MS-No.: egusphere- 2026-538

Title: Synergistic Use of Active and Passive Satellite Observations for Monitoring Urban Fossil Fuel CO2 Emission

Authors: Jinchun Yi, Yiyang Huang, Ge Han, Hongyuang Zhang, Zhipeng Pei, Haotian Luo, Yichi Zhang, Tianqi Shi, Siwei Li, and Wei Gong

Item-by-item reply to Anonymous Referee

March 30, 2026

Dear Jason Cohen and Reviewers,

We sincerely thank you and the reviewers for your constructive and thoughtful feedback on our manuscript. We have revised the manuscript accordingly and believe the revised version has improved in both clarity and scientific rigor. Please find below a detailed, point-by-point response to each reviewer comment.

1. The reviewers' comments are shown in black.

2. Our responses are also in blue, following each comment.

3. Descriptions of changes made to figures and tables are formatted in bold black text for clarity.

All modifications made to the manuscript are clearly marked in the revised version.

We greatly appreciate the reviewers' careful assessment and your editorial support. We hope the revised manuscript now meets the standards for publication.

Sincerely,

Jinchun Yi

On behalf of all co-authors

Wuhan University

Email: jinchun.yi@whu.edu.cn

Reply to Referee # 1

Dear reviewer:

Thank you very much for reviewing our manuscript and providing valuable feedback. We sincerely appreciate your patience and detailed comments. Under your guidance, we have carefully addressed each of your comments and made corresponding revisions and additions. All your words are in black and our item-to-item responses are in blue. We have included our revisions for some comments directly for your convenience.

Sincerely,

Jinchun Yi

On behalf of all co-authors

Wuhan University

This study develops a top-down inversion framework to estimate high-resolution, city-scale fossil-fuel CO_2 ($ffCO_2$) emissions by integrating active and passive satellite observations and coupling CO_2 and NO_x emissions through NO_x distributions and CO_2-to-NO_x emission ratios. It further evaluates how different approaches for deriving the CO_2-to-NO_x ratio influence the inferred $ffCO_2$ emissions, showing that optimized ratio estimation enhances emission accuracy and reduces uncertainties in both the ratio and the resulting inversions. The framework is applied to three major metropolitan areas—Beijing, Cairo, and Paris—with comprehensive supporting analyses provided. By improving the estimation of urban carbon emissions at the city scale and lowering associated uncertainties, this work offers valuable contributions to the community for more robust emission quantification and better-informed carbon mitigation strategies.

I recommend this manuscript for publication, with only a few questions and suggestions for consideration.

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.

Mandatory changes:

1: Since Aura OMI and OCO-2/3 are not utilized in this study, they should be removed from the figure. In addition, the connections between the different steps of the workflow are not sufficiently clear. The flowchart could be revised to improve clarity and logical progression.

ANSWER: Thank you very much for your comments. We fully agree with your suggestions regarding Fig. 1. In the revised manuscript, we removed Aura OMI and OCO-2/3 and revised the flowchart to improve its clarity and logical consistency.

