

Reply to Referee # 2

Dear reviewer,

We sincerely thank you for your thorough review and constructive comments on our manuscript. We truly appreciate the time and effort you have invested in evaluating our work. Your insightful suggestions have been very helpful in improving the clarity, depth, and overall quality of our manuscript. We have carefully addressed each of your comments and revised the manuscript accordingly.

Sincerely,

Jinchun Yi

On behalf of all co-authors

Wuhan University

This study proposes a framework to estimate urban fossil fuel CO₂ emissions by combining satellite-derived NO_x emissions with active CO₂ lidar observations. Specifically, NO_x emissions are first inferred from TROPOMI NO₂ columns using a mass-balance approach and then converted to prior CO₂ emissions using CO₂-to-NO_x ratios. The prior emissions are subsequently constrained by XCO₂ observations from DQ-1 through a Bayesian inversion coupled with WRF-STILT simulations. The framework is applied to three cities-Beijing, Paris, and Cairo, to evaluate the influence of different CO₂-to-NO_x ratio estimation methods on inferred emissions. The manuscript presents an interesting attempt to integrate active and passive satellite observations for urban CO₂ emission monitoring, and the discussion of different CO₂-to-NO_x ratios touches upon the key challenges in recent studies on indirect CO₂ emission estimates. However, several methodological aspects and the logical framework of the study require further clarification. The manuscript requires major revisions to address the following comments before it can be considered for publication.

ANSWER: Thank you for your positive comments. We really appreciate your encouragement and support. To facilitate the readers' understanding of this study, we have carefully revised the whole manuscript according to your comments.

Comments:

Title and Figure 1: The manuscript's main objective is somewhat unclear. Although the Abstract, Introduction, Section 4 and Section 5 emphasize the importance of uncertainties in the CO₂-to-NO_x emission ratio for FFCO₂ estimation, this key aspect is not reflected in the title or clearly illustrated in Figure 1. Clarifying the important role of the CO₂-to-NO_x ratio in the title and conceptual framework would improve the overall coherence of the manuscript.

ANSWER: Thank you very much for your comments. We agree with your observation that "the main objective of the manuscript is somewhat unclear." To address this, we have emphasized the CO₂-to-NO_x emission ratio in both the title and Fig. 1. The title has been revised to: Active and Passive Satellite Observations Coupled with Carbon-Nitrogen Synergy for Urban Fossil Fuel CO₂ Emissions Monitoring. This highlights the core idea of the study, namely, using the carbon-nitrogen synergy approach to invert urban fossil fuel CO₂ emissions. We have also revised the conceptual

framework in Fig. 1 according to your suggestion.

