Reviewer: 2

This work presents a robust uncertainty analysis for an established mass balance inversion scheme capable of inferring CO_2 emissions from TROPOMI's NO_2 measurements. While I do not take issue with the results presented in this manuscript, I found myself carefully re-reading the text multiple times to try and find information I felt to be crucial to the methodology. Some of the information was found after multiple readings while some remained elusive. The omission of certain points in the methodology section and its lack of organization made reading difficult. I have listed my comments, both major and minor, below.

Response:

We express our gratitude to the referee for constructive remarks regarding our manuscript. Below, we provide detailed responses addressing each point raised.

Major Comments

1. In Lines 38-40, the text mentions the "co-emissions characteristics in time and space" of NO_2 and CO_2 emissions, leveraging the linear relationship between the two (Yang et al., 2023; Fig. 1). However, in other work by the author (Li and Zheng, 2024; Paper highlight #2), they state that NO_x and CO_2 are inversely proportional (at least during COVID lockdowns). Upon first reading, this seems like a contradiction. Perhaps the relationship between NO_x and NO_2 emissions should also be discussed in the introduction, near lines 38-40. At least conceptually highlight the conversion from TROPOMI NO_2 to NO_x here, particularly how works (eqn. 2).

Response:

Anthropogenic NO_x and CO_2 are co-emitted, yet their sector-specific emission ratios differ, leading to potentially distinct trends in their total emissions. Specifically, emission controls implemented by the Chinese government have reduced NO_x emission factors (EFs) over time, while CO_2 EFs have remained stable, primarily due to their dependence on fuel type and combustion conditions. Thus, given the asynchronous changes in activity levels, NO_x EFs, and CO_2 EFs, differing trends in overall NO_x and CO_2 emissions are possible.

In the NO_x family, NO is the primary species emitted and undergoes rapid conversion to NO_2, which is also the component detectable by most satellites. Therefore, NO_2 effectively serves as a proxy for NO_x emissions in inversion studies. NO_x is a short-lived species, making its concentrations highly sensitive to emission sources. This enables the use of mass-balance methods to estimate NO_x emissions, which rely on the assumption of a linear relationship between NO_2 columns and local NO_x emissions (Cooper et al., 2017; Mun et al., 2023; Martin et al., 2003).

We have added some explanations in Lines 42-46, Lines 48-50, and Lines 98-100 in Manuscript.

Lines 42-46: "NO_2 forms rapidly after NO is emitted from sources and is also the primary nitrogen oxide detectable by most satellites (Ye et al., 2016). This makes NO_2 a reliable and widely adopted proxy in nitrogen oxides (NO_x = NO_2+NO) emission inversions. However, the co-emission of NO_x and CO_2 does not imply synchronized trends in their emissions, as the CO_2-to-NO_x emission ratios and activity trends vary across different sectors (Li and Zheng, 2024)."

Lines 48-50: "This short lifespan of NO_2 facilitates mass-balance approaches for estimating NO_x emissions, which rely on the assumption of a linear relationship between NO_2 columns and local NO_x emissions (Cooper et al., 2017; Mun et al., 2023; Martin et al., 2003)."

Lines 98-100: "A critical step in this process was establishing a linear relationship between NO_2 tropospheric vertical column densities (TVCDs) and anthropogenic NO_x emissions under the mass balance assumption (Eq. 2) through GEOS-Chem simulation (v12.3.0, https://geoschem.github.io/) at a horizontal resolution of 0.5°×0.625°."

2. Lines 46–50 claim that space-based observers of NO_2 have surpassed CO_2 observers in revisits, spatial resolution, and coverage. However, I question at least some aspects of this statement. While TROPOMI has a daily revisit time, it is restricted to a ~1:30 pm overpass time. The CO_2-observing OCO-3 instrument provides coverage at different times throughout daytime hours, providing the potential to elucidate diurnal emissions (albeit with a ~3 day revisit time). Additionally, OCO-3 has a higher spatial resolution than TROPOMI, on the order of 2km x 2km. Thus, it is my opinion that Lines 46-50 make unfair statements by not acknowledging the benefits of the OCO-3 instrument.

Response:

We have rephrased this sentence acknowledging the development of CO_2 satellites in Lines 53-60.

