The comments from the reviewer are given below in unedited text. Our responses are given in blue font. Any added figures are included in line.

The reviewer finds the manuscript hard to read and understand, due to the confusion in the terms used, the vague model description for emission estimation, and poor English in the manuscript.

1. Confusing terms used in the manuscript.

A) CH_4

In equations (1), (2) and (3), CH_4 represents methane concentration with the unit of ppm, while the in the main part of the manuscript, CH_4 just means methane in English. This creates unnecessary confusion for readers.

Thank you very much for kind reminder. We have revised all the "CH_4" according to your suggestion and now differentiate between methane concentration (hereafter delineated as [CH_4]), methane emissions (hereafter delineated as ECH_4), and methane as a word (hereafter is delineated as CH_4).

B) CH_4 emissions

Why one term "CH_4 emissions" is assigned to two different variables, one in the unit of ppm/min (ECH_4), and another one in the unit of kg/h ($E'CH_4$), and ECH_4 has never been clearly defined in the manuscript: is it the methane concentration change over time at the certain point? Or average methane concentration changer over time over a controlled volume? This is the first time that the reviewer has seen such a term is used to define emissions. Please define parameters with their actual physical meaning.

Thank you very much for this question, which can help us make better improvements to this article. In fact, E_{CH4} is clearly defined as methane emissions in the manuscript, with the unit being ppm/min, which was obtained using the mass conservation model. However, because there are new papers over the past few years which are now using the unit of kg/hour, in order to allow for more simple comparison, we have added Equation 7 and subsequent analysis of our results additionally using this unit. Note that adopting this unit has required additional assumptions to be made connecting our observed wind speed with an assumed area and height, which then leads to an increase in the uncertainty of the overall result. For this reason, we want to provide both sets of emissions data. In all cases, emissions of CH_4 are now clearly delineated as ECH_4 with the respective units determining whether or not the additional equation 7 is applied.

C) Temporal frequency

Why a frequency has a unit of minute? Should it be hertz (event per time) (Lines 136, 143)

The observational frequency of the raw data from the machine used herein is 1Hz, which means one observation is obtained per second, introduced in line 107 of the manuscript (https://www.merriam-webster.com/dictionary/hertz). In order to apply the mass conservation model to calculate methane emissions, the temporal derivative of methane concentration, wind speed, and direction data need to match. To ensure this is the case, the temporal concentration data was averaged to per minute, as mentioned in line 113. Herafter, the unit of per minute is used.

2. The Mass Conserving Model of Measured CMM and the 2-Box model

First, the model or models used to estimate methane emissions from one or multiples coal mines are not well described. The reviewer is expecting the following information to be clearly stated in the model:

A) Is the model dealing with one coal mine or multiple coal mines?

In this study, the Mass Conserving Model of Measured CMM is solving the mass conservation equation to derive the transported emissions observed at the place and time where the observations are taken. The number or locations of the sources are not relevant at this point. However, the Mass Conserving Model of Measured CMM is used to calculate methane emissions within a certain spatial area. The methane emissions measured in this spatial area may come from a fixed methane emission source (a coal mine), from two or more methane emission sources (multiple coal mines), or even other possible sources transported from far away or due to extreme and infrequent events. For example, there is a specific case given on line 305-310 in which the most reasonable explanation for an extreme computed emissions in terms of large magnitude and infrequent occurrence were likely the result of a leaky vehicle. Due to this reason, this specific extreme emission value was not subsequently used in the attribution steps.

The application of the subsequent 2-Box model allows attribution to be obtained based on the sources being one of two different adjacent coal mines. This is done using a statistical forward and backward analysis using all possible combinations of computed emissions and wind directions and speeds which are applicable to the known directions of the two adjacent coal mines. If there are multiple methane emission sources upwind occurring at the same place and time which were transported over the sources with the same time and direction, the 2-Box model may not be able to differentiate this. The model does however produce a statistical result of concentrations obtained using the computed methane emissions from each emission source. The distributions of computed concentrations are shown to be consistent with the observed distributions of measured concentrations, allowing confidence in the attribution.

B) Is the coal mine emission treated as point source or area source?

