Response to Referee #2

We would like to thank reviewer #2 for taking the time to review this manuscript and for providing valuable, constructive feedback and corresponding suggestions that helped us to further improve the manuscript.

In this author's comment, all the points raised by the reviewer are copied here one by one and shown in blue color, along with the corresponding reply from the authors in black.

This article provides coal mine methane emission estimates for three regions of the Shanxi province (China), using the recently-developed wind-assigned anomaly method with methane concentration observations from the TROPOMI satellite instrument. The results suggest that commonly-used emission inventories overestimate coal mine emissions in the area. The sensitivities of wind-assigned anomaly results to several of the method inputs and parameters are described.

The article is concise (too much actually, see below) and its English reads well. I think it is a relevant addition to the literature because (1) it confirms previous results on Chinese coal mine methane emissions with an original method; and (2) it builds more confidence and understanding of the wind-assigned anomaly method and its sensitivities.

I recommend the publication in ACP once all the following comments are addressed.

We thank the reviewer for this positive statement.

Significant comment on structure, method and data description, and naming

While concision is indeed a quality when writing a scientific article, the authors must be careful to provide enough information so that it can still be read as a standalone piece. In its current state, this article cuts too many corners in describing their datasets and methods to be read smoothly and requires, on this matter, a significant adjustment.

We have added more information in the manuscript as suggested by both referees.

Data set descriptions

The text keeps referring to "the bottom-up inventory" (in the abstract !!, line 20, and at lines 141, 147, 166, 171, 183, 228, 232, 236, 258 and 275) which is different from commonly-used EDGAR or CAMS-GLOB-ANT, but without ever properly presenting this different inventory. The reader has only the captions of Figures 2 or 4 to rely on to guess that "the bottom-up inventory" is actually work by Qin et al. (2023). Considering that the Qin et al. (2023) bottom-up inventory is a significant discussion reference, it needs to be clearly presented in the abstract, and presented and described in the main text.

For clarity, I would suggest to gather the descriptions of all three emission inventories (Qin et al. (2023), EDGAR and CAMS-GLOB-ANT) in a dedicated subsection of Section 2 "Data and method".

In addition, regarding naming, the expression "the bottom-up inventory" which is repeatedly used to refer to Qin et al. (2023) may be confusing to some readers as EDGAR and CAMS-GLOB-ANT can also be understood and referred-to as bottom-up inventories (e.g. Janssens- Maenhout et al., 2019). I would suggest to use the actual citation or a defined abbreviation/ acronym to refer to the Qin et al. (2023) bottom-up inventory in the text, and in Figures captions and labels.

Thanks to the referee for this suggestion. We have incorporated additional details about the "bottom-up inventory" in the abstract. Furthermore, we have introduced an additional subsection in the "Data and method" section to comprehensively describe all three emission inventories as suggested by the referee. In order to differentiate the presently mentioned "bottom-up inventory" from CAMS-GLOB-ANT and EDGAR, we have revised the name to "IPCC Tier 2 bottom-up inventory (Qin et al., 2023)". The figures are updated accordingly.

Method description

Subsection 2.2 named "Wind-assigned anomaly method", only briefly provides the Tu et al. (2022a) reference that actually gives the description of the wind-assigned anomaly method, and then just details the approximate "cone plume" model.

These few elements are insufficient for a standalone reading and understanding of the work performed in this study. While it is unnecessary to reproduce all the description and equations provided in the main text and appendices by Tu et al. (2022a), a 1-2 paragraph digest description of how the method works is at least expected.

The reading would be greatly improved if such a 1-2 paragraph digest description of the wind-assigned anomaly would mention and provide the minimally-required information on: (1) the background estimation and removal; (2) the principles of averaging TROPOMI data for two different wind field segmentations and making the difference of those averages; (3) the fitting of modelled against observed wind-assigned anomalies to estimate an emission scaling factor;and (4) uncertainty bar calculation.

We have added all information related with the wind-assigned method in Subsection 2.3-Dispersion model and 2.4-Background removal and wind-assigned anomaly method, as suggested by the referee.

