Response to Referee #1

We would like to thank reviewer #1 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.

Tu et al. present an analysis of TROPOMI methane observations over the coal-rich Shanxi province of China. They use their wind-assigned anomaly method to quantify regional methane emissions for three clusters of Shanxi coal mines. They compare their estimates with three bottom-up emission inventories, EDGAR v7.0, CAMS-GLOB-ANT, and a measurement-based coal mine methane inventory by Qin et al. (2023). They find good agreement with the Qin et al. estimates but much lower emissions (~factor of 2-3) than reported in the EDGAR and CAMS inventories.

The paper is interesting and a good fit for ACP, but in my view substantial changes are needed before it can be published. I see two major weaknesses. First, the methods need to be explained in much more detail, not merely by pointing to previous publications. I found it very difficult to follow the discussion of results because the paper does not adequately explain the wind-assigned anomaly method and its interpretation. Second, the uncertainty analysis appears to be incomplete. The authors report uncertainties <4% (<2% in two of three cases) on their regional methane emission estimates inferred from TROPOMI. These values are unrealistically low; regional emission errors reported elsewhere in the literature are routinely in the range ~20%-30%. I am therefore left with the impression that the authors have overlooked important sources of error, for example having to do with background subtraction and wind speed.

We appreciate that the referee provided us these valuable comments. With the assistance of these comments, we have tried to improve the manuscript accordingly.

More information has been added in the manuscript as suggested by both referees. The dispersion model and the wind-assigned anomaly method have been explained in detail in Section 2. To address concerns related to uncertainty analysis, we have not only considered background estimation error and satellite data noise, but have also discussed the uncertainties associated with the dispersion model and its inputs. The new derived errors are in the range of 20%-25%. These two issues will be discussed in detail below.

Specific comments

- L. 18-19: The reported uncertainties (<4%) are unrealistically small. Uncertainty in regional emissions derived from TROPOMI tend to be in the 20%-30% range (or more). There must be other, larger sources of error besides what is reported.
- Shen et al. (2022) used TROPOMI to estimate methane emissions for ~20 US oil and gas basins and reported mean errors of 30% based on an elaborate uncertainty analysis (https://acp.copernicus.org/articles/22/11203/2022/). Error bars for emissions from individual countries estimated by Shen et al. (2023) are of similar magnitude

(https://www.nature.com/articles/s41467-023-40671-6). TROPOMI analyses by Cusworth et al. (2022), Chen et al. (2023), and many others found similar results.

The previously reported uncertainties included only the contributions from background estimation error and satellite data noise. In the revised version of the manuscript, we also discuss the uncertainties arising from the background removal, dispersion model (cone plume model or Gaussian plume model) and its inputs (wind at different height level, different wind segmentation, the spatial variation of the winds and inventories as the apriori knowledge) and we propagate these uncertainties into our emission estimates. As a result, the total uncertainties of our emission estimates are determined to be 25% in Changzhi, 20% in Jincheng and 21% in Yangquan.

- L. 20: Which bottom-up inventory?

The bottom-up inventory computed based on the IPCC Tier 2 approach from Qin et al. (2023). This information has been included in the abstract and further detailed information is now provided in an additional subsection (Section 2.2).

- L. 21-22: That may be, but it's not entirely clear given the unrealistically small uncertainties reported for the TROPOMI emission estimates.

We have revised the given uncertainties, previously derived only from background estimation error and satellite data noise, to now encompass the specific uncertainties, including those arising from the background removal method, dispersion model and its inputs originated from wind and a priori inventory. The updated uncertainties are computed based on the error propagation, considering all the impacts mentioned above.

- L. 23: How do the estimates help to develop climate mitigation strategies?

The term "develop" might not be a good fit here. We changed this phrase to "provide additional insights (eg. a more realistic approximation based on the measurement dataset) into CMM emissions mitigation".

- L. 31: Would it not be more accurate to say that China is "the leading emitter", rather than just "one of" them?