In reality, there is no difference between the two, since all sources are area sources. It is just that point sources are emitted over a very small area. In this work we are treating the emissions using a Lagrangian parcel when computing the MCM^2 approach. We then switch to a specific set of assumptions of a box in space and time when converting units to kg h^{-1}.

C) Are the methane emissions from the coal mine considered stable or not?

Our computed emissions range in time from a minimum of 3 minutes to a maximum of 22 minutes for each event. There is not any single observed event which is sufficiently long to be stable at the per hour time scale. This is clear from the variation in the observations, and is the underlying reason why this work has focused on updating the mass balance equation at high-frequency. Gaps and differences in terms of magnitude and timing are presented in Figure 6 and in the underlying dataset accompanying the paper https://figshare.com/s/1a393772d7b72ae17e62.

D) What is the control volume that the model is applying?

The concept of a control volume is only applied when converting the emissions results from ppm/min to kg/hr. This is detailed in the manuscript in equations (7) and (8) and section 2.7. In specific, it assumes a plume rise height and area based on wind speed, boundary layer height, and angle of spread (itself a function of the wind speed), thus obtaining a hypothetical control volume. The point is that the plume cannot flow any higher or further than the wind speed allows it to be transported over the length of the total emissions event. The manuscript states the following:

"H is the height of the vertical rise that the emissions undergo within their first minute (m), A is the area (m^2) swept over an arc, which ranges linearly from a maximum of 60^o under slow wind conditions to a minimum of 30^o over very fast wind conditions, based on the wind speed when the direction is found to lead to successful attribution."

"Two different assumptions are made for the vertical extent of the plume rise, since the emissions are computed minute-by-minute which is shorter than the adjustment time throughout the entire boundary layer (Vaughn et al., 2018; Zinchenko et al., 2002). The first is to assume it has mixed within the bottom one fourth of the boundary layer, and the second is that it has mixed based on a steady vertical rise equal to one tenth of the horizontal wind. In this work, results using both assumptions will be presented."

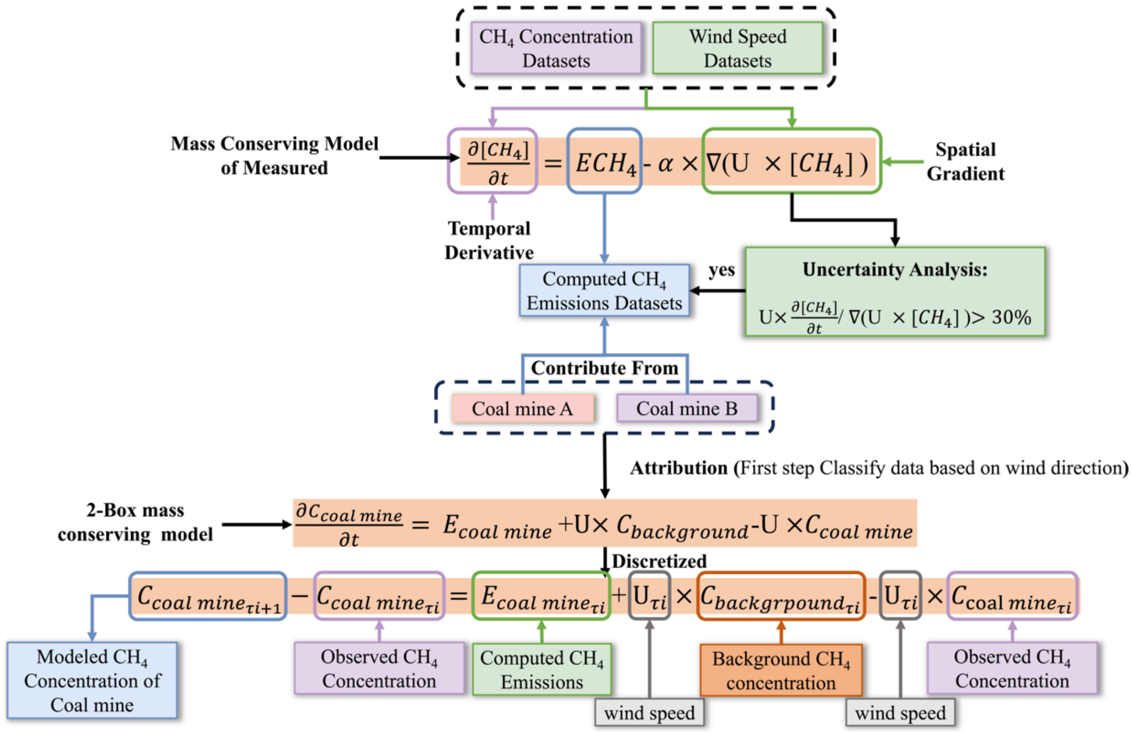
E) What are the boundary conditions and initial conditions if transit process is considered?