Besides, the discussion of using a Gaussian plume instead of a cone plume model included in this work shows that the cone plume model, in itself, is not essential to the wind-assigned anomaly method. For clarity, I would thus suggest to separate the descriptions of the wind-assigned anomaly method principles and approximate plume models in different sub(- sub?)sections, and so also move the description of the Gaussian plume model from Sect.3 to somewhere in Sect 2.

Two separated subsections about plume models (cone plume and Gaussian plume) and the wind-assigned anomaly method have been added in Sect. 2.3 and 2.4, as suggested by the referee.

Significant comments on Results and discussion

The Results and discussion section can be improved on three different aspects, detailed below.

Discussion on elevation features, approximate plume models and their opening-angle parameters

To my understanding, this application of the wind-assigned anomaly method in Shanxi brings a new additional interesting aspect that is not currently discussed in the paper. Works by Tu et al. (2022a,b) previously studied locations where methane can be transported in plumes, along elevation features. However, this new study area in Shanxi has elevation features all around the target sources, methane is being blown against these elevation features and piles up at the bottom of valleys sources.

I would expect that approximate plume models struggle more to reproduce realistic enhancements in mountainous areas with complex elevation features such as Shanxi, compared to flatter terrains like the ones near Madrid or around the Polish coal mines. Interestingly, the discussion in Sect 3.4.1 shows for Changzhi region that changing the approximate plume model from cone to Gaussian improves the wind-assigned anomaly result comparison to Qin et al. (2023). Furthermore, it also shows that increasing the opening angle of these approximation models from 60° upwards improves the comparison even more.

Does that also hold for Jincheng and Yangquan regions? Could the Gaussian plume model with wider opening angles, to some extent, be more appropriate to approximate transport in such mountainous areas with complex elevation features compared to the cone model with the lower opening angle of 60°? What could be explored to test such an hypothesis? Could test experiments with N2O help, like what was done near Madrid in Tu et al. (2022a) to show that wind-assigned anomaly works?

I do not obviously expect that all these questions will be precisely answered after completing the review process. However, I think that additional discussion elements on the appropriateness of different approximate plume models and/or of their opening angle parameter values, possibly in relation with complex elevation features in Shanxi, can be an interesting and valuable addition.

Thanks to the referee for suggesting a valuable approach to identify optimal opening angles. We have tested experiments with TROPOMI NO_2 in the study regions. However, the complexity of the spatial distribution of NO_2 sources in these regions became apparent. The NO_2 sources are in the Changzhi region (see Figure 1-left) differ significantly from those in Madrid, where the dominant sources are concentrated in the city center. Notably, the wind-assigned anomalies (i.e., the difference in the TROPOMI tropospheric NO_2 concentration under NW and SE wind regimes) does not exhibit a distinct bipolar plume (see Figure 1-right). Thus, the NO_2 test experiment unfortunately proves ineffective in the study area.

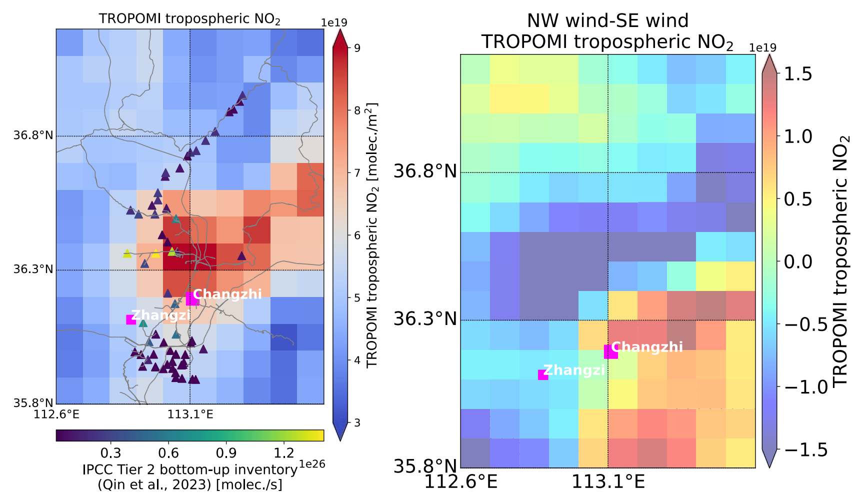


Figure 1: (left) spatial distribution of TROPOMI tropospheric NO_2, (right) wind-assigned anomalies (NW – SE) based on TROPOMI tropospheric NO_2 in Changzhi region.

