.

After seeing Figure 8 by Krol et al. (2024), I sort of understand why this model works near the emission source. However, it certainly cannot work far away from the emission source, as NO_x decays (and hence eventually also NO₂). That is also clear from Figure 8 in Krol et al. (2024). I would assume the model works until a steady state is reached, but I do not directly see any theoretical justification for it.

Equation (1) may be a useful empirical fit for the simulated near-field plume, where NO is still being converted to NO₂, but the manuscript does not provide sufficient physical justification for applying this form more generally to CO₂/NO₂. In particular, the interpretation of the asymptotic term m_0 is unclear: approximate NO_x partitioning equilibrium does not by itself imply a constant CO₂/NO₂ ratio farther downwind, especially as ongoing NO_x loss increasingly affects the NO₂ plume farther downwind. The authors should better motivate the validity range of Equation (1), clarify that it is an empirical approximation, and discuss where and why it may break down.

All in all, I think the authors should motivate the use of this model, discuss its validity, and potentially provide a theoretical justification. Also, how does the decay of NO_x affect the interpretation of the parameters m_0 , m_1 , and τ of the selected model?

adjusted

The key physical argument is as follows. Close to the source, NO_x/CO₂ is approximately constant since both are conserved tracers on short timescales. The CO₂/NO₂ ratio therefore mirrors the NO_x/NO₂ ratio analyzed by Meier et al. (2024), and the same functional form applies. The model breaks down farther downwind once NO_x loss through secondary chemistry becomes significant. For a typical wind speed of $\sim 5 \text{ m s}^{-1}$, a distance of $\sim 10 \text{ km}$ corresponds to a transport time of $\sim 2000 \text{ s}$, comparable to the NO₂ chemical lifetime under daytime OH concentrations. The model is therefore valid within the $\sim 16 \text{ km}$ analysis range used here. We have revised the manuscript to clarify this explicitly. We added the following text at (Page 6, Line 170) where the ratio model is defined:

“The functional form of Equation (1) is physically motivated as follows. Freshly emitted NO_x is dominated by NO, so the NO₂ concentration is initially suppressed relative to its photostationary-state value. As NO is oxidized to NO₂, the CO₂/NO₂ ratio decreases from an initially elevated value toward a near-field asymptote m_0 . The exponential term in Equation (1) describes this approach to photostationary-state partitioning, while CO₂ acts as a conserved tracer on these scales. We emphasize that this is an empirical near-field approximation whose validity rests on the assumption that NO_x loss through secondary chemistry—primarily NO₂ + OH → HNO₃—remains negligible within the analysis range. Any residual NO_x loss is implicitly absorbed into the fitted value of m_0 , which should therefore be interpreted as an observational near-field diagnostic rather than a true physical ratio. This assumption is supported

by the MicroHH simulation used in this study, which explicitly accounts for secondary chemistry losses. As shown in Figure 2c, the modeled CO_2/NO_2 ratio remains constant across the far field within the simulation domain, confirming that NO_x loss through secondary chemistry is negligible on the scales considered here. This is consistent with Fig. 8 of Krol et al. (2024) and validates the assumption within the mission requirements. We note that Meier et al. (2024) applied the same functional form to the NO_2/NO_x ratio, where the ratio rises from near zero at the source to a photostationary-state plateau. The advantage of the CO_2/NO_2 ratio is that the relevant chemistry parameters can be determined by a fit, whereas they must be known *a priori* when working with the NO_2/NO_x ratio.”

We further changed L394–395 from:

“Finally, we discuss the background ratio m_0 , which represents the CO_2/NO_2 ratio in the far downwind portion of the plume, where atmospheric processing has progressed to an approximate NO_x equilibrium.” to:

“Finally, we discuss the background ratio m_0 , which represents the empirical CO_2/NO_2 ratio in the far downwind portion of the plume within the near-field validity range of the model—i.e., where the $\text{NO} \rightarrow \text{NO}_2$ conversion is essentially complete but NO_x loss through secondary chemistry remains small.”

2 Technical and Minor Comments

1. **Title:** *I checked that AMT does not capitalize individual words in their titles. As a general suggestion, I would also recommend writing carbon dioxide and nitrogen dioxide instead of CO_2 and NO_2 .*

adjusted

We changed the title to sentence case following AMT style. We have retained the chemical formulas CO_2 and NO_2 in the title, as they are widely recognized abbreviations and their spelled-out forms would make the title unnecessarily long. The revised title reads:

“TANGO CO_2 and NO_2 observations: synergistic usage to improve emission quantification and characterize atmospheric chemistry”

2. **Abstract:** *With the same line of thought, CO_2 , NO_2 , CH_4 , and NO should also be defined in the abstract.*

adjusted

We have added the full names of CO_2 (carbon dioxide), NO_2 (nitrogen dioxide), CH_4 (methane), and NO (nitric oxide) on their first occurrence in the abstract.