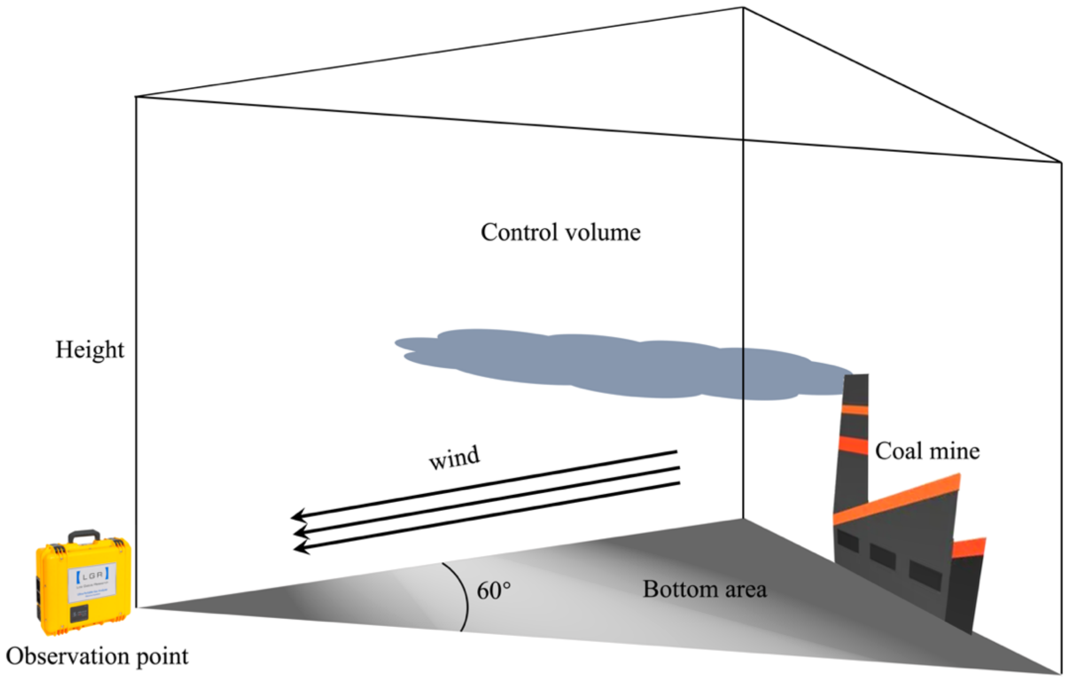
The 2-box model is a set of differential equations, requiring boundary and initial conditions. All possible combination of observed concentrations at the background locations, computed emissions and observed wind-fields which correspond to the appropriate physical direction of the two upwind mines are used to drive the model. All combinations are run until the values converge on a concentration. This distribution of results is then presented probabilistically. The details can be found in section 2.6 and equation (5) and (6), and the results are given in Figure 14 as well as in the dataset at https://figshare.com/s/1a393772d7b72ae17e62.

A scientific description of a mechanism physical transport model should include the following contents:

A) Control volume: the physical region where the model is applying. In the case of this paper, are we considering 3-dimensional box covering multiple coal mines and monitoring points with a height of boundary layer? An illustrative figure will help the readers and authors too.

Thank you very much for your recommendation. The underlying technical issues have been explained. We have now included the following illustrated figures.