Elevated concentrations of XCH_4 are notably centered in the heart of the Changzhi region, with slightly lower values observed in the southern areas, where coal mines are clustered (Figure 2 left). Taking into consideration the elevation features, the central part of the Changzhi region is characterized by flat terrain, while elevations rise in the northeast and southeast, as depicted in Figure 2 on the right. Thus, we expect that CH_4 does not accumulate at the bottom of valleys but tends to distribute across the entire flat terrain. The orientation of the mountains supports certain prevailing wind patterns in the study area, which is reflected by our segmentation choice.

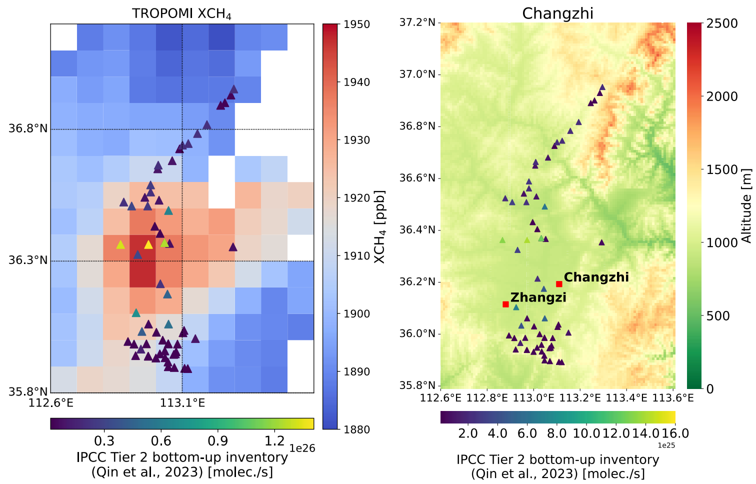


Figure 2: spatial distribution of XCH$_4$ (left) and altitude (right) from TROPOMI observations.

The impact of two distinct dispersion models (Gaussian plume and cone plume) on the estimated emission rates in distinct study regions is depicted in Figure 3. Notably, estimates show an upward trend with higher opening angle (fov) values for both models. Furthermore, the difference in estimates between the two models become more pronounced as fov values increase. The estimated emission occurs closest to the bottom-up inventory for higher fov in Changzhi region (i.e., 80º), while a closer match is observed for lower fov values in the Jincheng and Yangquan regions (i.e., 20º).

It appears difficult to decide whether using wider opening angles for approximating in such mountainous areas is the superior choice. Instead, it is more appropriate to view the opening angle as a contributing factor to uncertainty in estimating emission rates.