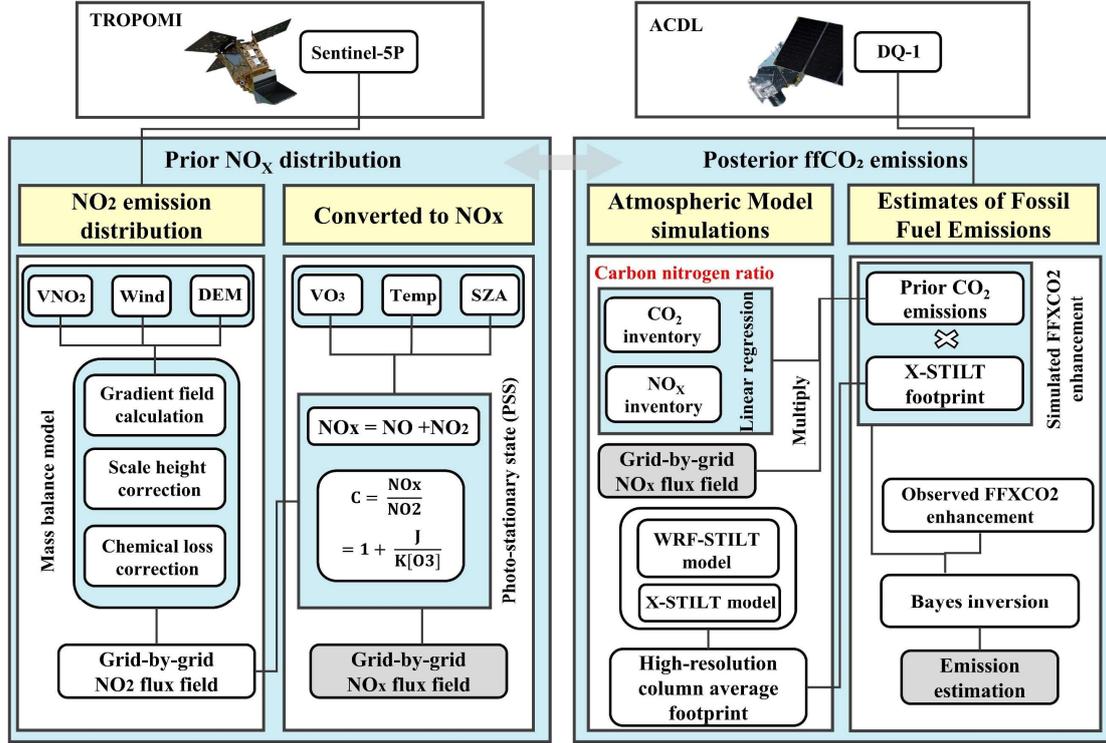


Figure 1 Technical framework diagram

Section 2 (Materials and Methods): The divergence-based approach is a flexible and low-cost method that has been widely applied in previous studies. However, it can be strongly affected by white noise in NO₂ observations due to the nonlinear nature of the divergence terms. In the manuscript, the authors evaluate this effect using the ratio of the standard deviation to the mean of the column concentrations. It would be helpful if the authors could further clarify the range of these ratios and provide additional discussion on their variability. Previous studies suggest that the magnitude of this uncertainty may vary considerably, typically ranging from approximately 10% to 40%. The authors are encouraged to provide appropriate references to support this range.

ANSWER: We have revised Appendix A5 accordingly:

The uncertainty of the NO_x inventory derived from the mass balance approach can be estimated using the error propagation law as follows:

$$\epsilon_{NO_x} = \sqrt{\epsilon_{\alpha}^2 + \epsilon_{NO_2}^2} \quad (1)$$

where ϵ_{α} represents the uncertainty in the NO_x/NO₂ ratio, its uncertainty arises from the uncertainties in the input parameters of the chemical model (Liu et al., 2022). And ϵ_{NO_2} denotes the uncertainty in the NO₂ flux field. The latter can be further decomposed as:

$$\epsilon_{NO_2} = \sqrt{\epsilon_{TROPOMI}^2 + \epsilon_{Wind}^2 + \epsilon_{Fit}^2} \quad (2)$$

Here, $\epsilon_{TROPOMI}$ is the uncertainty of the NO₂ column concentration, ϵ_{Wind} represents the uncertainty associated with the wind field, and ϵ_{Fit} accounts for the uncertainty in the fitted vertical

scale height and chemical lifetime. The uncertainty of NO₂ arises from multiple factors, including spectral fitting, stratospheric correction, AMF, clouds, vertical profiles, and surface albedo(Boersma et al., 2018; Verhoelst et al., 2021; Van Geffen et al., 2022; Boersma et al., 2004; Martin et al., 2002; Lu et al., 2025). In this study, we use the ratio of the standard deviation to the mean of the column concentration within the study area as a proxy for the TROPOMI observational noise, integrated over the time series. It should be noted that this proxy is calculated based on the oversampled gridded data (also referred to as Level-3) rather than the original Level-2 orbit data. In this study, we do not account for errors introduced during the oversampling of TROPOMI L2 data to the grid(Glissenaar et al., 2025). With appropriate gridding, the uncertainty in polluted areas can be reduced by approximately 20% compared with the original orbits(Sun et al., 2018). Wind field uncertainty is quantified through 10⁴ Monte Carlo perturbations of wind speed and direction, with the propagated standard deviation representing the flux variability. The fitting uncertainty is obtained by performing 10⁴ Monte Carlo draws of the grids involved in the fit, generating ensembles of scale heights and chemical lifetimes, with the final fitting error defined as the root mean square of the standard deviations of these ensembles.

