

Dear Editor and Reviewers,

We appreciate all of the time you have provided to us, as well as your insightful and valuable comments and recommendations. This process has helped us improve our research, and I am confident that the revised version of the manuscript will address all the concerns and queries. A concerted effort has been made to address all points raised in full. To facilitate the response, the unaltered original remarks are highlighted in yellow while our responses are highlighted in blue and our revisions in red.

Reviewer #1 (Comments to the Author):

One of the biggest sources of anthropogenic methane emissions is coal mining. Methane entrapped in coal seams and the surrounding strata is released during coal production. According to estimates from the US EPA (2019), 11% of all methane emissions from human activities worldwide come from the coal mining sector. Many studies contend that methane emissions from fossil fuels are currently underestimated. Furthermore, due to a lack of in-situ and field measurements, there is a considerable level of uncertainty surrounding these emissions.

This study has considered a large number of active coal mines in the Shanxi region, which is one of the significant coal mining regions of the globe, and has implemented a synergistic approach of using both top-down and bottom-up approaches plus validation from nearby ground-based measurements. The paper then goes on to develop correction factors to estimate the coal mine methane (CMM) emissions and compares these emissions with the commonly used CMM from the EDGAR and GFEI v2 datasets to address the biases and uncertainties associated with CMM emissions. This work therefore provides a platform by which a spatially, and temporally quantifiable set of CMM emissions can be obtained.

The major highlight and novelty of the paper is that it has conducted a robust uncertainty analysis, which is then used to create a bound on the CMM emissions on a mine-by-mine, type-by-type, and day-by-day basis. Both spatial as well as individual sites are analyzed, revealing that CMM is underestimated in general against currently widely used emission inventory datasets. In addition, the study approach also paves the way to correct top-down approaches and improve upon emission estimation uncertainties in general.

For these reasons, the paper provides extensive information and covers enough scenarios to produce the best set of observations possible, enabling policy makers to have the data needed to work towards CMM mitigation. This will allow for a more comprehensive and well-supported range of emissions to be controlled.

Thank you very much for this insightful background, detailed summary of the important points, and highlighting the overall findings from our paper. This is indeed one of the motivating factors behind why we have selected Shanxi as our study area. Shanxi accounts for approximately one quarter of all of China's coal mining and consumption, over an area which is relatively small. It is also a region which has not been studied as much by the community as a whole as other areas within China.

We also feel that a greater understanding of the spatial and temporal distribution of methane emissions, as well as their uncertainty from coal mines adds considerable value to atmospheric science studies, top-down emissions studies, global warming studies, design and usage of GCMs and CTMs, and other related academic disciplines. We hope that the work can help to provide a small step towards making more policy-relevant science.

I would recommend the paper for publication and consideration as an excellent paper, after a few more specific details are elaborated upon:

(1) The AD and rank sourced from the <http://nyj.shanxi.gov.cn/mkscnldxgscysxxgg/ggl> which is only accessible in Chinese. If there is no English data or translation available, can this be mentioned more clearly?

We will mention clearly in the revised version of the paper that there is no English version of the website available. We have made the rank data available on a mine-by-mine basis in this paper's data repository (<https://doi.org/10.6084/m9.figshare.23265644>).

(2) Could the ranking of mines and their corresponding coal emission types (used for EF calculation) be elaborated in a simpler way? What is the reason behind assuming Default mines EF can be weighted from high gas mine and low gas mine alone? Does this make a significant difference from a different assumption?

We have added in a couple of sentences to make the ranking of mines and corresponding coal emissions types easier to understand. We are also constrained by how the industry reports their data. For this reason, we do not want to take out the detail, but we agree that a simpler way to explain it is a good idea.

Assuming that default mines are weighted between those of high gas and low gas mines is a conservative estimation, which has been performed by others (Kirchgessner et al., 2000; Wang et al., 2013; Li et al., 2014). To explore the impact of relaxing this assumption, we have re-calculated the default coal mines using the EF from the high gas mines instead of a weighted mixture. While this change is across all of the mines, in reality the change at some mines is very small, while at others it is not insignificant. The total net increase in emissions is computed to be 1.71(1.67-1.82) Tg/year over Shanxi as a whole, compared with the initial total value of 6.80(4.41-7.77) Tg/year, leading to a final net total emission of 8.51(4.41-9.47) Tg/year.