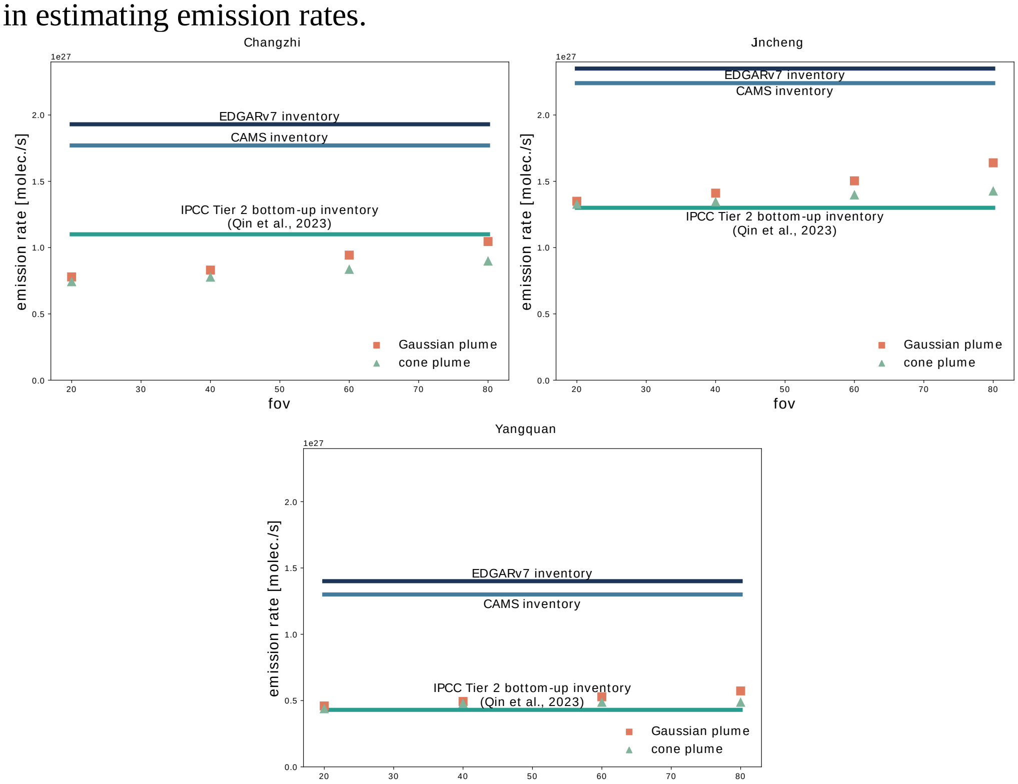


Figure 3: Estimates of emission rates in Changzhi, Jincheng and Yangquan regions with respect to different opening angles based on cone plume and Gaussian plume. The three different inventories are presented as well.

To further investigate the wind pattern on the complex terrain, we subdivide the study area in the Changzhi region into three subregions (specifically, areas between 35.8 º – 36.3º N, 36.3 º – 36.8º N, 36.8 º – 37.2º N). The corresponding wind rose plots are illustrated in Figure 4. The wind patterns in the southern and central subregions, where most coal mines are located, exhibit a similar pattern, whereas the northern subregion tends to feature more wind from the NW direction and less from the SE direction. Thus, the wind distribution appears generally homogenous in areas with dominant emission sources, while the complex terrain demonstrates a more pronounced impact on the northern region where less coal mines are located.

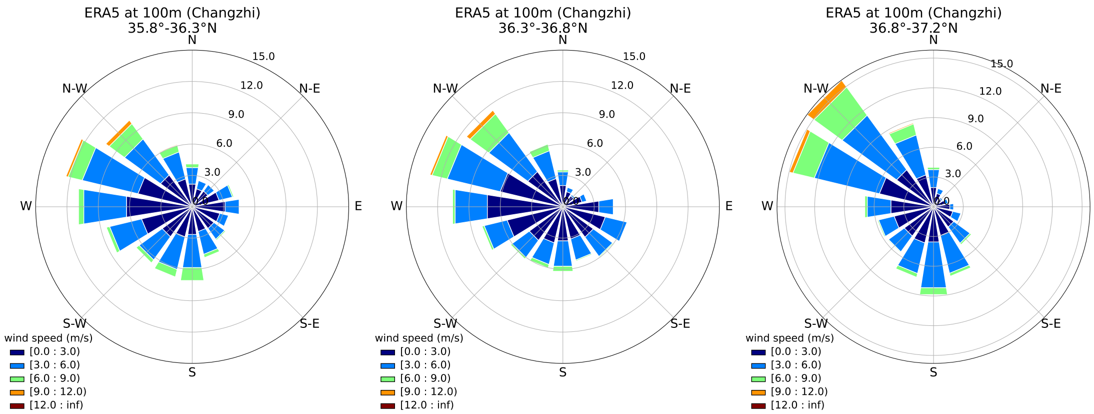


Figure 4: Wind roses plots for local daytime (08:00–18:00 UTC) from May 2018 to April 2023 for the ERA5 model wind for three subregions in Changzhi. The region range is given in the title of each subfigure.