Using the method described above, we quantified the overall uncertainty of NO_x prior emissions for three cities, as well as the contributions from individual components, with the detailed results summarized in Table X. It should be noted that the uncertainties reported here represent aggregated values for the entire urban area, rather than detailed uncertainties for individual grid cells.

Based on the uncertainty calculations, the total uncertainty follows the order Beijing (23.79%) > Paris (18.15%) > Cairo (14.31%). A closer look at the contributions of individual components reveals that NO₂ column concentrations and the wind field are the dominant sources, together accounting for more than 66.7% of the total uncertainty. This is attributable to the nature of data-driven dispersion models, in which uncertainties in wind and concentration directly govern the overall uncertainty(Sun, 2022). The nonlinear gradient operations in dispersion models (e.g., second-order difference operators) can amplify white noise in the original concentration field, while in existing emission quantification models, wind fields are considered a major source of uncertainty due to sparse monitoring sites and model errors(Huang et al., 2025).

Among the three cities, NO₂ column inversion uncertainty is highest in Beijing. Unlike Cairo, where high surface reflectivity eases retrievals, Beijing is located in the highly polluted North China Plain with elevated AOD, which increases the difficulty of passive column inversion. In addition, Beijing's complex terrain contributes to the highest wind field uncertainty (~17%) among the three cities. The NO_x/NO₂ uncertainty is roughly similar across the three cities, consistent with previous studies using NU-WRF (~1.4 ± 0.1). In contrast, the uncertainty associated with first-order chemical lifetime and vertical scale height is the lowest among all components (~1%). This is different from earlier studies (~15%) (Liu et al., 2022)and reflects the benefit of the data-driven fitting approach proposed by Sun et al. (see main text). Since no new assumptions were introduced in the current study, this uncertainty arises solely from the linear fitting model.

Table A3 The overall uncertainty of NO_x emissions and the uncertainties of individual components were derived using the dispersion model.

	NO _x /NO ₂ uncertainty(%))	NO ₂ uncertainty(%))	Wind uncertainty(%))	Fitted uncertainty(%))	Total uncertainty(%))
Beijing	6.51	15.49	16.76	1.67	23.79
Cairo	4.79	11.64	6.76	0.78	14.31
Paris	5.02	13.67	10.76	1.21	18.15

Section 2 Materials and Methods: The manuscript uses observations from DQ-1 satellite and Sentinel-5P TROPOMI. However, the data description does not clearly specify whether the two satellites have similar overpass times or how the temporal differences are handled. Since both CO₂ and NO₂ can exhibit noticeable diurnal variability, differences in satellite overpass times may potentially introduce additional biases when applying the Bayesian inversion for the CO₂-to-NO_x emission ratio. For example, Sentinel-5P TROPOMI typically has an overpass time around 13:30 local time, while the DQ-1 satellite observations used in this study appear to correspond to nighttime conditions (e.g., around 23:00 on 19 August, according to the case shown). It would be helpful if the authors could clarify the overpass times of the satellites used and explain whether any temporal matching or adjustment has been applied to ensure the consistency of the observations. Several studies have discussed these in appropriate way:

ANSWER: Thank you very much for raising this important point. Owing to our oversight, the temporal matching issue involved in the inversion process was not clearly described in the main text. We will add a detailed explanation and include the corresponding clarification in the revised manuscript.

It should be noted that this study uses the monthly NO_x emission fields derived from a TROPOMI NO₂-driven divergence method, rather than directly comparing TROPOMI NO₂ observations with DQ-1 measurements. The two studies you mentioned both rely on simultaneous CO₂ and NO₂ observations from OCO-2/3 and TROPOMI. Therefore, they need to address the “temporal matching or adjustment has been applied to ensure the consistency of the observations.” as you pointed out.