Lines 53-60: "Moreover, remote sensing technologies for NO_2 remain generally more mature, as indicated by the broader coverage and improved signal-to-noise ratio in column concentration observation (Macdonald et al., 2023; Cooper et al., 2022). Recent advancements in CO_2 satellite technology are promising, such as the Orbiting Carbon Observatory-3 (OCO-3), which can generate CO_2 maps with a resolution of up to 1.6 km × 2.2 km and monitor CO_2 columns at different times throughout the daytime to elucidate diurnal emission patterns (Taylor et al., 2023), while its spatial coverage may not be sufficient for large-area inversions at high temporal resolution."

3. Furthermore, this paper does not take into account the most recent efforts to measure sector-specific CO_2 emissions at a sub-annual scale (see Roten et al., 2023 for example). The title of this work "Air Pollution Satellite-based CO_2 Emission Inversion: System Evaluation, Sensitivity Analysis, and Future Perspective" suggests that the focus will be on the uncertainty/error of the posterior CO_2 estimates. There is little discussion of the current uncertainties of these measurements, approximated with "direct" CO_2 observations, not NO_x. Results should be presented in light of recent OCO-2, OCO-3, etc work. Several publications include city- and sub-city-level emission estimates using CO_2 observations, not CO_2 approximations. Consider uncertainties determined by Yang et al., 2020 and Ye et al., 2020 presenting constraints on CO_2 emissions using CO_2 observations directly. (Of course, results presented here are sector-specific. Yang and Ye are not.)

(Roten: https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2023GL104376)

(Yang: https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2019JD031922)

(Ye: https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2019JD030528)

Response:

We have added some discussion of CO_2-observing CO_2 emission inversion in the Discussion Section (Lines 432-436).

Lines 432-436: "Notably, remarkable advancements have been achieved in estimating subnational CO_2 emissions through CO_2-observing satellites, such as sectoral CO_2 assessments with OCO-3 (Roten et al., 2023), and urban emission optimizations utilizing the Orbiting Carbon Observatory-2 (OCO-2) (Yang et al., 2020; Ye et al., 2020). Yet, reducing uncertainties at subnational scales remains an ongoing challenge."

4. The authors should consider reordering the methodology sections. For example, moving 2.1 (Base Inversion) after 2.2.4 and updating the text would let Sections 2.2.1-2.2.4 provide more context in the presentation of equations 1-4. The way the methodology is currently presented is quite confusing. I found myself rereading these sections multiple times to really understand what was going on. Several of these sections are missing helpful information. For example, the section titled "Prior Emission Inventory" (2.2.1) never actually mentions the name of the inventory being used. This made tracking down information difficult throughout my reading of the manuscript. Furthermore, for readers who are unfamiliar with the MEIC inventory, a figure like Fig. 1 of Roten et al., 2023 would be helpful.

Response:

The original structure of the Methods section is organized as follows: we begin with an overview of the inversion methodology, using the Base inversion as a foundational example. This is followed by a detailed explanation of the rationale and methodology behind the sensitivity tests. To enhance clarity in discussing the total of 31 tests, we categorized the tested parameters into four classes based on their functions within the system. These categories include changes in prior updates, coarser model resolution, modifications to satellite observational constraints, and other systematic parameters, as depicted in Figure 1. To clarify our approach and reduce misleading, we have added more details about the methodology and re-order them in Section 2.1 (please refer to the Manuscript to track the changes in Section 2.1), added some explanatory notes, and revised the subtitles of Sections 2.1 and 2.2 as follows:

Sub-titles of 2.1: "2.1 Inversion methodology and Base inversion"

Line 87: "We use the Base inversion as a case to provide a detailed explanation of this inversion system."

Sub-titles of 2.2: "Sensitivity settings"

Line 152-158 "The sensitivity inversion experiments comprise 31 tests designed to provide a comprehensive evaluation of the system. To facilitate a clearer discussion of their impacts, we categorized these tests into four classes based on their roles within the system: prior information, GEOS-Chem model resolution, satellite observational constraints, and inversion system parameters (Fig. 1 and Table 2). Each test is conducted as a controlled experiment, where only one parameter is altered while the rest remain the same as their Base inversion setting. The rationale behind the settings and their design will be elaborated in the following sections."