Methane exhibits a long atmospheric lifetime, showing its influence on a climatic scale rather than an annual one. Meanwhile, certain emission sources, such as those originating from the military, are presently excluded from consideration. Therefore, we suggest the term "one of the leading CH_4 emitters" to more accurately convey the significant impact of methane emissions in the broader context of climate implications, given its persistent nature and the exclusion of specific sources.

- L. 33: China did not sign the 2021 Global Methane Pledge, so for clarity it would be best to use another word besides "pledge" here.

The word has been replaced with "committed". China has also signed other agreements and this information has been added in the manuscript.

> "China has demonstrated its commitment to addressing CH_4 emissions by signing key international agreements such as the Kyoto Protocol in 1998 and the Paris Agreement in 2016, reflecting its dedication to global efforts in mitigating climate change. Additionally, in 2021, China committed to reduce CH_4 emissions under the Glasgow Agreement and intended to develop a comprehensive and ambitious National Action Plan with the goal of

achieving a substantial impact on methane emission control and reductions in the 2020s (USDoS, 2021)."

- L. 33-35: It would be useful to include a reference for the Glasgow Agreement. Perhaps something like this 2021 US State Department press release: https://www.state.gov/u-s-china-joint-glasgow-declaration-on-enhancing-climate-action-in-the-2020s/

Thank you. This reference has been added.

- L. 68: It's unclear what "solar radiation […] radiated from the Earth" means.

original sentence: "The instrument utilizes passive remote-sensing techniques to measure solar radiation reflected by and radiated from the Earth across the ultraviolet (UV), visible (VIS), near-infrared (NIR), and short-wave spectral (SWIR) bands (Veefkind et al., 2012)."

the sentence has been changed to "The instrument utilizes passive remote-sensing techniques to measure the backscattered solar radiation across the ultraviolet (UV), visible (VIS), near-infrared (NIR), and short-wave spectral (SWIR) bands (Veefkind et al., 2012)."

- L. 88: What wind speed is used? The speed at 10-m? 50-m? Something else?

Wind data at 100 m is used here. This information has been added and it is also mentioned in Section 3.3. Uncertainty of wind at different height, e.g., 10 m, has been also discussed as part of the error analysis.

- L. 95: Regions of China or of Shanxi?

Thank you. It should be "regions of Shanxi". This has been corrected.

- Figure 1: Suggest increasing font size for legibility.

Thank you. The figure has been updated.

- L. 104-105: What are those estimates by Qin et al. (2023) based on? A brief description of the dataset would be valuable.

Thank you. The description of the dataset has been added in Section 2.2.

> "Qin et al. (2023) used both public and private datasets from over 600 individual coal mines in Shanxi Province. The IPCC Tier 2 approach is applied to calculate the corresponding CH_4 emissions based on 3-5 sets of observed emission factors, thereby establishing a range of bottom-up estimation of CMM on a mine-by-mine basis. In the following work, the bottom-up inventory computed from the median emission factors (E5) will serve as a prior information in the wind-assigned method for estimating emissions, referring to IPCC Tier 2 bottom-up inventory. In their study, an eddy-covariance tower was installed in Changzhi during two two-month periods to derive an average observed CH_4 flux. Based on the in-situ measurements, a series of scaling factors at different percentiles of the observational distribution (i.e., 10%, 30%, 50%, 70%, 90%) were generated. These scaling factors were subsequently employed to update the preliminary Tier 2 bottom-up inventory (Qin et al., 2023). The scaling factors for a specific percentile of the observational distribution show minimal variations among different coal mines, suggesting these factors can be treated as constant values across the ensemble of coal mines at each percentile. Our wind-assigned method emphasizes the proportional share of emissions per mine rather than absolute values, resulting in estimated CMM emissions that do not significantly differ whether using the Tier 2 bottom-up inventory or one of the scaled inventory datasets. In additional to the current

IPCC 2 Tier bottom-up inventory, the scaled inventory is also provided as an additional reference point in this work."

- L. 105-106: I do not understand the sentence beginning "Near 30 small coal mines…"

original sentence: "Near 30 small coal mines scatter in the mountain area in the south and the emissions are relatively small with 24 orders of magnitude in molec. s^{-1}".