Hight: V×T Bottom area: $\pi \times R^2 \times \frac{1}{6}$

V: Vertical wind speed, assuming the vertical wind speed is 1/10 horizontal wind speed here

T: Observation/calculated integration time for methane emissions, between 3 minutes to 8 minutes in this work

R: Radius of a circle, the distance transmitted by the horizontal wind speed during the integration time used to calculate each methane emission

B) Key assumptions of the model (3-dimension transport or one-dimension only? Transit or stable?) which will lead to

We have addressed these points above in more detail. A summary is given below.

The emissions model is a one-dimensional spatial and one-dimensional temporal model, which is not assumed to be stable in time, and is therefore computed on a minute-by-minute basis. The 2-box attribution model is assumed to be 1-dimension in space, and is solved to equilibrium in time using all possible combinations of observed emissions, wind speed, background concentration and direction.

The final conversion to the units of kg h^{-1} is assumed to be 1-dimensional in time with transport filling a control volume, computed on a minute-by-minute basis, and then aggregated to per hour.

Thank you very much for your asking us to be clear on these points.

C) Control equations base on mass balance and simplified by key assumptions

Our equations fully satisfy mass balance, and are the same equations used in regional mass-balance models such as WRF-CHEM, and global mass-balance models such as GEOS-CHEM, CESM, etc. There are key assumptions made that the transport is one-dimensional in space and on-dimensional in time, although the equations are updated on a minute-by-minute basis. The divergence accompanying the observed minute-by-minute changes in the wind speed and direction are accounted for in this work.

D) Boundary equations and initial conditions

We believe that the model is explained in the manuscript. We have worked hard to improve our communication, readability, illustration, and more, and hope that the reviewer and the community has a clearer understanding at the present time.

We believe that some of the assumptions below stated by the reviewer indicate that the reviewer may be accustomed to a subset of mass balance models which are not comprehensive and have already made certain critical assumptions which we do not make in our work. We will take the time to slowly and carefully address each point, explaining how and where our model works to address the questions raised. In fact, our equations can be simplified into such a similar model, but we chose to not do so due to the fact that the observed wind and chemical concentrations are rapidly changing throughout the entire dataset, which disallows these assumptions to be made.

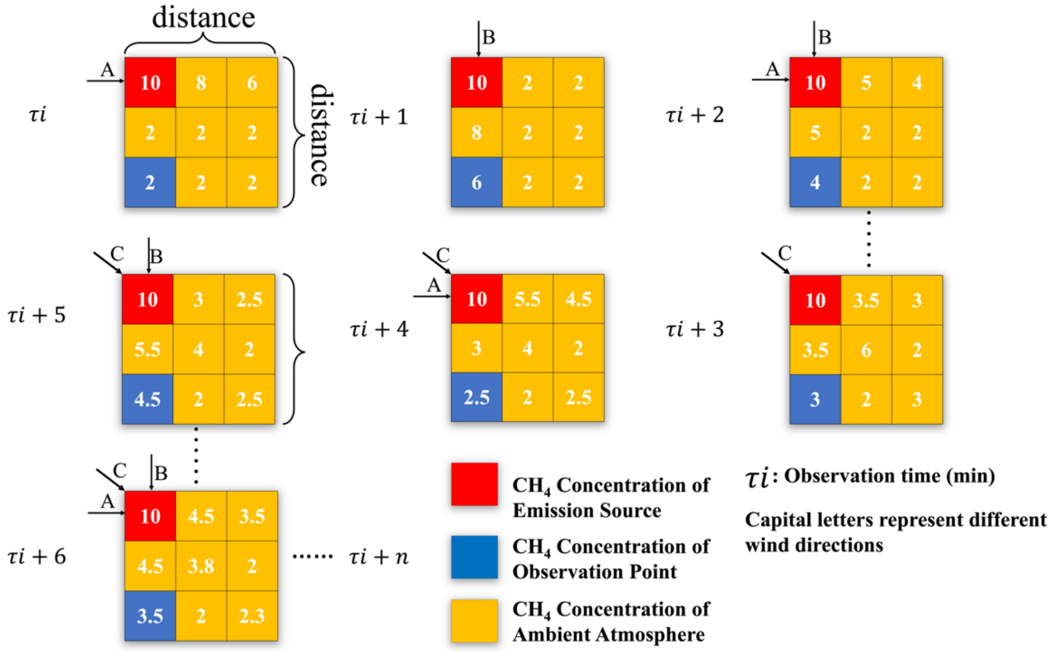
Secondly, the model itself is questionable. Since the model is not clearly defined in the manuscript, the reviewer cannot assess it accordingly. But in principle, the changes in methane concentration (ppm/min) downwind of a coal mine should be contributed to:

1) Variation of coal mine methane emission rates (not the absolute emission rate, but the changes)

This is not necessarily true. For example, you can have a case where the emissions is constant but the downwind observed methane concentration does change in time. This can happen when there is a divergence in the wind field. Please observe figure R2 below to highlight this case. In this case, the change in concentration downwind (in ppm/min or ton/min) will change as a function of the absolute emissions rate. Based on our wind speed and direction data, we indicate clearly that this condition is observed to occur frequently in our study area.

Note that our equation includes this effect of both absolute emissions and changes in Emissions. Our equation includes both an active time derivative and spatial gradient of the wind times the concentration. Note that the spatial gradient expands into multiple terms due to the chain rule. This allows for both the absolute values and the spatial and temporal changes in the computed emissions and the observed wind speed direction and the concentration to all matter.

Simple Examples: Absolute Emissions is Constant



Absolute emissions is constant, but the wind direction/speed change, the CH_4 concentration change can be observed

2) Changes in wind speed and direction (again, not the absolute value)

Again, the absolute value of wind speed and direction can be constant and can have an impact on the downwind methane concentration per minute change. This does occur under two different situations. First, when there is a time change in the emissions. However, it also does occur when there is a stable divergence anywhere in the air column above the observation site. In this case, both the emissions and the wind fields are constant, however, a vertical wind flow is induced. Similarly, a change in the vertical mixing between cleaner air above and dirtier air below, or a complete lofting of the emitted particles rapidly from the surface upward, so that they are not ever measured at the ground site. On top of this, there could be a total column change in the air pressure, especially if the observations are located near a mountainous area, as occurs in this work. These effects were observed within the area and times of the study herein. In all of these case, it is essential to know the absolute value of the wind speed and direction.

Note that our equation includes this effect of both the absolute values and changes in wind speed and direction to all be included. See above for detailed explanation.

3) Noise created by the instrument itself.

We have intentionally excluded all changes smaller than 30% to ensure that we are not considering observational error. This value could be changed or a sensitivity experiment carried out. We have decided to be very conservative in this case.

In other words, the changes in methane concentration (ppm/min) at a certain downwind point is not related to the absolute methane emission rate (kg/h) upwind, how can we estimate the methane emission rate (kg/h) from the downwind methane concentration change (ppm/min)?

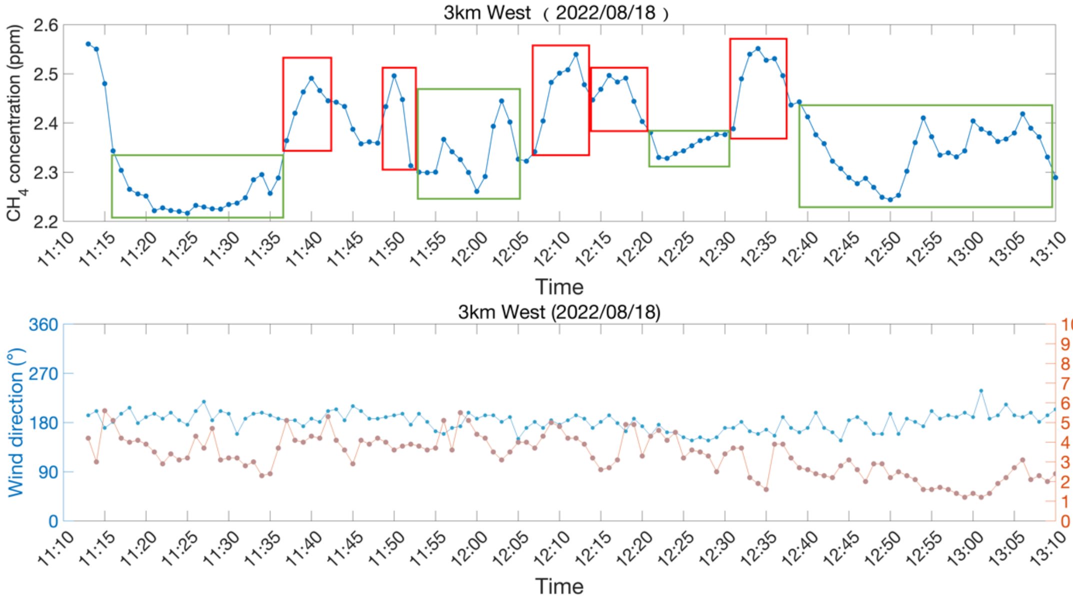
Consider an extreme ideal scenario: a coal mine emits 1000 kg/hr as a points source, the wind is precisely eastward at constant speed of 1 m/s. The instrument downwind measures the true in-situ methane concentration without any noise. In this case, we will observe constant methane concentration with 0 methane concentration changes, and lead to 0 methane emission rate from the model described in the current manuscript, which does not make sense.