To further investigate the sensitivity of the wind spatial variation, the wind data at the central point (36.5º N, 113º E for Changzhi, 35.5º N, 112.75º E for Jincheng and 38º N, 113.5º E in Yangquan) is used as a representative value to represent the wind for the entire study area. This substitution results in a decreased estimated emission rate of 11% (emission rate: 7.5×10^{26} molec. s^{-1}) in Changzhi, an increased emission rate of 7% (1.5×10^{27} molec. s^{-1}) in Jincheng and of 8% (5.3×10^{26} molec. s^{-1}) in Yangquan. The discussion of this uncertainty has been included in Subsection 3.3.3 in the revised manuscript.

Revising uncertainty estimates to account for method-related errors

Currently, the uncertainty estimates, which seem to correspond to the error bars in Figure 6, amount to only a few percents of the total emission estimates: 1.9%, 1.4% and 3.7% for Changzhi, Jincheng and Yangquan regions, respectively. From Sect. 2 (see significant comment on method description above), I can only guess that these uncertainty estimates include the contributions of background estimation error and satellite data noise, as performed in Tu et al. (2022a,b).

Thanks to the referee for emphasizing the uncertainty issues. The current uncertainty, as mentioned by the referee, includes only the contributions of background estimation error and satellite data noise. In response to this concern, we have expanded the discussion on additional sources of uncertainty in the revised manuscript (see section 3.3) and have accordingly update the total uncertainties.

In addition, Sect. 3.4 discusses the impact of (1) changing the approximate plume model to the Gaussian plume model, and perturbing the opening angle; (2) changing the wind product from ERA5 to NCEP, and changing the direction segmentation; and (3) changing the a priori inventories. Besides, other parameters may influence the results as well, such as the height of the wind speed, as discussed in Tu et al. (2022b): why is 100m chosen in this work, whereas 10m was used for Madrid area in Tu et al. (2022a), and 330m for Poland, in Tu et al. (2022b)?

Concerning wind at different height levels, we employed wind data at 10 m for the Madrid area due to the availability of meteorological station data measuring wind at this height. The in-situ measured wind at 10 m was employed together with ground-based measurements (EM27/SUN) to estimate local emission rates.

In the Poland study, we firstly used XCH_4 from the CAMS and its corresponding emissions to assess the wind-assigned anomaly method. Because the study area was larger (so we expect more vertical mixing), we assumed that using winds at a higher level of 330 m would provide a superior description of the transport on larger scales. Sensitivity analyses investigating the uncertainty connected to the altitude choice were conducted in both studies.

Overall, the sensitivity tests reported here result in emission rates changing from -5% up to +12%, which is larger in magnitude than the maximum of the currently reported uncertainties. As the choice of many method inputs and parameters can be somewhat arbitrary (ERA5 winds against NCEP, Gaussian against cone plume, 60° against 80° fov, wind speed height, etc), the uncertainty estimates provided in this work need to be revised to account for the contribution of these method-related choices and uncertainties.

For example, ways to account for these method-related uncertainties can be through the definition of a comprehensive quantification ensemble, which explores reasonable ranges for different method inputs and parameter values, such as done by e.g. Schuit et al. (2023), or to sum different contributions in quadrature, such as done by e.g. Cusworth et al. (2021).

Revised uncertainties are expected to be higher. However, these larger uncertainties may actually help to better compare with emission rates reported by Qin et al. (2023), and to assess how significant the difference found between EDGAR/CAMS-GLOB-ANT and wind-assigned anomaly results is.

Thanks to the referee for this important comment. The model and the input data represent the primary contributors to uncertainties. To encompass a comprehensive understanding of these uncertainties, we divided our analysis into four key components in the revised manuscript: 1) background removal (10^{th} lower percentile of overall satellite observations each day as the background); 2) dispersion model, including Gaussian against cone plume and variations in fov; 3) wind information, covering wind at different heights, wind data from different sources, wind segmentation and spatial variations; and 4) inventory, serving as the a priori knowledge (CAMS-GLOB-ANT inventory against the IPCC Tier 2 bottom-up inventory (Qin et al., 2023)).