The coal mines in the south of Zhangzi emit relatively smaller CH_4 than the others. There are approximately 30 small coal mines scattered in the mountainous area in the south. The emissions from these mines are relatively small, with values less than 1×10^{25} molec. s^{-1} per mine.

The sentence has been rephrased in the manuscript to "There are near 30 small coal mines scattered in the mountain area in the south and each mine has a relatively low emission rate, measuring less than 1.0×10^{25} molec. s^{-1}."

- L. 110: Qin et al. (2023) used eddy covariance measurements to construct their facility-scale inventory. Would it be appropriate to describe their work as a measurement-based inventory of coal mine emissions?

The referee correctly points out that the final inventory in Qin et al. (2023) uses a mixture of bottom-up and top-down approaches, i.e., using the in-situ measurements to update the preliminary inventory derived from the IPCC Tier 2 approach. From these observations, a series of scaling factors at different percentiles of the observational distribution (i.e., 10%, 30%, 50%, 70%, 90%) were generated. The scaling factors for a specific percentile of the observational distribution show minimal variations among different coal mines, suggesting these factors can be treated as constant values across the ensemble of coal mines at each percentile. Our wind-assigned method emphasizes the proportional share of emissions per mine rather than absolute values, resulting in estimated CMM emissions that do not significantly differ whether using the Tier 2 bottom-up inventory or one of the scaled inventory datasets. In additional to the current IPCC 2 Tier bottom-up inventory, the scaled inventory is also provided as an additional reference point in this work.

We have added detailed information about the bottom-up inventory in Section 2.2. Additionally, to distinguish this bottom-up inventory with other inventories, like the scaled inventory in Qin et al. (2023), CAMS-GLOB-ANT or EDGARv7.0, we use "IPCC Tier 2 bottom-up inventory (Qin et al., 2023).

- Subsection 3.2: Suggest moving this subsection to section 2 (data and methods), including description of the Qin et al. (2023) dataset.

Thank you. We have moved the subsection 3.2 to Section 2 and added more information about the inventory from Qin et al. (2023) in the text as suggested by the referee.

"Qin et al. (2023) used both public and private datasets from over 600 individual coal mines in Shanxi Province. The IPCC Tier 2 approach is applied to calculate the corresponding CH_4 emissions based on 3-5 sets of observed emission factors, thereby establishing a range of bottom-up estimation of CMM on a mine-by-mine basis. In the following work, the bottom-up inventory computed from the median emission factors (E5) will serve as a prior information in the wind-assigned method for estimating emissions, referring to IPCC Tier 2 bottom-up inventory. In their study, an eddy-covariance tower was installed in Changzhi during two two-month periods to derive an average observed CH_4 flux. Based on the in-situ measurements, a series of scaling factors at different percentiles of the observational distribution (i.e., 10%, 30%, 50%, 70%, 90%) were generated. These scaling factors were subsequently employed to update the preliminary Tier 2 bottom-up inventory (Qin et al., 2023). The scaling factors for a specific percentile of the observational distribution show minimal variations among different coal mines, suggesting these factors can be treated as constant values across the ensemble of coal mines at each percentile. Our wind-assigned method emphasizes the proportional share of emissions per mine rather than absolute values, resulting in estimated CMM emissions that do not significantly differ whether using the Tier 2 bottom-up inventory or one of the scaled inventory datasets. In additional to the current IPCC 2 Tier bottom-up inventory, the scaled inventory is also provided as an additional reference point in this work."

- Figure 4 (left): A log y-scale would be helpful here.

The figure has been revised, so as the Figure A- 2 for EDGARv7 inventory.

- Section 3.3 and Fig. 5: Significantly more explanation is needed on how the wind-assigned anomalies are calculated and how the wind direction segmentation is performed and what these things mean. The methods section on the wind assigned anomaly method only describes the cone plume model. Section 3.3 is very difficult to follow, and readers shouldn't need to read the authors' previous papers to understand what is going on; the paper should be readable on its own. It's unclear to me what the middle panel of Fig. 5 is showing. How are the estimated emissions distributed between the different coal mines within a cluster? How are the emissions calculated from the TROPOMI wind-assigned anomalies? Are all the mines scaled up/down together following the spatial distribution of the underlying inventory?