In fact, the extreme ideal scenario you haven assumed is not found to exist anywhere within the work done here for a 1-hour period. Since your assumption is in terms of kilograms per hour, then the wind must meet this assumption for an entire hour. Otherwise, the situation would not be accurate. Since the measurements are used over a long period of time, eventually the wind changes direction, and for this reason will be picked up as a change in time. On top of this, since the original equation is based on a gradient, adding in a second instrument or co-locating at a second location even just a few meters away would immediately pick up the gradient and register a vastly different number. Since in reality all air, even in your assumed case, as some amount of diffusion, therefore in reality the signal would be picked up. In fact, if the horizontal gradient across your assumed ideal line is really as strong as you say (1000kg/hr on the line and 0kg/hr off the line) then the diffusion will be very large. This will be picked up by the derivative of the second order term (wind times concentration) very effectively. This theory is in fact supported by the equations used by mass flux observational systems (Wofsy et al., 1993).

In the process of observation, there were indeed situations where CH_4 concentration and wind speed changed very little over ten- to thirty-minute-long periods. Over these time periods, since the variation was under 30%, we decided to not compute emissions. This is an aspect of our quality control, to ensure that machine noise or measurement error do not lead to erroneous emissions estimations. All quality control steps applied before using the Mass Conserving Model to compute the CH_4 emissions are detailed in the manuscript in equations (4) and section 2.4.

However, we selected four different directions and multiple observation points around the coal mine for CH_4 concentration observation, with a high time frequency of 1 Hz.

Here is an example of some of the data we have obtained. Note that in the case below, we have only found that the variation in time is sufficient to compute emissions occurs over the times boxed in red. The other times still exhibit changes in concentration, but are not included in the emissions calculation since the respective change in wind times concentration is smaller than 30%.



If a model cannot deal with simple scenarios, it cannot treat the complex situation when transit process is considered.

We fully agree with you. We invite the models of the type you are referencing to use our data, and compute emissions at higher frequency. We would also like to see how the uncertainty range changes for the computed emissions. We look forward to working together more to compare and contrast different approaches under more realistic conditions. We believe that this is important, since we cannot find any case in our results in which the data is long enough as to compute emissions continuously over 1 hour. We have made a set of assumptions to transform our units into the emissions per hour basis, as described in section 2.7.

We believe that the approach in this paper offers a unique approach to compute CH_4 emissions at high frequency (even if it is rough and has uncertainty). We have derived and computed an unbiased way to consistently compute at high temporal frequency emissions, including variability, and successfully attributed these emissions to two different substantial coal mine based sources.

2. Language and logical issues in the manuscript

A) Lines 40, 41: CH_4 emission estimates are highly uncertain in both space and time (Brandt et 41 al., 2014; Saunois et al., 2020b).

When people talk about the spatial and temporal variability of oil gas emissions, they are referring to the real emissions, not the emission estimates.