For the purposes of sensitivity analysis this work also calculates the total methane emissions assuming that each mine of default rank also may have EF values the same as high rank mines, in turn providing an upper bound on the emissions estimation, as compared to the more conservative approach applied herein (Kirchgessner et al., 2000; Wang et al., 2013; Li et al., 2014).

To explore the impact of relaxing this assumption, substituting the EF from high gas mines instead of the more conservative weighted mixture approach was used to determine an upper end constraint

on the overall methane emissions. The total net increase in emissions is computed to be 1.71(1.67-1.82) Tg over Shanxi as a whole, leading to a final net total emission of 4.41-9.47 Tg.

(3) Para 70. Typo error “emissions”

This has been fixed. Thank you!

(4) As many other parts of the world, in particular India, the USA, Indonesia, Australia, and Russia also heavily depend on coal as a fossil fuel, they also actively contribute to global CMM. Across these regions, some have coal and geography similar to those in Shanxi, while others do not. How could the work herein be applied to these other regions? What changes would need to be adapted, and what methods and analytical techniques could be retained? Could the authors clarify which input data would be required as a baseline to adapt and replicate this approach (as different nations may have different ranking systems or produce different types of coal)? What is the overall potential for applying this strategy to other parts of the world?

Thank you very much for this interesting and insightful question. We believe that it also helps us to extend the work herein, and are happy to have had the chance to dig deeper into it to address your concerns.

Over regions which have similar geography, coal type, mining process, and deep underground mining as Shanxi, we believe that the process could be reproduced using some of the fitting factors that we have already established within this work. Having access to surface emissions flux estimations, located next to exit ventilation shafts would be required to ensure that the fitting factors are both representative and effectively capture the temporal variability.

In regions which have a different geography, different coal type, use different mining processes, and mine via the surface or less deep underground than in Shanxi, the process may require more assumptions to be usable. In particular the issues of the rank, the AD, how the underground gas is managed for safety reasons, any technologies applied to vent the gas or water in the mines, etc. would lead to different forcing factors. However, if a sufficient amount of above ground flux observations were applied, so as to successfully sample the distribution over these differences both underground and in-situ in the atmosphere, then the overall methodology should be successful.

Improved a priori emissions baseline data would be essential to establish this. In the case of Shanxi, their baseline data is quite rigorous since it has been recorded for safety reasons over a long amount of time. Our own placement of the flux observations was done in coordination with the coal mine itself, so we were able to ensure that we had a proper sampling of the vented air. In both cases, without a decent a priori or access to an appropriate sampling strategy and location, the methodology may not work.

Since the difference between the high-frequency measured CH₄ fluxes in 2021 and 2022 is very small, and the mines herein have similar geological conditions, therefore this work assumed the observed changes well represent the actual observed temporal variation.

Additionally, iteration between the a priori or second guess observed emissions and the underground and overground observations is required to ensure that the top-down and bottom-up parts are in sync with each other. Such a baseline must be extended over a long period of time to more properly account for the “fat tail” distributions encountered. Additionally, if non-community wide known datasets or machines are used, then a more rigorous discussion of the uncertainties will be required. This is particularly so in regions with vastly different geographic conditions.

However, some of the observations were uncertain, had irregularities, or otherwise were not trustworthy. When data was not of sufficiently high quality to support the flux calculation via the WPL correction, the data from that entire day was discarded.

However, we believe that with the right tools and access, that all of these technical issues can be overcome, and the method can be successfully applied around the world.

The rank of a mine is determined from the underground observations of the gas and the airspeed of ventilation, the type of gas, and measurements of risk.

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Reviewer #2 (Comments to the Author):

Review of “Individual Coal Mine Methane Emissions Constrained by Eddy-Covariance Measurements: Low Bias and Missing Sources”.