Sub-titles of 2.2.1: "Modifying prior emission estimates"

Sub-titles of 2.2.2: "Employing coarser model resolution"

Sub-titles of 2.2.3: "Changing satellite observational constraints"

Sub-titles of 2.2.4: "Tests on inversion system parameters"

Besides, we have added a Fig. S2 displaying MEIC inventory in SI as suggested.

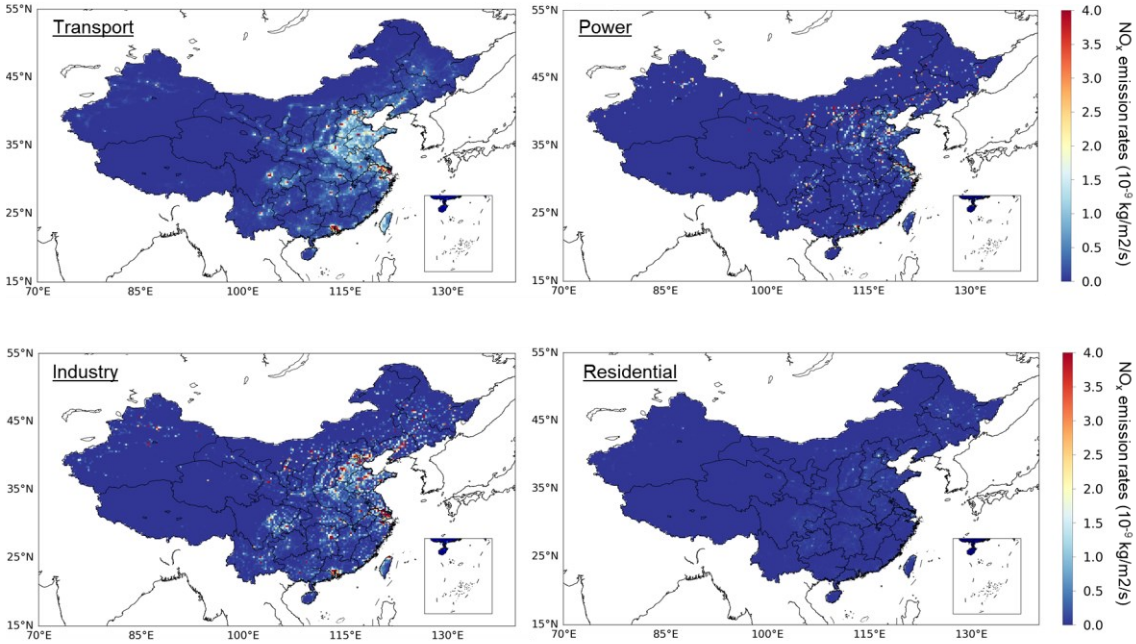

Figure S2. Sectoral NO_x emissions from MEIC inventory in 2019 (0.25°×0.25°).

5. From Line 114, "… while the CO_2 EFs are assumed to remain unchanged". If the emissions of NO_2, NO_x, and CO_2 are linked (Lines 38-40) what is the logic behind the assumption that CO_2 EFs remain unchanged? Should a scaling factor not be applied as well? This is not well explained.

Response:

The co-emission of CO_2 and NO_x does not imply aligned trends in their emission factors (EFs). NO_x EFs have consistently declined due to targeted end-of-pipe controls, with research documenting a continuous decrease in NO_x emissions in China since 2012, supporting this downward trend in NO_x EFs. In contrast, CO_2 EFs are primarily influenced by fuel type and combustion conditions, which have remained stable over time. We have added explanations in Lines 135-139 in Manuscript and Text S1 in SI.

Lines 135-139: "The CO_2-to-NO_x emission ratios in 2022 are updated by reducing NO_x emission factors (EFs) while keeping CO_2 EFs unchanged based on 2019 MEIC. The default assumption that the reduction rate halves annually is due to the limited potential for further reductions. In contrast, the CO_2 EFs are assumed to remain unchanged, as they are primarily determined by fuel type and combustion conditions (Cheng et al., 2021) (details seen in Text S2)."