3. **Abstract:** *In general, I believe the method names should be written in lower case. So, please change “Integrated Mass Enhancement (IME)” to “integrated mass enhancement (IME).” Note that in the manuscript, the method is defined quite many times.*

adjusted

We agree and changed it. The acronym IME is now defined only once, at its first occurrence in the abstract; duplicate definitions in Sections 4 and 5 have been removed. And we also use the acronym in the title of section 4.6.

4. **Abstract and many other places, especially in the Results:** *In the English language, there is no space before “%”.*

adjusted

We have removed all spaces before “%” throughout the manuscript.

5. **Abstract L3:** *“Here.” I think this is true in general, so you could maybe write “In general” instead of “Here.” This is also related to the discussion in line 37.*

adjusted

We changed the “Here” (Abstract L3, “Here, NO_2 plume structures exhibit. . .”) to “In general”.

6. *Abstract and in general: According to Table 1, the bias is always lower in the “CO₂ with NO₂ mask” method. Should it be reported?*

adjusted

We agree that reporting the bias characteristics in the abstract strengthens the message. We have added a brief statement to the abstract noting that the masking approach consistently yields lower absolute biases than the reconstruction approach, particularly for weak sources near the TANGO detection limit. In the conclusions this result is already discussed and does not need to be updated.

7. *Introduction: CO₂ and NO_x are defined, but please also define NO, NO₂, CH₄, CO, and O₃. You can then also remove the definitions from the beginning of Section 2. In Section 3.1, you might want to define CO and C₃H₆ (propane?).*

adjusted

We have added definitions of nitric oxide (NO), nitrogen dioxide (NO₂), methane (CH₄), carbon monoxide (CO), and ozone (O₃) at their first occurrence in the introduction. We note that the species-name definitions the reviewer refers to (“from the beginning of Section 2”) appear in the submitted manuscript at the start of Section 2 (TANGO mission), where carbon dioxide, methane, and nitrogen dioxide are expanded for CO₂, CH₄, and NO₂ respectively. Since we now define these in the introduction, we have removed the redundant expansions from the Section 2 opening. We have retained the units-convention sentence at the start of Section 4 (“Throughout this section, CO₂ and NO₂ denote retrieved column densities (mol m⁻²) ...”), as it serves a distinct technical purpose and is not a species-name definition. In Section 3.1 we have clarified that CO is carbon monoxide and that C₃H₆ is propene (not propane; propane is C₃H₈, whereas C₃H₆ is propene, an unsaturated hydrocarbon commonly used in atmospheric combustion simulations).

8. *Introduction: GOSAT-GW and EnMAP should be defined. Note that you do not mention CO₂M in the introduction but suddenly mention the mission in lines 520 and 523. Maybe a short mention could also be useful here?*

adjusted

We have added the full name of GOSAT-GW (Greenhouse gases Observing SATellite – for Greenhouse gases and Water cycle) at its first occurrence in the introduction. Regarding EnMAP: it is already defined in the abstract as “Environmental Mapping and Analysis Program (EnMAP)”; we have additionally expanded the acronym at its first occurrence in the introduction body. Regarding CO₂M: in the submitted manuscript it appears only in the Conclusions without prior introduction or definition. We have added a brief mention and definition of CO₂M (Copernicus CO₂ Monitoring mission) in the introduction, so that it is consistently introduced before use throughout the paper.

9. *L35: “representing the first global platform for co-located greenhouse gas and air pollutant monitoring.” I see what you mean here, but you might want to also highlight the resolution, as some could argue that SCIAMACHY was the first one.*

adjusted

We changed L35 from:

“representing the first global platform for co-located greenhouse gas and air pollutant monitoring”

to:

“representing the first global platform combining dedicated high-resolution greenhouse gas and air pollutant monitoring at spatial resolutions sufficient to resolve individual point source plumes”

10. *L37–45: Would it be more accurate to write “estimate NO_x emissions from NO₂ observations” instead of “convert NO₂ observations to NO_x”? Also, I just wanted to note that there are many more studies than are cited here.*

adjusted

We have adopted the suggested phrasing “estimate NO_x emissions from NO₂ observations” throughout the introduction, as it more accurately describes the inference step. We acknowledge that the citation list at L37–45 is non-exhaustive and change the sentence from “Several of the previous studies use NO₂ measurements ...” to “Several of the previous studies, among others, use NO₂ measurements ...”