Thanks to the referee for this comment. We have added additional details about the dispersion model and the wind-assigned anomaly method in Section 2.

The middle panel of Fig. 5 represents wind-assigned anomalies, representing the difference in TROPOMI enhancements between two wind segmentation. It is important to note that the estimated emission in this context is a total value for the entire study region, rather than a spatial distribution. The inventory, serving as a priori knowledge, provides the CMM emission fractions instead of their absolute values. Thus, as mentioned by the referee, all the mines can be collectively scaled up or down based on their emission share and spatial distribution in the underlying inventory.

- How is the TROPOMI methane background subtracted? Background subtraction tends to be a major source of error in regional emission estimation.

The background consists of a constant value, a temporal linear increase, a seasonal cycle, a daily signal, and a horizonal signal. This encompasses the consideration of both temporal and spatial variations in the background removal. The description of background removal has been added in Section 2.4.

"It is of importance to separate the increase of the atmospheric CH_4 concentration due to local emissions from the accumulated atmospheric CH_4 background concentration (the CH_4 atmospheric lifetime is in the order of 12 years). A Jacobian matrix is introduced to reconstruct the background according to a few background model coefficients, i.e., a constant CH_4 value and superimposed disturbances: a temporal linear increase, a seasonal cycle determined by the amplitude and phase of the three frequencies 1/year, 2/year and 3/year, a daily signal (same value for all data measured during a single day), and a horizonal gradient (same value for any time but dependent on the horizontal location) (Tu et al., 2022a). In the following discussion, the satellite enhancements refer to the residual signal as deduced from TROPOMI CH_4 observations after subtracting the modelled background (Figure 4 lower panel)."

The referee is right that the background subtraction can be a major source of error in emission estimation. To further study the impact of background subtraction on emission estimates, the 10th lower percentile of overall satellite observations each day is considered as an alternative choice for setting the background value for the study area on that day. The enhancements (TROPOMI XCH_4 - background) and the wind-assigned anomaly computed from the enhancements based on the 10th percentile are compared to those values based on the spatial and temporal variation (default calculation in this study), as presented below. The gridded enhancements computed from the 10th percentile show higher values (21.5 ppb in average) than those based on the spatial and temporal variation method. When comparing the resulting wind-assigned anomalies, an excellent correlation with a R^2 value of 0.98 and a slope near unity is found. The wind-assigned anomalies approach, which derives emissions from differences of XCH_4 observations associated with opposite wind orientations, helps to effectively mitigate systematic errors associated with background subtraction.

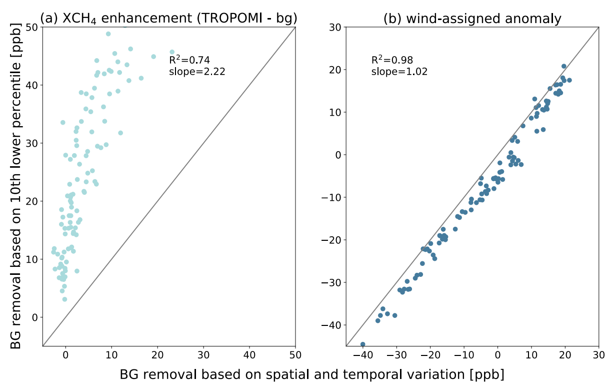
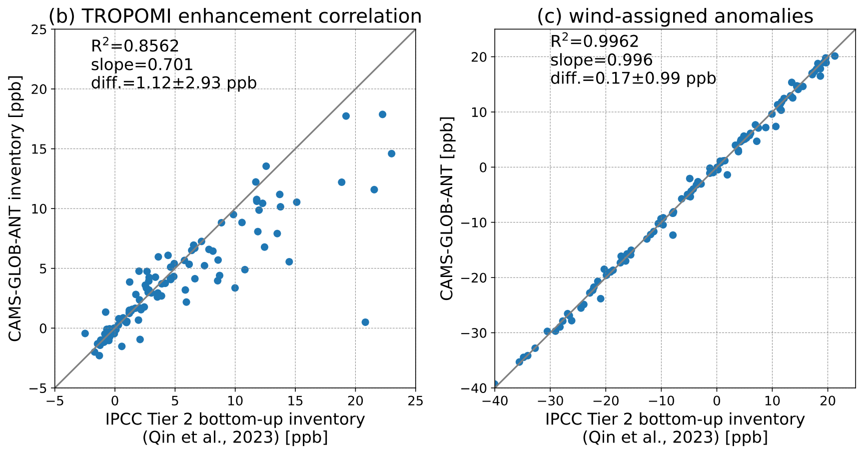