Our work at emissions estimation is to quantify the actual emissions and an uncertainty bound at high frequency. We are talking about the same thing. It is the rate at which the CH_4 escapes into the atmosphere from the source. The units can be ppm/min, ton/hour, etc., but the actual thing is the same.

We have removed the word "highly" next to uncertain to be a little bit less strong. But the fact of the matter is that there is a lot of work we have done (and others as well) which indicate clearly that there are issues of spatial mis-match, temporal mis-match, and absolute value mis-match between various platforms and observations.

If the issue is the difference between bottom-up and top-down emissions estimates, then this is an interesting area. In reality the results from both should match each other, unless there is a fundamental mis-match in how emissions are being talked about and discussed.

B) Lines 40-45: For these reasons, new approaches to quantify, reduce uncertainty, and attribute CH_4 emissions are necessary and can provide support for policies aiming to control and mitigate 45 CMM (Cao, 2017).

New approaches are necessary not because the emissions change with time and location, nor the fat tail distribution. It is because we need more accurate and economical tools.

Re: The original content of lines 40-45 in the manuscript is: "CH_4 emission estimates are highly uncertain in both space and time (Brandt et al., 2014; Saunois et al., 2020b). They also generally have a fat tail distribution, wherein a small number of samples have extremely large emissions that overwhelm emissions under average conditions (Duren et al., 2019; Plant et al., 2022). For these reasons, new approaches to quantify, reduce uncertainty, and attribute CH_4 emissions are necessary and can provide support for policies aiming to control and mitigate CMM (Cao, 2017)."

As you said, we need more accurate and economical tools. One of the reasons why is that many current tools are not capable of inverting the emissions at high frequency, or under complex meteorological conditions. However, as some recent work has demonstrated (Li et al., 2023; Qin et al., 2024; Qin et al., 2023), emissions variability in time may be a dominant factor behind annualized or other long-term emissions estimates. For this reason, quantifying changes at high temporal frequency alone directly contributes to being more accurate ant economical.

For example, coal mines, which are sources of methane emissions, do not emit the same amount of methane all times of day and all days of the year. There are impacts due to mine size, production, underground geology, possible emergency situations underground, water table, changes in energy, coal, materials, or other demand, efficiency of coal bed gas use or retrieval, and other possible factors as well. Satellite remote sensing approaches to computing methane emissions currently have lower spatial and temporal resolution compared to the high-frequency ground observation method used in this study. The transit time of the satellite is fixed every day. Clouds or aerosols vary day-to-day. Some of these factors of variability may introduce an extremely high event from being sampled, leading to an over (or under) sampling of such events. Additionally, low emissions events may not be distinguishable from background noise or other urban-based sources which have advected into the same grid. This could lead to an over (or under) sampling of such very low events. Given all of this, emissions data observed by satellites from coal mines cannot even meet a daily temporal frequency, let alone and hourly frequency. Therefore, we further need methane emission observation methods with higher temporal and spatial resolution to achieve more accurate estimation of methane emissions and help improve the results which are obtained from satellite estimations.

C) Line 54: Uncertainties are rarely assessed holistically or in detail (Cohen and Prinn, 2011; Cohen and Wang, 2014).

Be careful to make such a claim. Almost all methane measurement papers have one section addressing and reporting their measurement uncertainties.

Uncertainties consist of measurement uncertainties, model uncertainties, and parametric uncertainties. Thank you for your words of caution. We will more carefully word this part. However, we have reviewed many such methane measurement papers recently published, and find that a significant number of these papers do not address the uncertainties in the underlying model assumptions raised in and addressed by this work, as well as issues with parametric uncertainty in general.

D) Line 55: Airborne remote sensing is a highly technical and costly approach to record CH_4 fluxes from...

Where does the claim on "costly" come from? Actually, aerial approach has been widely employed due to its relative low cost comparing to other approaches. It will definitely less expensive to deploy than the method described in the current manuscript.

Airborne remote sensing requires the use of an aircraft, which is a very expensive item. Furthermore, even if they have sufficient funds, many research teams do not have access to an aircraft or to the ability to fly without extensive permissions. On top of this, just possessing the aircraft does not mean that performing remote sensing from an aircraft platform is simple: it requires an extensive number of disposables, a large number of people hours, and extensive training, adding to the cost. We are happy to share with the reviewer that we have spent far less for this entire experiment than it would cost us to just rent an aircraft capable of being flown over our region for one day.