11. *L59–60: I find that the approaches “first” and “second” go the other way around here.*

adjusted We changed:” The first approach uses NO₂ observations to define spatial masks for CO₂ integration, exploiting NO₂’s superior signal-to-noise ratio for plume detection. The second approach fits an exponential model to the CO₂/NO₂ ratio. Multiplying the model with observed NO₂ fields yields a CO₂ fields with reduced noise.” to

”The first approach fits an exponential model to the CO₂/NO₂ ratio. Multiplying the model with observed NO₂ fields yields a CO₂ field with reduced noise. The second approach uses NO₂ observations to define spatial masks for CO₂ integration, exploiting NO₂’s superior signal-to-noise ratio for plume detection. ”

12. *L64: According to Table 1, the bias is always lower for the “NO₂ plume masking” approach.*

adjusted

Table 1 confirms that the masking approach yields lower absolute biases than reconstruction at all emission strengths. The sentence at L64 previously read:

“We demonstrate that NO₂ plume masking provides superior emission estimates for CO₂ sources < 5 Mt yr⁻¹, while ratio reconstruction enables interpretable chemistry characterization for stronger emissions.”

The phrase “superior emission estimates” refers primarily to precision at low emissions, but does not capture that masking also has lower bias across all emission strengths. We have revised this to:

“We demonstrate that NO₂ plume masking yields both lower biases and comparable precision for CO₂ sources ≤ 12.5 Mt yr⁻¹, while ratio reconstruction uniquely enables interpretable chemistry characterization for emission approximately ≥ 5 Mt yr⁻¹.”

13. *L124: I noticed that you have used a special environment for CO₂ and NO_x/CO₂ here. Maybe some \chem should be used in general?*

adjusted

The inconsistent format is corrected.

14. *L152: “CO₂ and NO₂ denote retrieved column densities (mol m⁻²) after background subtraction.” I was wondering what the native units of the TANGO retrievals are. I was expecting to see ppm for CO₂. Anyways, in my studies, I have noted that it is very important to remove the CO₂ background in ppm; otherwise, you might run into problems later. Also, removing the background is a much more important step than is given credit here, and making small mistakes with the background definition can cause large errors in the final emission estimates. This is particularly the case with methods like IME that integrate over a large area.*

adjusted

We agree that background subtraction is a critical step and deserves more acknowledgment. The native TANGO retrievals will be expressed as total column mole fractions (in ppm for CO₂), which are converted to column densities (mol m⁻²) using the total air column density before background subtraction. We have added a clarifying sentence at L152 ”We note that in our simulation study the true background is known exactly from the LES, so background subtraction errors do not contribute to our uncertainty estimates; for real data errors in the background definition propagate directly into the integrated emission estimate.”

15. *L152: “The approaches require CO₂ and NO₂ observations on a common spatial grid.” If you were to use line densities, it would not be so strict.*

not adjusted

We agree that working with line densities would relax the requirement for a common spatial grid. However, for the TANGO instrument configuration considered here, the co-registration errors introduced by regridding are negligible compared to the retrieval noise (see Figure 6). We therefore retain the gridded formulation, as it simplifies the processing (e.g. common plume mask) without introducing a meaningful error. We note that a line-density formulation would still require interpolation to a common along-track coordinate, and also within the individual grids.

16. *L161: As the equation ends with “,”, I assume that this paragraph should start with “here” and not “Here.”*

adjusted

Corrected to lowercase “here”.

17. *L162: “Asymptotic background value.” This concept is quite difficult for me to comprehend, as NO_x (and eventually also NO_2) decays and CO_2 does not. I completely understand this concept for the NO_x/NO_2 ratio.*

adjusted

This concern is directly related to the major comment and is addressed there. The term “asymptotic background value” has been replaced by “empirical far-field value within the near-field validity range of the model” throughout Section 4.1 and the results discussion.

18. *Equation (6): Please use “argmin” instead of “min.” Also, as your covariance matrix S_y is diagonal, it might be clearer to write the sum-of-squares function as $\sum\{(y_i - F(s_i; \mathbf{x}))/\sigma_{y_i}\}^2$ instead of using matrices.*

partially adjusted

We have replaced `min` with `argmin` in Eq. (6), implemented via `\DeclareMathOperator*\argmin{\arg\,min}` added to the manuscript preamble, as `\argmin` is not defined by default in `amsmath`. We have retained the matrix notation, as it is consistent with the error propagation formalism used throughout Section 4 and is standard in the retrieval literature we build on (e.g., Hansen, 2010, Rodgers, 2000).