Figure 1: XCH_4 enhancements (TROPOMI - background) and its corresponding wind-assigned anomaly using different background removal methods. The grey line corresponds to the 1:1 line.

- L. 231: What "calculated average" value is being reported here? It's unclear what the ppb values refer to.

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.



- The uncertainty analysis varying the plume model, wind product, and inventory is a good start, but not sufficient. What is the sensitivity to background subtraction scheme? What about wind speed levels (e.g., using the 10 m wind rather than 100 m)?

To assess the impact of background removal sensitivity, the study now employs the 10^{th} lower percentile of overall satellite observations as alternative choice for the daily background in the study area, deviating from the approach outlined in Section 2.3, which separately considers spatial and temporal variations. The substitution of the background removal method results in a 7% increase in estimated emission rates in Changzhi, a 6% increase in Jincheng and a 9% increase in Yangquan.

Lower estimates are observed in three regions when employing the wind at 10 m. Specifically, there is a 12% decrease in estimation strength in Changzhi, 11% in Jincheng and 4% in Yangquan. These differences can be attributed to reduced wind speed at lower level, resulting in measured lower wind speed of 15% in Changzhi, 17% in Jincheng and 10% in Yangquan.

Both of these sensitivities are thoroughly addressed in the Uncertainty analysis (Section 3)

- How are the current error values calculated? Do they represent 1-sigma errors? Reporting the uncertainty as the range of estimates from a broader estimation ensemble might be clearer.

The current error values derived from the background removal and the satellite noise. To calculate the uncertainty of the background signal, we first compute the difference between the satellite observations ($\boldsymbol{y}$) and the modeled background ($\mathbf{K}^{*}{}_{\mathrm{BG}}\hat{\boldsymbol{x}}_{\mathrm{BG}}$), and then determine the mean square value ($\mathbf{S}_{\mathrm{y,BG}}$) from its elements representing observations unaffected by the plume. The uncertainty of the background model coefficients can be calculated as $\mathbf{S}_{\hat{\boldsymbol{x}}_{\mathrm{BG}}} = \mathbf{G}_{\mathrm{BG}}\mathbf{S}_{\mathrm{y,BG}}\mathbf{G}^{T}{}_{\mathrm{BG}}$. The $\mathbf{G}_{\mathrm{BG}}$ is the gain matrix.

The observed wind-assigned anomaly $\Delta\boldsymbol{y}_{\mathrm{plume}}$ is a column vector, obtained as the product of satellite signals $\boldsymbol{y}_{\mathrm{plume}}$ and the operate $\mathbf{D}$, which represents the binning, the averaging, the wind-assigned Δ-maps calculations and the data number filtering. The uncertainty covariance can be written as:

$$\Delta\mathbf{S}_{\mathrm{y,plume}} = \mathbf{D}\mathbf{S}_{\mathrm{y,plume}}\mathbf{D}^{T}$$

A Jacobian $\Delta\boldsymbol{k}$ represents the wind-assigned anomaly model, aiding in generation of the wind-assigned anomaly $\Delta\boldsymbol{y}_{\text{plume}}$, i.e., $\Delta\boldsymbol{y}_{\text{plume}} = \Delta\boldsymbol{k}\boldsymbol{x}$. Here, the coefficient x represents the scaling factors for adjusting the a priori emission rates to achieve the best agreement with the observed plume. Thus, a row vector can be derived as:

$$\boldsymbol{g}^{\boldsymbol{T}} = (\Delta\boldsymbol{k}^T \Delta\mathbf{S}^{-1}{}_{\text{y,plume}} \Delta\boldsymbol{k})^{-1} \Delta\boldsymbol{k}^T \Delta\mathbf{S}^{-1}{}_{\text{y,plume}}$$