In contrast, our study mainly addresses the temporal matching between the monthly NO_x emission fields derived from the divergence method and the STILT footprints. Specifically, we use the temporal profiles provided by the New High Resolution Temporal Profiles in EDGAR dataset (https://edgar.jrc.ec.europa.eu/dataset_temp_profile) to distribute the monthly NO_x emissions to each hourly time step of the STILT footprint simulations. This approach allows us to obtain the corresponding NO_x emission field for the study region for each hourly STILT footprint.

We have added a clarification in Section 2.2.2 (3) of the revised manuscript. (Page 12, Line 284-287)

Figure 3: It is difficult to discern the spatial patterns in b, c, and d without city boundaries. Could the authors clarify where the boundaries are? Additionally, regarding the fitted CO₂-to-NO_x ratio, was any filtering applied? For instance, in Paris, there appear to be numerous grid cells with values

below 200, yet these low values do not seem to appear in Figure 3e. If such low ratios were filtered out, it should be explained that how the remaining grids can represent the CO₂-to-NO_x ratio for the entire city.

ANSWER: Thank you very much for your comment. In the revised manuscript, we have added the city boundaries to Fig. 3. Regarding your question, “Additionally, regarding the fitted CO₂-to-NO_x ratio, was any filtering applied?”, no filtering was applied during the analysis.

You also mentioned that “For instance, in Paris, there appear to be numerous grid cells with values below 200, yet these low values do not seem to appear in Figure 3e.” This is because Figs. 3b–d and Figs. 3e–g represents two different methods for calculating the CO₂-to-NO_x ratio. Figs. 3b–d show the gridded CO₂-to-NO_x ratios obtained by directly dividing the CO₂ emissions by the NO_x emissions from the CAMS inventories for each grid cell. In contrast, Figs. 3e–g show the overall CO₂-to-NO_x ratio obtained from a linear fit between the CO₂ and NO_x emissions across all grid cells. Therefore, the ratios from these two approaches do not correspond one-to-one within the same city. This explains why many grid cells in the Paris region have ratios below 200 in Figs. 3b–d, while these low values are not reflected in Fig. 3e.