The total uncertainty is computed through error propagation, similar to the approach of summing contributions in quadrature as done by Cusworth et al. (2021). This computation yields a total uncertainty of 25% in Changzhi, 20% in Jincheng and 21% in Yangquan.

Discussion of results against previous satellite-based top-down estimates

Satellite-based estimates of methane emissions are currently being studied and developed at different scales, with different datasets and different methods, by several groups across the world. For example, Chen et al. (2022) report a downward correction of coal mine emissions in China compared to UNFCCC reports, partly driven by Shanxi; or Zhang et al. (2021) report a 30% decrease in their posterior estimates for China, 60% of which are attributed to coal mines,and provide an extensive list of other studies supporting a consistent result.

These previous studies and/or others need to be mentioned in this article. They could for example be included in the Introduction, and their relevant messages cited and discussed when presenting the wind-assigned anomaly results. As they give a similar picture of overestimated Shanxi coal mine methane emissions in EDGAR/CAMS-GLOB-ANT, the overall article message would be even more highlighted, while at the same time building more confidence in the wind-assigned anomaly method.

Thanks to the referee for bringing this valuable information to our attention. We have added related information in the introduction and the results and discussion part as recommended by the referee.

> In introduction:
> "Qu et al. (2021) highlighted significant challenges in their satellite inversion over southeast China characterized by elevated seasonal rice emissions that coincide with extensive cloud cover and potential misallocation of coal emission. A recent study from Chen et al. (2022) reported a downward correction in CMM emissions (-15%) in China compared to the United Nations Framework Convention on Climate Change (UNFCCC) reports, partly driven by Shanxi. Zhang et al. (2021) documented an overestimation of anthropogenic emissions from China, revealing a 30% decrease in the posterior estimates, with approximately 60% of this downward correction attributed to coal mining."
>
> In Results and discussion:
> "Our CMM estimates in these three regions fall within the 30th and 70th percentile range of the updated emission rates in the study by Qin et al. (2023). In addition, our results are consistently lower than the CAMS-GLOB-ANT and the EDGARv7 inventories. This result agrees with previous studies. For instance, a -15% underestimation compared to the UNFCCC has been reported by Chen et al. (2022). Additionally, Zhang et al. (2021) documented a 30% decrease in their posterior estimates for China, with 60% attributed to coal mining. This pattern of overestimation in anthropogenic emissions, in comparison to China's inventory, has been observed in previous research, utilizing GOSAT inversion and various versions of the EDGAR inventory as a priori estimates (Miller et al., 2019; Maasakkers et al., 2019). This divergence may be attributed to two reasons: (1) missing observation of strong CMM emissions during the TROPOMI overpass. It is important to note that CMM emissions exhibit a strong dependency on coal mine activities, which vary over time. The TROPOMI data provide instantaneous observations, capturing CH_4 concentrations at a specific moment (local time ~ 13:30), thereby leading to limitations in detecting strong CMM emissions during both morning and afternoon periods. (2) the CMM utilization have been largely improved in the last decade, since the national government issued specific targets in the national 12th and 13th five-year plan (Gao et al., 2021; Lu et al., 2021)."

Minor corrections and questions

- Section 2.1 "TROPOMI dataset": Please provide the data quality filters applied to select the TROPOMI data included in this work.

Thank you. The information has been added.
"A data quality filter (qa = 1.0) is applied to characterize the data during clear-sky and low-cloud atmospheric conditions."

- Line 148: Please delete the "latest" adjective for EDGAR v7, as EDGAR v8 has just been released (https://edgar.jrc.ec.europa.eu/dataset_ghg80).

Thank you for providing this information. Corrected.

- Figures 4, 6 A-3 and A-5: Please use a consistent label to designate emissions from Qin et al. (2023), “bottom-up inventory” on one side, and “shaft emission” on the other.