The background uncertainty ($\boldsymbol{\epsilon_{BG}}$) and the noise in the satellite data ($\boldsymbol{\epsilon_n}$) can be estimated as:

$$\boldsymbol{\epsilon_{BG}} = \sqrt{\boldsymbol{g}^T \mathbf{D}\mathbf{K}_{\text{BG}} \mathbf{S}_{\hat{\text{x}}_{\text{BG}}} \mathbf{K}^T{}_{BG} \mathbf{D}^T \boldsymbol{g}}$$

$$\boldsymbol{\epsilon_n} = \sqrt{\boldsymbol{g}^{\boldsymbol{T}} \mathbf{D}\mathbf{S}_{y,n} \mathbf{D}^T \boldsymbol{g}}$$

The uncertainties in the updated manuscript include additional errors introduced by the dispersion model and its input data through the error propagation. This information has been updated in the conclusion section.

- L. 246-250: These sentences are contradictory. Should the first sentence only refer to the “bottom-up” inventory (which, again, does not seem to me to be a “bottom-up” inventory – rather a measurement-based inventory).

Thanks to the referee. The sentences have been modified.

> “The estimates obtained derived through the wind-assigned anomaly method demonstrate comparability with the IPCC Tier 2 bottom-up inventory (Qin et al., 2023). Compared to the estimates, the inventory shows relative differences of 31%, -7%, and -12% in Changzhi, Jincheng, and Yangquan, respectively.”

Typos

- 14: “process” → “progress” ?

corrected.

- 27: “emission” → “emissions”

corrected.

- 51: “emissions” → “emission estimates”

corrected.

- 53: “from satellite” → “from satellites”

corrected.

- 70: “unprecedented high spatial resolution…” → “unprecedented combination of high spatial resolution…”

corrected.

- 6: “bottum” → “bottom”

The typo in the legend of fig. 6 has been corrected.

- 243: “achived” → “achieved” ?

corrected.

- 252: “boarded” → “bordered” ?

corrected.

References

Chen, Z., Jacob, D. J., Gautam, R., Omara, M., Stavins, R. N., Stowe, R. C., Nesser, H., Sulprizio, M. P., Lorente, A., Varon, D. J., Lu, X., Shen, L., Qu, Z., Pendergrass, D. C., and Hancock, S.: Satellite quantification of methane emissions and oil–gas methane intensities from individual countries in the Middle East and North Africa: implications for climate action, Atmos. Chem. Phys., 23, 5945–5967, https://doi.org/10.5194/acp-23-5945-2023, 2023.

Cusworth, D. H., Thorpe, A. K., Ayasse, A. K., Stepp, D., Heckler, J., Asner, G. P., Miller, C. E., Chapman, J. W., Eastwood, M. L., Green, R. O., Hmiel, B., Lyon, D., and Duren, R. M.: Strong methane point sources contribute a disproportionate fraction of total emissions across multiple basins in the U.S., Earth ArXiv, https://doi.org/10.31223/X53P88, 2022.

Shen, L., Gautam, R., Omara, M., Zavala-Araiza, D., Maasakkers, J. D., Scarpelli, T. R., Lorente, A., Lyon, D., Sheng, J., Varon, D. J., Nesser, H., Qu, Z., Lu, X., Sulprizio, M. P., Hamburg, S. P., and Jacob, D. J.: Satellite quantification of oil and natural gas methane emissions in the US and Canada including contributions from individual basins, Atmos. Chem. Phys., 22, 11203–11215, https://doi.org/10.5194/acp-22-11203-2022, 2022.

Shen, L., Jacob, D.J., Gautam, R. et al. National quantifications of methane emissions from fuel exploitation using high resolution inversions of satellite observations. Nat Commun 14, 4948 (2023). https://doi.org/10.1038/s41467-023-40671-6