This paper is conducted to quantify the emissions of methane from coal mines in Shanxi, China, which is treated as a significant source of methane emissions due to its high coal production. The research employs a combination of bottom-up and top-down methodologies to estimate coal mine methane (CMM) emissions. Data from 636 coal mines are scrutinized, and the measurements from an eddy-covariance flux tower is utilized to constrain the CMM results. When juxtaposed with global datasets such as EDGAR and GFEI v2, the study yields several intriguing findings.

The authors have presented an innovative set of methods to combine the in-situ observations to retrieve the CH₄ emissions in Shanxi Province with only one flux tower. However, there are so many complex landscapes in Shanxi Province, such as the Taihang Mountains, and valleys alongside the rivers, will this only one CH₄ flux tower data represent the real emissions within the whole Shanxi Province? As this is an innovative method to help gain emissions, the plausibility of applying its measurements to the other coal mines in Shanxi Province should be clearly clarified.

We agree completely that additional flux observation sites in Shanxi will increase the overall ability to capture the actual emissions variability. Although there are mountain ranges and river valleys in Shanxi Province, most of the coal mines are underground mines constrained to six major coal fields (all of which are from the same historical Carboniferous-Permian period). Therefore, focusing on these different coal field types with their slight differences in geological type may help us to have a better handle on the overall variability. In the real world there will be

some variability on a mine-by-mine basis, especially over relatively short time scales compared with the processes occurring both in terms of the mining itself, as well as the hydrological and geophysical processes that are rapidly adjusting underground due to the impact of the mining itself. However in Shanxi the mines have been developed in the same standardized manner (inclined well development and combined development) and mined following similar processes (caving mining). However, based on surface observations taken using LGR and a mass-conserving emissions framework applied at high frequency around multiple coal mines in Shanxi, it is revealed that the emissions products are still safely located within the ranges observed by the flux tower in this work. For these reasons, we assume that the emission patterns of coal mines in different geographic regions are consistent at the macro level. It is true that there may be changes in the overall shape of the PDF, and addressing this is planned for future work. To these ends, we appreciate that further observations should lead to deeper and more robust results, and in an ideal world with sufficient funding, would propose placing observations in the future to cover at least one observation site per each coal field, as well as across the low gas, high gas, and abandoned types within each coal field.

However, at the present time, this is beyond the scope of this current work. One issue faced will be related to cost, since establishing sufficient observations to more deeply account for the variability, will require a minimum of 40 to 50 days of high frequency observations at each new proposed location. A second issue is access, since many mines will not allow outsiders to place instruments on their property, and therefore it may not even be possible to get observations.

For the comparison of global scale CH₄ data (EDGAR and GFEI) with the CMM results derived in this paper, the timing of these emission inventories could be taken into account to increase the reliability of the comparison results.

EDGAR and GFEI data are limited in terms of when they provide data, as detailed in Section 2.6. In all cases, we use the a priori inventory which is closest in time to the data available. It is important to note that new mines do not suddenly open or change their amount, processing, and type of methods of opening new areas of the coal field for access at a very high time resolution. Therefore, for those sites identified, they are not expected to have changed much over the past few years difference between the reported times of EDGAR and GFEI, and the observations times made in this work. The important finding is that there are many mines which exist which are reported by EDGAR and/or GFEI as having zero emissions, which can never be scaled. A second important finding is that the variability of the emissions over month-to-month or higher time frequency is not available from EDGAR and GFEI. This compared with day-to-day and 30-minute to 30-minute variability which are many times larger. It is clear that without representing this variability, it is not really possible to guarantee the net total CH₄ emissions over Shanxi.

EDGAR is a bottom-up global database of anthropogenic emissions of greenhouse gases and air pollutants that provides estimates based on data reported by European Member States or by Parties under the UNFCCC, using the IPCC methodology. EDGAR provides emissions of CH₄ at 0.1° × 0.1° spatial resolution and both monthly and annual time resolution over the domain studied (Crippa, 2021). GFEI allocates methane emissions from oil, gas, and coal at 0.1° × 0.1° spatial resolution using the national emissions reported by individual countries to the UNFCCC and mapping them to infrastructure locations (specifically using IPCC Sectors 1B1 and 1B2).