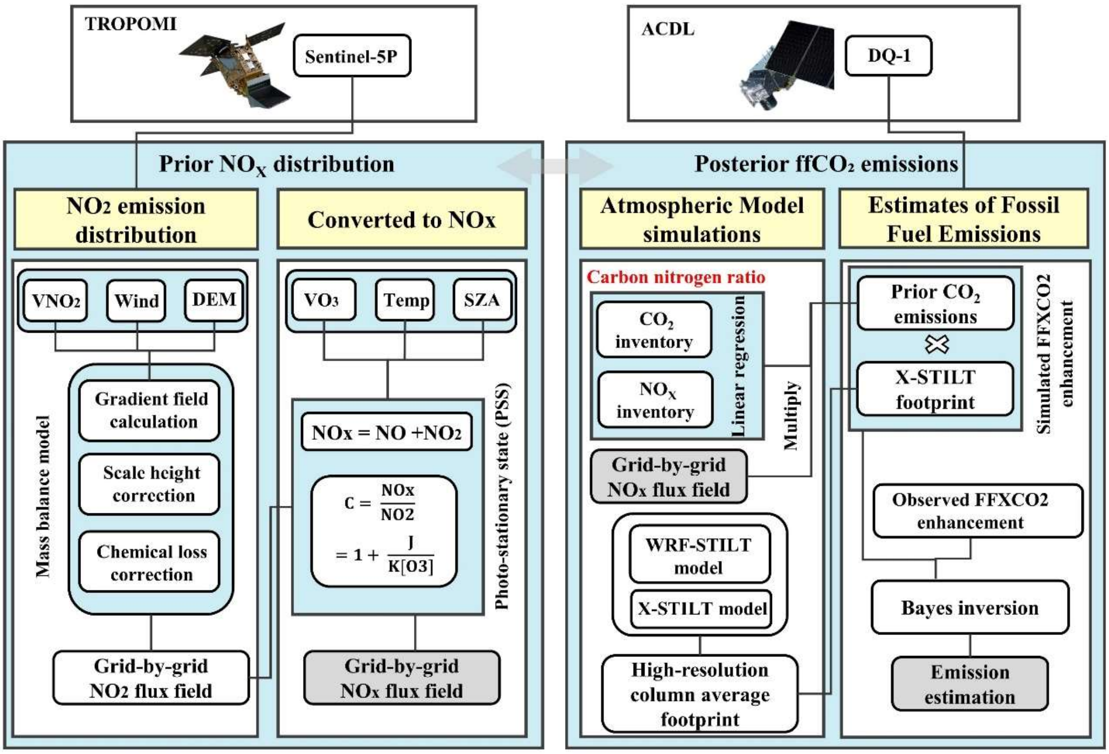

Figure 1 Technical framework diagram

Section 2.1.1 ACDL Products: The statement "which enables direct 'top-down' observations of atmospheric CO_2" is misleading. While the use of an active lidar sensor is indeed a technological breakthrough, it does not inherently enable "direct" top-down emission estimates. This phrasing should be revised for accuracy.

ANSWER: Thank you very much for your comments. We fully agree with your comment. In the revised manuscript, we will refine this description to make it more accurate. Specifically, we will replace “which enables direct 'top-down' observations of atmospheric CO_2” with “which enables active top-down observations of atmospheric CO_2.” (Page 5, Line 106)

P11, Eqs. (4)–(5): Section 2.2.2 (2) is difficult to follow due to insufficient explanation of the parameters in Eqs. (4)–(5). Please revise these equations and clearly describe each variable and its physical meaning.

ANSWER: Thank you very much for your comments. We followed your suggestion and clearly described each variable in Equations (4) and (5) and its physical meaning. (Page 12, Line 272-276, Line 283)

Section 3.2.3: The posterior uncertainties reported in the text do not match the ones stated in table 2. The uncertainties reported in table are significantly larger. What is the reason?

ANSWER: Thank you very much for your comments. This was our oversight. The posterior uncertainty reported in Table 2 is not a percentage uncertainty; however, we mistakenly added the symbol “%” in the column header. We have corrected this issue in the revised manuscript. The uncertainties in the table are calculated as the posterior C/N ratio multiplied by the percentage uncertainty, whereas the values reported in the text represent the percentage uncertainty. Therefore, the uncertainties listed in the table are significantly larger than the values reported in the text. (Page 24 Table 2)

Recommended minor changes:

P1, L23: XCO_2 should be defined at its first occurrence in this paper.

ANSWER: Revised (Page 1, Line 21)

P2, L49–51: I would recommend adding more references by citing more relevant papers.

ANSWER: Revised (Page 2, Line 50)

P3, L63: Please clarify what specific measurement limitations are being referred to here.

ANSWER: We have replaced "due to the measurement limitations" with "because the CO_2-to-NO_x ratio used in this study is calculated from CO_2 emissions and NO_x emissions, there is currently a lack of accurate top-down measurement methods" (Page 3, Line 65-67)

P5, Fig. 2: It may be more appropriate to relocate this figure to Section A1, where the parameter details are described.

ANSWER: Thank you for your suggestion regarding Fig. 2. We appreciate your comment; however, we prefer to keep the figure in its current location in the main text because it provides important context for the ACDL productions and helps readers follow the analysis more easily. The detailed parameter descriptions are still provided in Section A1 for reference.

P7, L174: Sun et al. (2018) does not appear to be directly relevant to flux estimation. More appropriate references would include Sun et al. (2022; https://doi.org/10.1029/2022GL101102) and Ayazpour et al. (2025; https://doi.org/10.1029/2024JD042817), which focus specifically on flux estimation methodologies.

ANSWER: Revised (Page 7, Line 177-178)

P9, L202-212: It would be helpful to elaborate on how the scale height and chemical lifetime are determined.