19. *Equation (11): Note that this is essentially plume masking, as can also be seen from Figure 2c,d.*

adjusted

We have added a sentence after Eq. (11) explicitly noting that the pixel selection criterion is equivalent to plume masking applied to the signal-to-noise ratio of each species, consistent with the visual impression from Figure 2c,d.

20. *Equation (14): This equation probably should end with “.”.*

adjusted

Added a full stop after Eq. (14).

21. *Section 4.5: I found this section quite short. Although this is not maybe the main part of the manuscript, you could connect your work to the masking done by Kuhlmann et al. (2019) and Varon et al. (2018). Both papers are already cited and use a “neighboring” approach.*

adjusted

We have expanded Section 4.5 to include a comparison with the neighboring-pixel masking approaches of Kuhlmann et al. (2019) and Varon et al. (2018), noting the conceptual similarity and the key difference that our threshold is applied to the NO_2 field rather than the target species itself.

22. *Section 4.6: Just to note that the IME method used by Santaren et al. (2025) is different from the one introduced by Varon et al. (2018), although they have the same name. Varon et al. (2018) do not have a similar distance concept as used here. They use $L = \sqrt{A}$ and LES simulations to calibrate the effective wind speed. For the version used by Santaren et al. (2025), and essentially this paper, you might want to check the derivation from Kuhlmann et al. (2024). Anyways, Equation (16), as presented, is not very intuitive, as it already mixes the theory with practical choices like pixel area. It might be more intuitive to write the integral directly and say that, fundamentally, emission = mass/lifetime, and then continue with $E = \text{mass} \times (U/L)$, and only after that explain how the integral is calculated by summing in practice.*

adjusted

We agree with this clarification on the IME lineage. We have added a note in Section 4.6 distinguishing the Varon et al. (2018) formulation (which uses $L = \sqrt{A}$ and LES-calibrated effective wind speed without an explicit distance concept) from the distance-based variant used here, which follows Kuhlmann et al.

(2024). Note that Kuhlmann et al. (2024) is a new reference not previously cited in the manuscript; we have added it to the bibliography. We have also restructured the presentation of Eq. (16) to first motivate the result from fundamentals—emission rate equals integrated plume mass divided by residence time, $E = M/t_{\text{trans}}$ with $t_{\text{trans}} = L/U$ —before introducing the discrete pixel sum. The citation for the distance-based formulation is updated accordingly from Varon et al. (2018) alone to Kuhlmann et al. (2024), Santaren et al. (2025).

23. **L261–262:** *The way I read this is that you do not use masking for the “reconstructed” approach. I think this is quite a dangerous approach, especially with real observations, and is prone to systematic errors. Why not use both approaches at the same time? I think it should be written clearly how the plume is selected.*

adjusted

We have clarified the text at L261–262. For the reconstruction approach, pixel selection is implicitly performed through the signal-to-noise filter in Eq. (11), which excludes pixels where either CO₂ or NO₂ falls below twice the respective noise level. This is functionally equivalent to a plume mask. We have made this explicit and noted that the same NO₂-derived mask used in the masking approach can optionally be applied to the reconstruction approach as well; for consistency in our Monte Carlo experiments we use the SNR filter throughout.

24. **L296:** *There is some extra space after “(.”.*

adjusted

Corrected.

25. **Figure 2:** *As your model is essentially 1D, would it make sense to also illustrate line densities?*

not adjusted

We acknowledge that line densities offer a compact 1D representation. However, as discussed in our response to comment 15, regridding errors are negligible for the TANGO configuration considered here, and the gridded 2D fields shown in Figure 2 more directly illustrate the spatial structure of the plume and the quality of the model fit across the full retrieval domain. We therefore retain the 2D representation, which we consider more informative in this context.

26. **Figure 2:** *I do not understand why you want to create a CO₂ reconstruction outside the essential plume area. As panel e is panel b times panel d, should you not also present your model for the full image?*

adjusted

We agree with the reviewer and changed Figure 2. Now the model and the reconstruction is reduced to the plume area.

27. **Figure 8:** *You could define the red line in Figure 8. I assume it comes from Equation (11). You could mark the source location with “x”. How do you make a CO₂ reconstruction upwind from the source?*

adjusted We added to the figure caption: “The red line indicates the plume mask used and the source location is indicated by the black x.”

28. **L385:** *I think it would be safe to note that this is not the CO₂/NO_x ratio. In the MicroHH setup, you assume that 95% of NO_x is emitted as NO.*

adjusted

We have added a parenthetical remark at L385 “This does not correspond to the true CO₂/NO_x emission ratio, since in the MicroHH setup 95% of the emitted NO_x is assumed to be released as NO rather than NO₂.”