(Scarpelli and Jacob, 2021). This work makes specific comparisons between the computed CMM emissions and results using EDGAR version 6.0 and GFEI version 2.0.

Ideally, correction factors could be applied by EDGAR and GFEI to account for the annual-scale changes in emissions in a top-down way, and then applied in the comparison. At the present time we are only aware of this having been done for NO_x (Li et al., 2023; Qin et al., n.d.) and BC (Wang et al., 2021), and not for CH₄.

If a new a priori dataset is known to the reviewer, we will be happy to include a comparison in our revised version.

When the CMM results of this study is compared with those of Edgar and GFEI, it is observed that the emissions reported in this paper are derived from point sources, whereas the emissions reported by the other two inventories are aggregated within a grid range. Consequently, a pertinent question arises as to whether the point sources considered in this study encompass all coal mine sources within the grid. If not, it is plausible that the higher emissions reported by Edgar could be attributed to this discrepancy.

In the small number of grids in which EDGAR and GFEI is higher than our coal mine methane results, this is a possibility. We have examined known urban sources of CH₄ based on CEMS and explained one or two specific grids with this issue. Overall, the number of such grids where this occurs is small. If the values from EDGAR and GFEI are trustworthy on the grids they are given on, and on those specific grids the values are larger than the CMM values given herein, then it could prove a secondary constraint on anthropogenic small CH₄ source emissions, which are not included in the CEMS inventory. This could be a further important finding to explore. Thank you!

What we observe however is that on many grids, the difference between EDGAR and GFEI and our results is such that our results are larger than EDGAR and GFEI, even though both account for mines and anthropogenic sources, meaning that their CMM emission value is very low, that they are missing the locations of mines, or a combination of both. After a careful re-examination, we believe that the bias is mainly influenced by the location of the emissions in EDGAR and GFEI not matching with the data that we have.

Besies, some minor comments should be corrected inside the manuscript.

Minor comments:

The line number is shown with mistakes so that I cannot label the issues or errors with the line number correctly, the authors should correct it.

The line numbers have been changed throughout the paper. Thank you for bringing this to our attention.

More details of the time periods (55 days in two years) of the data used and the filtering method should be mentioned in Section 2.

Thank you for helping us to make the data easier to understand. We have added more details to section 2.

The total length of observations was longer than 55 days, and was spread roughly evenly over the two time periods from 24th October to 21st December, 2021 and from 15th August to 31st August, 2022. However, some of the observations were uncertain, had irregularities, or otherwise were not trustworthy. When data was not of sufficiently high quality to support the flux calculation via the WPL correction, this data was discarded. Specifically, the entire day of data was filtered and removed from the subsequent analysis. At the end, 55 days of data remained.

Line 85 in Page 11, “flux-tower” should be “flux tower” which corresponds to the title of Section 2.3.

All instances of “flux-tower” have been changed to “flux tower”. Thank you.

Line 22 in Page 12, “smapling” should be “sampling”.

Thank you for pointing out this improvement.

Some sentences should be decreased to be clear. Such as, Line 38 in Page 13, “Since the differences between the measured high frequency CH4 fluxes is very small between 2021 and 2022”, and Line 60 in Page 14, “which have CMM emissions which are not known in terms of their geospatial locaction.”

This sentence has been decreased in size. The entire paper has been checked again for similar issues to improve communication and readability. Thank you.

Some writing or spelling errors in the context and captions of figures and tables, such as “whichis much” in Line 70 in Page 29 and “R5)” in Line 2 in Page 25. The authors should recheck and correct them.

Thank you. We have carefully looked over all of the captions of figures and tables. We have addressed both your specific improvement, as well as others.

The CH4 data in both EDGAR and GFEI are used to compare with the CMM results, but few details on the two emission inventories are mentioned in the manuscript.

We have added more details on the two emissions inventories in the methods section.

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I express my appreciation for the time, effort, and deep insights provided by the reviewers, and hope the responses herein will meet the rigorous standards of ACP. If there are any remaining questions, we look forward to taking the time to carefully engage and continuously improve.

Best Regards,

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