Text S1. Bottom-up estimates

To derive a sector-specific prior, we update the 2019 Multi-resolution Emission Inventory for China (MEIC) (Zheng et al., 2018) using a range of activity data. The bottom-up estimation follows two primary steps: first, we apply monthly updates based on year-on-year national activity ratios obtained from the National Bureau of Statistics (https://data.stats.gov.cn/english/easyquery.htm?cn=C01); second, we disaggregate monthly emissions into daily estimates using multi-source data. The specific data sources used in this bottom-up approach are detailed in Table S1.

For emission factors (EFs), we assume a yearly halving of the reduction rate in NO_x EFs. Since 2012, NO_x emissions have sharply decreased due to effective pollution control measures with many end-of-pipe devices; however, the rate of decline has slowed in recent years, reflecting the diminishing potential for further reductions (Geng et al., 2024; Li et al., 2023). As such, the default assumption is that the reduction rate in NO_x EFs halves each year, consistent with the limited potential for continued reductions. By contrast, CO_2 EFs are assumed to remain constant over time, as they are primarily influenced by fuel type and combustion conditions (Cheng et al., 2021).

6. In Lines 88-89: "assuming that each grid's emission variability was primarily driven by its dominant source sectors (contributing over 50%)…". What about situations where no sectors make up more than 50% of a grid cell? Hypothetically, what if Power, Industry, Residential, and Transport all made up 25% of a grid cell? Do these situations not exist in the prior emission inventory? If not, why not? How is an observation-driven posterior estimate assigned to a grid cell when it doesn't meet the criteria?

Response:

For grids without a sector contributing over 50%, we excluded them from sectoral scaling factor calculations, instead applying scaling factors derived from grids meeting this criterion. Notably, over 80% of the grids have a sector contributing more than 50%, indicating a clear dominant sector for the majority of grids.

The overall NO_x emissions remain unaffected by this threshold parameter, as they are determined prior to disaggregation into sectors (Eq. 1). The threshold mainly impacts the sectoral distribution and the CO_2 emissions conversion process. We assessed the threshold's effect by adjusting it to 40% and 60% (thre_40% and thre_60%), and the results show that only residential emissions exhibit sensitivity due to their relatively low share of total emissions (Fig. 4 and Fig. S13).

We have added this explanation in Lines 123-126 in Manuscript.

Lines 123-126: "For grids without a sector contributing over 50%, we excluded them from sectoral scaling factor calculations, instead applying scaling factors derived from grids meeting this criterion. The number of these grids accounts for less than 20% of total grids, making their impact negligible."

Minor Comment

7. For readers who are not familiar with the mass balance inversion method, providing an additional citation, or explicitly pointing the reader to an additional resource, would be more helpful than simply citing Zheng et al., 2020 and Li et al., 2023. Pointing the readers to a paper such as Mun et al., 2023 or something similar will help make the connection between the inversion system being discussed and the corresponding equations 1-4.

(Mun: https://www.sciencedirect.com/science/article/pii/S1352231022004940)

Response:

We have added some introduction to the mass balance method in Lines 48-50 in the Introdution.

Lines 48-50: "This short lifespan of NO_2 facilitates mass-balance approaches for estimating NO_x emissions, which rely on the assumption of a linear relationship between NO_2 columns and local NO_x emissions (Cooper et al., 2017; Mun et al., 2023; Martin et al., 2003)."

8. Remove the word "here" in Line 59.

Response:

We have removed "here" in Line 69 (original 59) as suggested.

9. Add "of" in Line 77. "ten-day moving average of anthropogenic NO_x and CO_2"

Response:

We have added "of" in Line 89 (original 77) as suggested.

10. I understand the need to be succinct in Lines 78-81 regarding the scaling of emission sectors; however, it is my opinion that a little more information should be included here. The authors should consider including an extra statement explaining where these indicators came from. Were they from external an external inventory? Where they part of MEIC? Does MEIC contain sector-specific information already?

Response:

We have added more details regarding the bottom-up estimates in Text S1, along with Table S1 in SI, which outlines the data sources for activity levels.