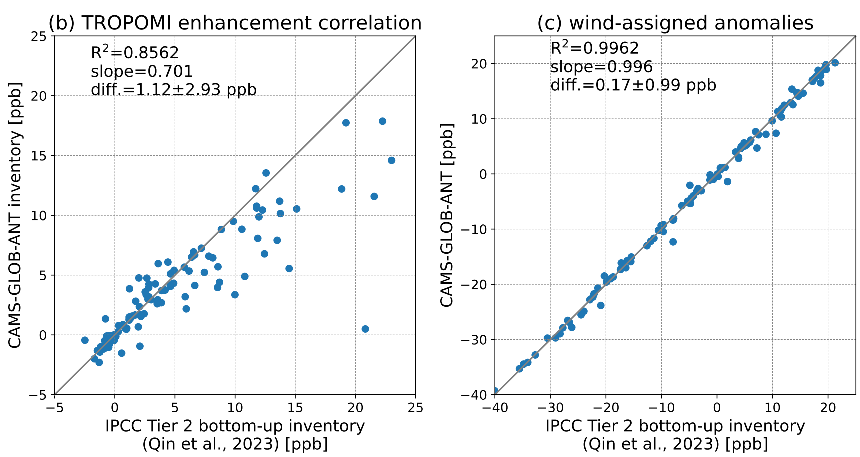
Thank you. Corrected.

- Figure 7: Please start the y-axis range to 0 in order to facilitate comparison with Figure6.

Thank you. The figure has been updated.

- Sect 3.4.3: It is unclear whether the “calculated average” (line 231) refers to simulated or observed averaged enhancement, please precise. If it is simulated, the fact that enhancements are lower with CAMS-GLOB-ANT whereas this inventory prescribes nearly twice as high emissions is quite counter-intuitive and surprising. Is this explained by the sentence lines 234-235 about similar background estimation errors that compensate in the wind-assigned difference? If so, could you please reformulatethis explanation and move it a few lines earlier in the text, when “calculated average”values are compared?

The “calculated average” refers to the observed enhancement (i.e., TROPOMI XCH_4 – background). This information has been detailed in the text. The a-priori inventory information, including the location and emission rates of the sources, has a small impact on the estimation of the background, as illustrated in the left figure below. The difference (1.12 ± 2.93 ppb, R^2 = 0.8562) arising from the use of different inventories as the a priori, is effectively mitigated (R^2 = 0.9962) when comparing the wind-assigned anomalies, as illustrated in the right figure below. It is because the systematic errors in background removal is compensated by computing the differences of enhancements under different wind field segmentations. These figures are presented in Figure A- 14 in the updated manuscript.



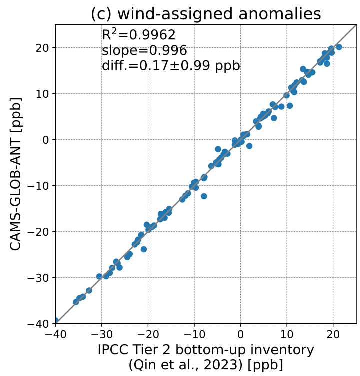
- Line 236: I think there is a typo, isn’t 8.5 x 10^27 supposed to be 8.5 x 10^26 instead? Otherwise, it would mean an order of magnitude difference…

Thank you. Corrected.

- Figure A-10: Are marker supposed to be missing in the scatter plot (right panel). If not,can you please add them, otherwise explain why there is no marker in the scatter plot?

We appreciated the referee for bringing this to our attention. In addition to the missing marker, we have also recognized that the current correlation figure, similar to the Figure 5(c) (or Figure A-6) in the manuscript, might raise confusion for readers.

The existing right figure differs from Figure 5(c) in the manuscript, which illustrates anomalies derived from the cone plume model and TROPOMI observations. It presents the correlation of modelled wind-assigned anomalies using different inventories, i.e., the bottom-up inventory from Qin et al. (2023) and the CAMS-GLOB-ANT inventory. It aims to display how different inventories affect anomalies based on the cone plume model. To avoid potential confusion for readers, the correlation plot, featuring different inventories, is presented in a distinct plotting style and as a subfigure in Figure A-14.



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