In addition, aircraft based remote sensing makes many assumptions about the atmospheric column between the aircraft the surface source. How different thin clouds, aerosols, changes in surface reflectance, and other confounding factors are handled is highly technical. It also adds to the sources of model uncertainty as well as parametric uncertainty. Thank you for your work using aircraft observations!

We hope that this work can present a new opportunity and perspective to the community to also make better use of high frequency surface-based measurements.

E) Lines 61, 62: … but only after being calibrated by upward looking remotely sensed measurements…

Aren't almost all instruments need to be calibrated before adoption?

Different calibrations and methods are required under different atmospheric and geographic conditions. The comment in the paper being referred to is the EM27/SUN. This upward looking instrument is used to calibrate TROPOMI xCH_4 columns. The EM27 itself also needs to be calibrated, in general against other EM27 devices in-situ. One source of uncertainty is that the calibration itself may also vary across different environmental conditions.

F) Lines 73-74: This work employs a high-frequency surface-based observation platform of CH_4 concentration, which is portable, economical, and unaffected by most environmental factors

What is the proof for "economical"? Do not make any claims that you cannot support.

Based on our costs of analyzing CH_4 in Shanxi, China using this method, flux tower observations, surface based remote sensing and satellite based remote sensing, we have found the approach herein to be relatively less costly. The reasons for its economic viability are as follows: (1) The instruments are relatively inexpensive and readily available to rent and/or purchase; and (2) after setup and appropriate training, these analyzers can be run by a single student, reducing personnel costs.

F) Lines 78-79: Continuous 79 observations were made around known coal mines, unknown sources, and of background conditions.

Delete "of"?

Thank you for your suggestion. It has been revised in the manuscript.

Lines 80-90: High-frequency emissions calculated using these data were used to drive a 2-box model to attribute 81 emissions to the known mine and a second low production mine previously thought insignificant.

The model used the data to derive … not the data is used to derive the model.

Thank you for your suggestion. It has been revised in the manuscript.

G) Lines 86-89. While the authors are talking about Changzi basin, why do we claim "province-wide" background are high?

We have access to additional surface and other observations showing that the values of CH_4 are high province-wide. However, we agree with this change, since the focus of this work is on the Changzi basin. We have hereafter changed "province-wide" to "city-wide" or "basin-wide".

J) Line 92: Observations were positioned along concentric…

Should be "instruments" be positioned, not observations

Thank you for your suggestion. It has been revised in the manuscript.

There are so many similar issues in the manuscript, and the reviewer will stop here at Page 5. The reviewer recommends having somebody proofread the English in the manuscript.

References:

Li, X., Cohen, J. B., Qin, K., Geng, H., Wu, X., Wu, L., Yang, C., Zhang, R., and Zhang, L., 2023, Remotely sensed and surface measurement-derived mass-conserving inversion of daily NOx emissions and inferred combustion technologies in energy-rich northern China: Atmospheric Chemistry and Physics, v. 23, no. 14, p. 8001-8019.

Qin, K., Hu, W., He, Q., Lu, F., and Cohen, J. B., 2024, Individual coal mine methane emissions constrained by eddy covariance measurements: low bias and missing sources: Atmospheric Chemistry and Physics, v. 24, no. 5, p. 3009-3028.

Qin, K., Lu, L., Liu, J., He, Q., Shi, J., Deng, W., Wang, S., and Cohen, J. B., 2023, Model-free daily inversion of NOx emissions using TROPOMI (MCMFE-NOx) and its uncertainty: Declining regulated emissions and growth of new sources: Remote Sensing of Environment, v. 295, p. 113720.

Wofsy, S. C., Goulden, M. L., Munger, J. W., Fan, S.-M., Bakwin, P. S., Daube, B. C., Bassow, S. L., and Bazzaz, F. A., 1993, Net Exchange of CO_2 in a Mid-Latitude

Forest: Science, v. 260, no. 5112, p. 1314-1317.