29. *L510: You did not consider any errors related to wind speed. Errors in wind speed dominate the total error budget and can easily be more than 30% alone. In your IME method, the effect of wind speed is linear, so a 30% error in wind speed means a 30% error in the final emission estimate. You might want to also mention this.*

adjusted

We agree that wind speed uncertainty is a dominant error source in operational emission quantification and that its omission from our error budget was an oversight. We have added a paragraph at L510 noting that wind speed enters the IME estimate linearly through the transit time $t_{\text{trans}} = L/U$, so a 30% error in the effective wind speed directly translates to a 30% error in the inferred emission rate. Our study isolates the precision attributable to measurement noise and spatial/temporal co-location errors; wind speed uncertainty is an additional and often dominant contribution that must be accounted for in operational deployments. We cite representative references on wind speed uncertainty in satellite-based emission estimates to guide the reader.

3 Additional corrections identified during revision

During the revision process we conducted a systematic check of the manuscript beyond the issues raised by the reviewer. The following additional corrections have been applied.

1. **Subsection title capitalisation.** AMT uses sentence case for section and subsection titles. Several subsection titles in the Results section used title case and have been corrected:
 - “Emission Estimation Performance” → “Emission estimation performance”
 - “Method Comparison: Masking versus Reconstruction” → “Method comparison: masking versus reconstruction”
 - “Chemistry Parameter Retrieval” → “Chemistry parameter retrieval”
 - “Robustness to Spatial and Temporal Misalignment” → “Robustness to spatial and temporal misalignment”
 - “Application to EnMAP Observations” → “Application to EnMAP observations”

The subsection title “Emission estimation using the Integrated Mass Enhancement method” also contained the capitalized method name and has been changed to “IME method” consistent with the correction described in minor comment 3.

2. **Bare `\cite` command.** One citation in the Conclusions used the bare `\cite{Landgraf2025}` command rather than `\citep{Landgraf2025}`. This produced incorrect formatting in the natbib author-year style. Corrected to `\citep{Landgraf2025}`.
3. **Inconsistent spacing before units.** The manuscript uses `\,yr` (thin space) in 75 instances for Mt yr^{-1} but `\~yr` (non-breaking space) in 4 instances, all in the abstract and introduction. These 4 instances have been changed to `\,yr` for consistency.
4. **Inconsistent CO_2/NO_2 ratio direction in m_0 discussion.** In the paragraph discussing m_0 (Section 5.3), the manuscript alternately refers to the “ CO_2/NO_2 ratio” and the “ NO_2/CO_2 ratio” when both should consistently refer to CO_2/NO_2 , since m_0 is defined as the asymptotic value of $F(s; \mathbf{x}) = [\text{CO}_2]/[\text{NO}_2]$. Specifically, the sentence “it represents the NO_2/CO_2 ratio far downwind ... restoring the ambient NO_2/CO_2 ratio” has been corrected to CO_2/NO_2 throughout.
5. **Figure caption equation cross-reference error.** The caption of Figure 2 (method demonstration) references “Eq. 4” for the smoothed ratio from the model fit. However, Eq. (4) in the manuscript is the error propagation formula for the ratio uncertainty, not the ratio model itself. The correct reference is Eq. (1) (the ratio model $F(s; \mathbf{x})$). The caption has been corrected to read “Eq. (1)”.
6. **“ NO_2/CO_2 ” in ratio model description (Section 4.1).** In the state vector description, m_0 is described as “the background CO_2/NO_2 ratio downwind”. This is consistent with the model definition. However, in the m_0 discussion in the Results we verified and corrected inconsistencies as noted in item 4 above.

7. **Figure caption for EnMAP observations lacked site identification.** The caption of the EnMAP results figure (Figure 8 in the submitted manuscript) referred to the three observed industrial sites as “case 1”, “case 2”, and “case 3”, which provides no information to the reader without cross-referencing Table 2. We have replaced these anonymous labels with the actual site names and observation dates throughout the caption. The revised caption now introduces all three targets explicitly at the outset—Matla power plant, Mpumalanga, South Africa (5 October 2023; panels a, d, g, j), PP10 power plant near Riyadh, Saudi Arabia (11 July 2023; panels b, e, h, k), and PP9 power plant near Riyadh, Saudi Arabia (15 July 2023; panels c, f, i, l)—and uses these names in the emission estimates rather than case numbers. The phrase “particularly visible in case (i)” in the NO₂ row description has also been corrected to “particularly visible in panel (i)”.
8. **The EnMAP observations in Fig. 8 were plotted in a wrong orientation** This is corrected to agree with Borger et al. (2025).

References

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