Text S1. Bottom-up estimates

To derive a sector-specific prior, we update the 2019 Multi-resolution Emission Inventory for China (MEIC) (Zheng et al., 2018) using a range of activity data. The bottom-up estimation follows two primary steps: first, we apply monthly updates based on year-on-year national activity ratios obtained from the National Bureau of Statistics (https://data.stats.gov.cn/english/easyquery.htm?cn=C01); second, we disaggregate monthly emissions into daily estimates using multi-source data. The specific data sources used in this bottom-up approach are detailed in Table S1.

For emission factors (EFs), we assume a yearly halving of the reduction rate in NO_x EFs. Since 2012, NO_x emissions have sharply decreased due to effective pollution control measures with many end-of-pipe devices; however, the rate of decline has slowed in recent years, reflecting the diminishing potential for further reductions (Geng et al., 2024; Li et al., 2023). As such, the default assumption is that the reduction rate in NO_x EFs halves each year, consistent with the limited potential for continued reductions. By contrast, CO_2 EFs are assumed to remain constant over time, as they are primarily influenced by fuel type and combustion conditions (Cheng et al., 2021).

Table S1. Data sources used in the bottom-up estimates.

Steps	Corresponding MEIC sector	Adopted data	Data source
Monthly emission estimation*	Power	Thermal power generation	National Bureau of Statistics (https://data.stats.gov.cn/english/easyquery.htm?cn=C01)
	Cement	Cement production	
	Iron	Iron production	
	Other industry	Manufacturing value added	
	On-road	Road Freight turnover	
	Off-road	Construction area	
Dissolving monthly emissions into daily	Residential/ Residential-bio	Population-weighted heating degree day	Calculation based on the 2m temperature data from the ERA5 dataset
	Power/ Cement/ Other industry	Coal consumption	(Wu et al., 2022)
	Iron	Operating rates of electric furnace	The custeel database (https://www.custeel.com/)
	On-road/ Off-road	Baidu migration data	The Baidu database (https://qianxi.baidu.com/)

*Production index are used to differentiate January and February from the combined first two months' data in the National Bureau of Statistics.

11. The source of the 40% reduction is confusing (Lines 105-106). Only after reading the rest of the paper did I realize that this was from one of the sensitivity tests. (Again, the authors need to focus on the logical flow of information in the text.)

Response:

The 40% reduction in simulation is used to quantify the response of NO_2 concentration to the changes in anthropogenic NO_x emission (β), building on previous works. In our previous tests, this perturbation magnitude seems to have a limited impact on final estimates within the tested range of 30-50%. We have added a brief explanation in Lines 110-112 in Manuscript. Besides, we have made adjustments in Methods to clarify the logical flow, please refer to the response to Comment 4.

Lines 110-112: "The 40% reduction was selected after a series of sensitivity tests, which demonstrated that this perturbation level exerts a limited impact on the β estimates (Zheng et al., 2020)."

12. Section 2.2.1 does not mention the spatial resolution of the inventory.

Response:

The original MEIC inventory has a resolution of 0.25°×0.25°, which we aggregate to 0.5°×0.625° to align with the resolution of the prior and the GEOS-Chem model. We have added this explanation in Lines 94-96 and Lines 165-166.

Lines 94-96: "Notably, to reconcile the resolution between the prior emissions and the model, we aggregated the original MEIC emissions from a resolution of 0.25°×0.25° (Fig. S2) to 0.5°×0.625°."

Lines 165-166: "The prior provides the sectoral profile for subsequent emission attribution. We conducted a comprehensive examination of associated parameters when updating the prior from 2019 MEIC (0.5°×0.625°),"

13. In Line 172, consider changing "policies" to "protocols". The use of "policies" has political connotations.

Response:

We have changed the "policies" to "protocols" in Line 206 (original 172) as suggested.

14. In Line 245, add "the" before "tests' impact".

Response:

We have added "the" in Line 278 (original 245) as suggested.

15. From Line 252, "A reduction in NO_x increases rNO_x". Why is this the case? I do not follow.

Response:

rNO_x represents the reduction ratio of NO_x emission factors (EFs); thus, a greater reduction in NO_x EFs corresponds to a higher rNO_x value. We have explained this parameter in Line 143.