ANSWER: Thank you for your comment. This process is already described in the manuscript: "To suppress excessive noise in single-day fits, we perform monthly regressions and adopt the temporal and spatial mean over the month as the final estimate, representing an aggregate over the full spatial domain, the entire month, and the troposphere. The retrieved scale height and first-order chemical lifetime are then applied back into Equations 4 and 6 to obtain the final gridded NO_x vertical fluxes." We believe this description provides sufficient detail on how the scale height and chemical lifetime are determined.

P12, Eq. (11): Please specify the definition of Sobs.

ANSWER: S_{obs} is a diagonal matrix, with the diagonal entries representing the observational error variances ε_{obs}^2 for each orbit. (Page 13, Line 318)

P16, L336 we don't say 'concentration' when we talk about emissions.

ANSWER: Thank you for your comment. We agree that the term "concentration" is not appropriate when referring to emission. We have revised the manuscript to replace it with “emission” wherever necessary. (Page 17, Line 367)

P16, Table 1: Are the chemical lifetime and scale height values spatially averaged across each city?

ANSWER: Yes

P28, L548: How is the prior uncertainty of the CO_2-to-NO_x ratio treated in experiments M4–M6?

ANSWER: We have stated in the main text that M4 and M6 are the same as M2, and M5 is the same as M1. (Page 11, Line 243-254) Their uncertainty calculation methods are also identical; the only difference lies in the emission inventories used.

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 NOx emissions with active CO2 lidar observations. Specifically, NOx emissions are first inferred from TROPOMI NO2 columns using a mass-balance approach and then converted to prior CO2 emissions using CO2-to-NOx 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-NOx 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-NOx 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-NOx 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-NOx 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 CO_2-to-NOX 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 CO_2 Emissions Monitoring. This highlights the core idea of the study, namely, using the carbon–nitrogen synergy approach to invert urban fossil fuel CO_2 emissions. We have also revised the conceptual framework in Fig. 1 according to your suggestion.

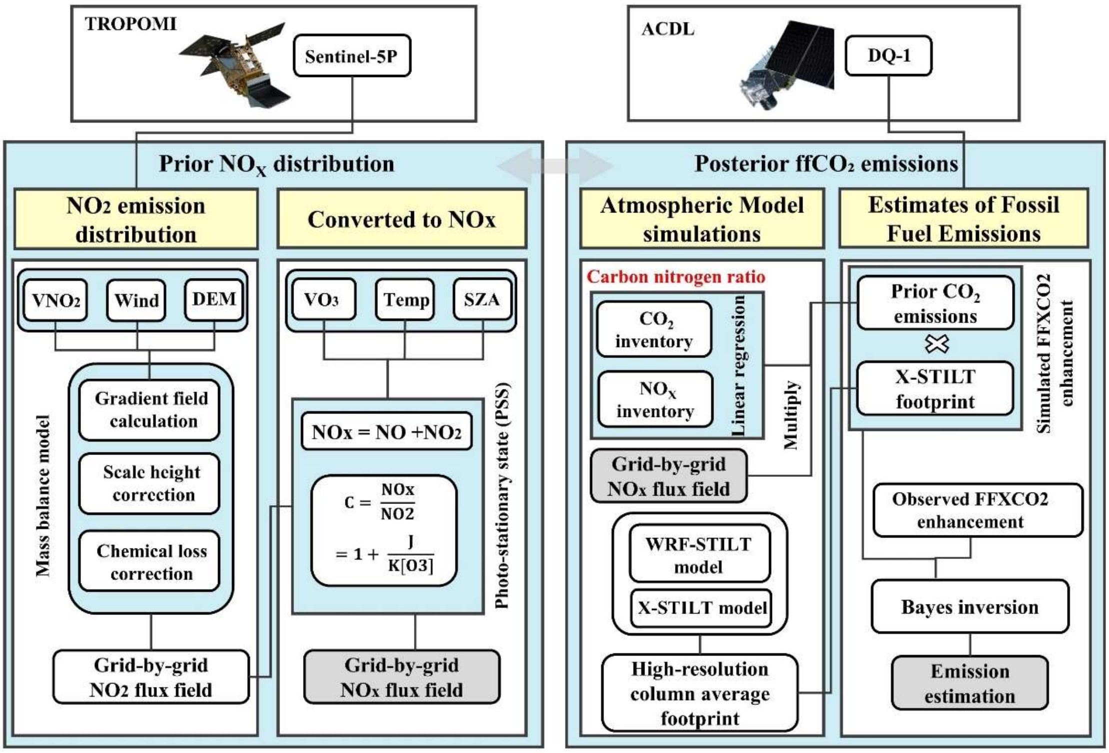

Figure 2 Technical framework diagram

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:

$$\varepsilon_{NOx} = \sqrt[2]{\varepsilon_{\alpha}^{2} + \varepsilon_{NO2}^{2}} \tag{1}$$

where ε_{α} represents the uncertainty in the NO_x/NO_2 ratio, its uncertainty arises from the uncertainties in the input parameters of the chemical model(Liu et al., 2022). And ε_{NO} denotes the uncertainty in the NO_2 flux field. The latter can be further decomposed as:

$$\varepsilon_{NO2} = \sqrt[2]{\varepsilon_{TROPOMI}^{2} + \varepsilon_{Wind}^{2} + \varepsilon_{Fit}^{2}} \tag{2}$$

Here, $\varepsilon_{TROPOMI}$ is the uncertainty of the NO_2 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 NO_2 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 10^4 Monte Carlo perturbations of wind speed and direction, with the propagated standard deviation representing the flux variability. The fitting uncertainty is obtained by performing 10^4 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 NOx 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 NOx/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 NOx emissions and the uncertainties of individual components were derived using the dispersion model.

	NO_x/NO_2 uncertainty(%)	NO2 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-NOx 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 NOx emission fields derived from a TROPOMI NO_2-driven divergence method, rather than directly comparing TROPOMI NO_2 observations with DQ-1 measurements. The two studies you mentioned both rely on simultaneous CO_2 and NO_2 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 NOx 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 NOx emissions to each hourly time step of the STILT footprint simulations. This approach allows us to obtain the corresponding NOx 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 13, Line 305-308)

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-NOx 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-NOx 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-NOx 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 CO_2-to-NOx ratio. Figs. 3b–d show the gridded CO_2-to-NOx ratios obtained by directly dividing the CO_2 emissions by the NOx emissions from the CAMS inventories for each grid cell. In contrast, Figs. 3e–g show the overall CO_2-to-NOx ratio obtained from a linear fit between the CO_2 and NOx 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.