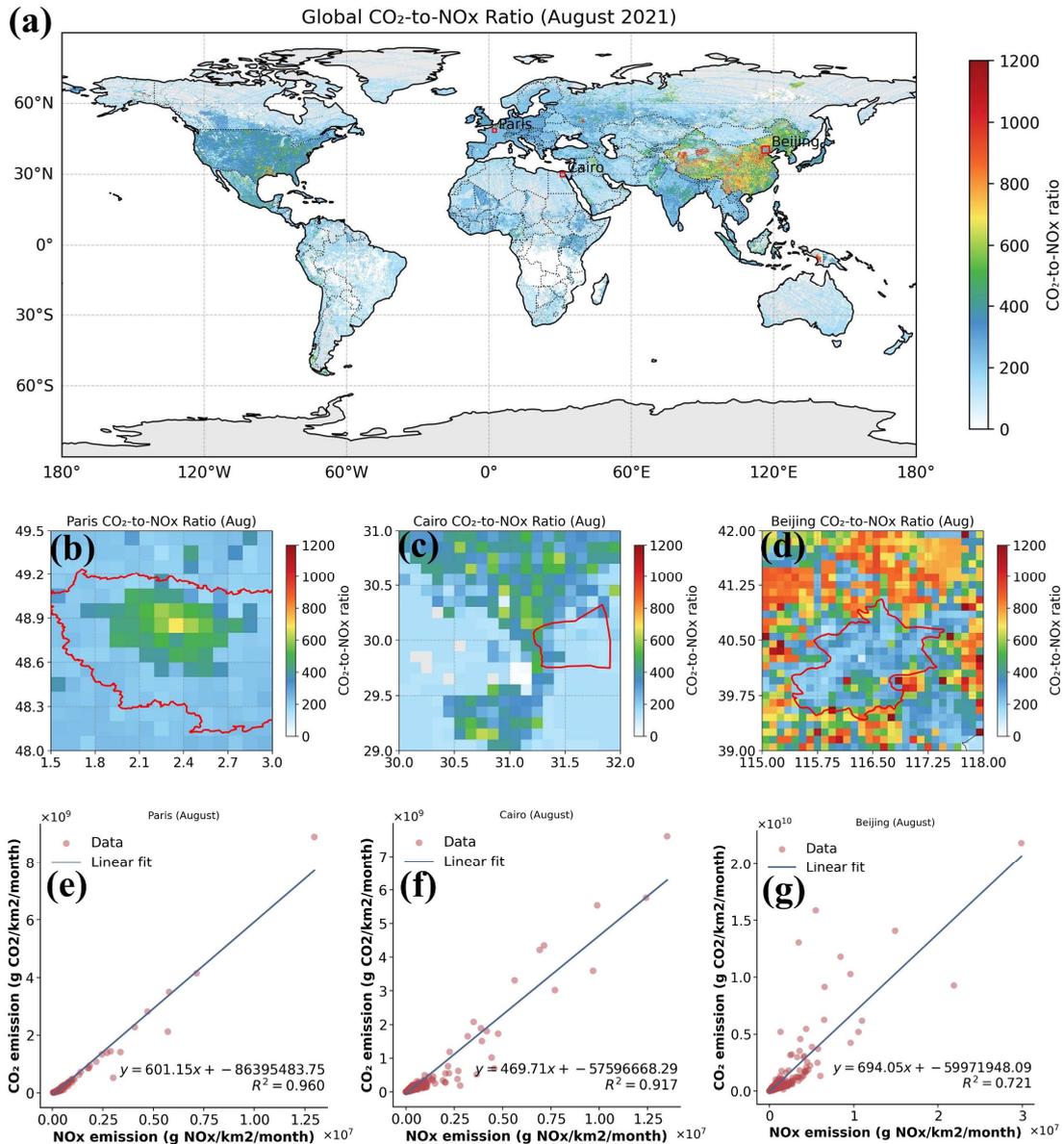


Figure 3 Schematic diagram of prior CO₂-to-NO_x ratio calculation methods. Panel (a) shows the global gridded CO₂-to-NO_x ratio derived from GMES data. Panels (b)–(d) present the gridded CO₂-to-NO_x ratio for Paris, Cairo, and Beijing (the red lines indicate the boundaries of each city). Panels (e)–(g) display the overall CO₂-to-NO_x ratio fitting results for the three cities. We used the Île-de-France administrative boundary to depict Paris in the figures, rather than the city proper. Although our actual study area only covers a subset of Île-de-France (1.5–3° E, 48–49.5° N)

Section3: The manuscript emphasizes the high-resolution of the derived emission estimates. However, several key parameters in the framework, such as the CO₂-to-NO_x ratio, seems to be treated as fixed values at the city scale (only one value per city for the studied month), without accounting for potential temporal or spatial variability. In addition, although the study focuses on monitoring urban fossil fuel CO₂ emissions, no figures are provided to illustrate the spatial distribution of CO₂ within these cities. Including such information would better support the claims of high-resolution emission estimates.

ANSWER: Thank you very much for your comment. First, regarding your point that “The manuscript emphasizes the high-resolution of the derived emission estimates. However, several key parameters in the framework, such as the CO₂-to-NO_x ratio, seems to be treated as fixed values at the city scale (only one value per city for the studied month)” we would like to clarify that the high resolution we refer to is primarily spatial resolution—for example, the 5 km NO_x emission fields derived from the divergence method, coupled with the high-resolution atmospheric transport model WRF-STILT, which together produce high-resolution urban CO₂ plumes.

We agree that the CO₂-to-NO_x ratio may exhibit potential temporal and spatial variability. However, there is currently no open-access emission dataset that provides daily or hourly CO₂ and NO_x emissions, which would allow calculation of the ratio for each STILT footprint time step. It is also important to note that our inversion scales the total city emissions as a whole. Therefore, instead of assigning a CO₂-to-NO_x ratio to each emission grid, we applied a fixed ratio for the entire city (as described in the Methods section, where different approaches to calculate the ratio are introduced).