Line 143: "$rNO_{x\ s,i,y}$ is the reduction ratio in NO_x EFs by sector from 2019 to 2022 derived from the bottom-up estimation."

16. In Line 273, I think "parameters" should be singular: "parameter".

Response:

We have corrected the "parameters" to "parameter" in Line 306 (original 273) as suggested.

17. In Line 307, “mode” should be “model”.

Response:

We have changed the “mode” to “model” in Line 340 (original 307) as suggested.

18. How are the cities arranged in Figure 5? Are they arranged by longitude?

Response:

The original arrangement was based on the IDs of China's provinces. We have now modified it to follow an area-based sequence, as the area is one of the key factors influencing regional emission estimates in this methodology.

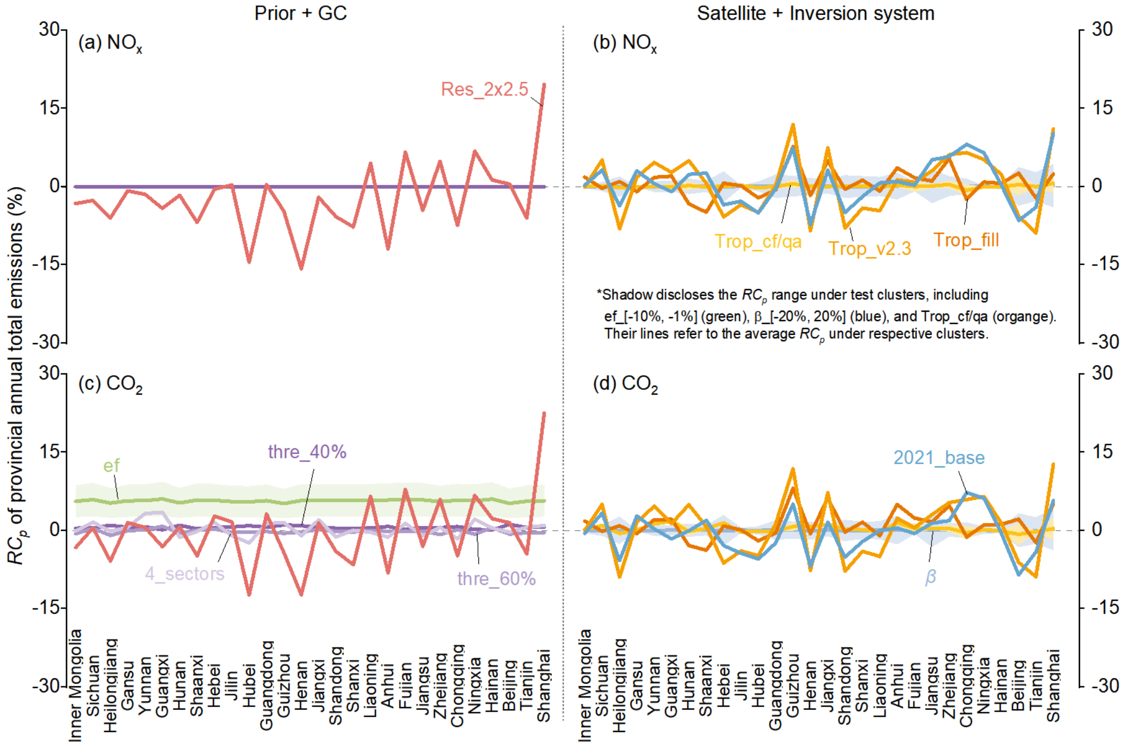

Figure 5. Response of provincial annual total NO_x and CO_2 emissions to different tests. (**a**) and (**b**) show RC_p of NO_x emissions incurred by tests. (**c**) and (**d**) are plotted for CO_2 emission as (**a**) and (**b**). Lines refer to the RC_p caused by the corresponding test or the averaged RC_p caused by corresponding test clusters (ef_[-10%, -1%] and β_[-20, 20%]), and the shadow refers to the RC_p range in test clusters. Only provinces with enough TROPOMI observations are shown here (i.e., grids with NO_2 TVCDs larger than 1×10^{15} molecules/cm^2 cover more than 90% of anthropogenic NO_x emissions within provinces). The provinces are arranged by area.

Reference:

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