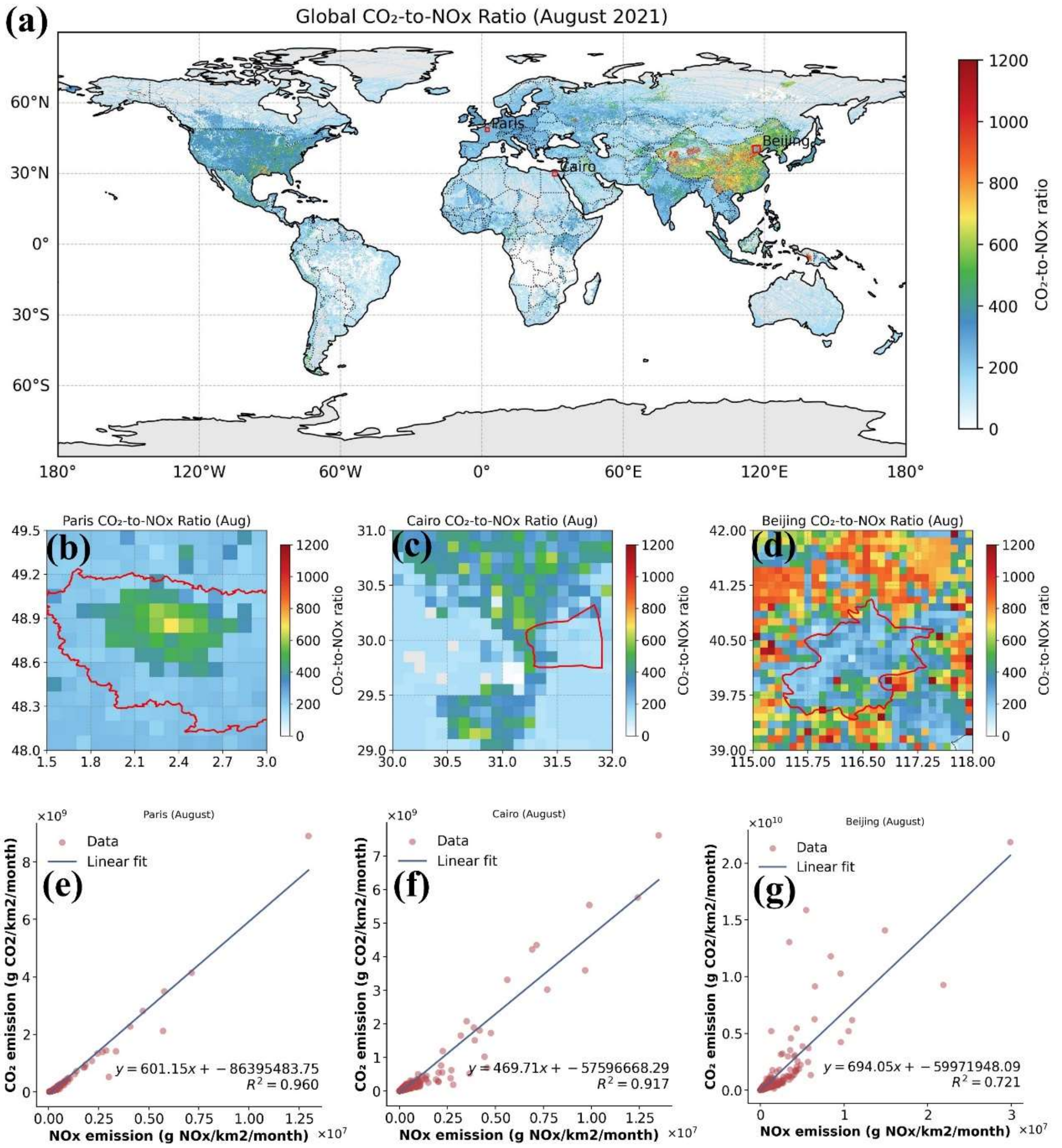

Figure 3 Schematic diagram of prior CO_2-to-NO_x ratio calculation methods. Panel (a) shows the global gridded CO_2-to-NO_x ratio derived from GMES data. Panels (b)–(d) present the

gridded CO_2-to-NO_x ratio for Paris, Cairo, and Beijing (the red lines indicate the boundaries of each city). Panels (e)–(g) display the overall CO_2-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-NOx 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-NOx 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 NOx emission fields derived from the divergence method, coupled with the high-resolution atmospheric transport model WRF-STILT, which together produce high-resolution urban CO_2 plumes.

We agree that the CO_2-to-NOx ratio may exhibit potential temporal and spatial variability. However, there is currently no open-access emission dataset that provides daily or hourly CO_2 and NOx 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 CO_2-to-NOx 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 CO_2-to-NOx ratio by performing a Monte Carlo perturbation of the gridded CO_2 and NOx 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 CO_2 within the cities. In addition, the colored shading in Figs. 5, 6, and 7 illustrates the simulated urban CO_2 plumes and concentrations during several hours prior to the satellite overpasses.

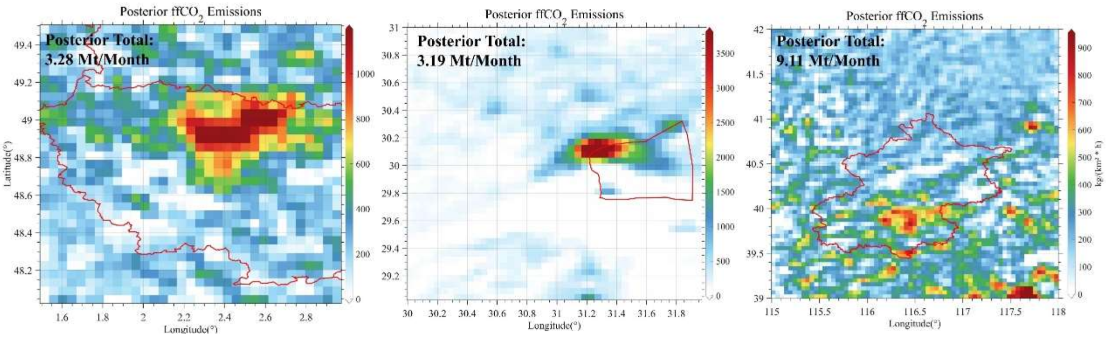

Figure A1: Posterior fossil fuel carbon dioxide emissions for each city. The red lines outline city boundaries, while the colored shading indicates carbon dioxide emission distribution.

Section 4: According to the proposed framework, CO2 emissions are estimated using NOx emissions together with the CO2-to-NOx 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-NOx 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-NOx 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-NOx 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 CO_2 emissions using both active and passive satellite observations, and to discuss the prior and posterior uncertainties associated with different CO_2-to-NOx 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 CO_2 emissions for different cities and provide the posterior CO_2 emission distributions for each city in the appendix A7.

It should be noted that the posterior CO_2 emissions for each city are calculated using the posterior CO_2-to-NOx ratio obtained with the M1 method.

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