While this choice may introduce uncertainties associated with temporal and spatial variability, we account for such uncertainties in the prior CO₂-to-NO_x ratio by performing a Monte Carlo perturbation of the gridded CO₂ and NO_x emissions 10,000 times, which incorporates variability across space to some extent.

Regarding your second point, “In addition, although the study focuses on monitoring urban fossil fuel CO₂ emissions, no figures are provided to illustrate the spatial distribution of CO₂ within these cities. Including such information would better support the claims of high-resolution emission estimates.” we will provide the posterior emission for each city in the appendix A7 to show the spatial distribution of CO₂ within the cities. In addition, the colored shading in Figs. 5, 6, and 7 illustrates the simulated urban CO₂ plumes and concentrations during several hours prior to the satellite overpasses.

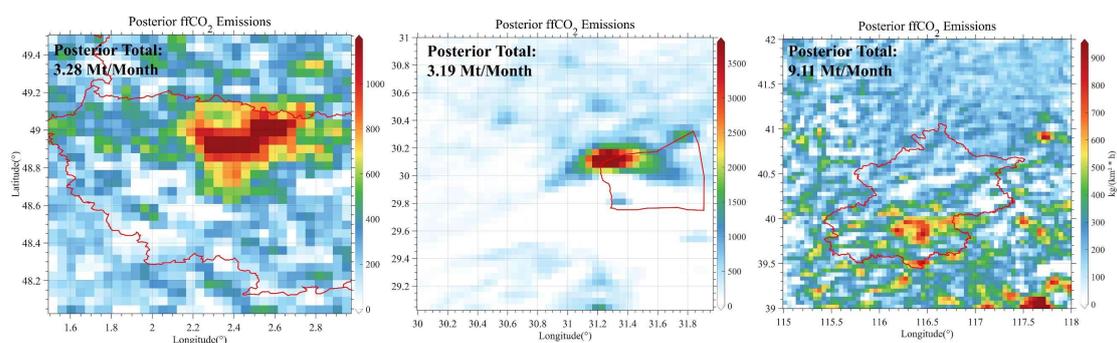


Figure A1: Posterior fossil fuel carbon dioxide emissions for each city. The red lines outline city boundaries, while the colored shading indicates carbon dioxide emission distribution.

Section 4: According to the proposed framework, CO₂ emissions are estimated using NO_x emissions together with the CO₂-to-NO_x emission ratio. Therefore, the determination and evaluation of this ratio appear to be a key step in the overall methodology. However, the discussion of the CO₂-to-NO_x ratio is mainly presented in Section 4 and Section 5, after the CO₂ emission results are shown. From a methodological perspective, it might be clearer if the definition, evaluation, and potential adjustment of the CO₂-to-NO_x ratio were introduced earlier, as part of the methodological framework leading to the CO₂ emission estimates.

ANSWER: We sincerely thank you for your careful review and comments. We will adjust the manuscript structure accordingly to improve the clarity of the argument.

Section 4: The manuscript discusses both the prior CO₂-to-NO_x ratio and the posterior ratio obtained from the Bayesian inversion. However, it is not explicitly stated which of these ratios is ultimately used to derive the final CO₂ emission estimates. If the prior ratio is used for the final CO₂ calculation, it raises the question of why considerable effort was devoted to constraining the posterior ratio, which apparently is not applied in deriving the final emissions.

ANSWER: We sincerely thank you for your careful reading and comments. This study aims to present an inversion framework for estimating urban anthropogenic CO₂ emissions using both active and passive satellite observations, and to discuss the prior and posterior uncertainties associated with different CO₂-to-NO_x ratio calculation methods.

Regarding your comment that “However, it is not explicitly stated which of these ratios is ultimately used to derive the final CO₂ emission estimates.” we acknowledge that this was not explicitly clarified in the main text. We appreciate your raising this point. In the revised manuscript, we will clarify the satellite-constrained CO₂ emissions for different cities and provide the posterior CO₂ emission distributions for each city in the appendix A7.

It should be noted that the posterior CO₂ emissions for each city are calculated using the posterior CO₂-to-NO_x ratio obtained with the M1 method.

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