**Dear Editors and Reviewers**

The point of this work is based on the fact that all observations have uncertainty, and due to the non-linear nature of atmospheric mass conservation, the computations which convert observed atmospheric column concentrations and associated uncertainty into emissions produce a non-linear response on computed emission uncertainty. We specifically aim to conservatively yet explicitly include these uncertainties on the emissions computed, so as to yield a realistic and reasonable final product, with a reasonable range of values.

To our knowledge, this is the first work done which has attempted to apply this basic science-philosophy approach to estimate emissions of CH4 using observations of XCH4 from satellite. Our group has just published a paper using a similar approach (with quite different chemical and dynamical driving terms) to estimate emissions of NOx using observations of column NO2 also from TROPOMI (Lu et al., 2025a). In general, we know that the uncertainties in the retrieval of the XCH4 will be done to uncertainties and incomplete scientific knowledge of clouds, aerosol, and surface terms. We know that these uncertainties must be smaller than the uncertainties of retrieved gas products, since the total impact on radiation streams from these three is an order of magnitude larger than that of the gasses.

We have specifically published work using aerosol products from TROPOMI, OMI, and MISR where we demonstrate that the aerosol uncertainty is at least 5% (Tiwari et al., 2025; Liu J et al., 2024; Liu Z et al., 2024) and demonstrate using column and surface observations that the variability on inverted radiative products is also at least of a similar amount (Tiwari et al., 2023; Guan et al., 2025). In general, we have found that TROPOMI NO2 products, which are considered one of the most reliable gaseous products, has an uncertainty ranging from at least 10% (Qin et al., 2023; Lu et al., 2025b; Wang et al., 2025; Li et al., 2023), to possibly as high as 40% or more (Boersma et al., 2018; Pollard et al., 2022).

Additionally, the differences between the observations from TROPOMI and from the long-term WMO surface station are up to nearly 10% as explained in the initial version of the paper published. We now also include air-core observations and find that these closely match the surface observations and also have a difference of around 10% (or possibly even more) when compared with TROPOMI. Given all of the above reasons, we believe that using a 10% uncertainty for XCH4 is reasonable.

We specifically believe that we should not need to compute the exact uncertainty for each contributing factor in the inversions, as this is an issue with the underlying physics based radiative transfer model itself. It is interesting work, and we hope that those who produce the column products can continue to pursue ever improved concentration

inversions, which better represent the underlying uncertainties. The point is that uncertainty is meant to account for both what is known, as well as other sources of uncertainty in terms of what is unknown, or what is currently not able to be measured.

In response to the reviewer's comments, we have provided comparisons with aerosol extinction, aerosol absorption, and surface albedo, and demonstrate that in fact that our emissions are found to be robust and reliable in areas where these factors all have less of an impact on the retrieval, while many of the grids found to not have reliable emissions have a larger impact from these driving factors. This demonstrates that our philosophical approach is in fact reasonable. The community still does not fully understand the physics and composition of the atmosphere, and therefore there are still physical mechanisms and calculation approaches, as well as parts of the atmospheric composition which in reality are related to the retrieval of XCH4 but are still not certain. This applies not only to the aerosol extinction, aerosol absorption, and surface albedo, but also to other factors we currently cannot readily observe including water vapor, and carbon monoxide, which are currently not well known.

For these reasons, we believe that our approach is reasonable and conforms to high standards of scientific enquiry. We do believe in satellite products and want the community to feel confident in their use. For this reason, our work proposes an unbiased framework by which a reasonable accounting of their uncertainty can be robustly applied, and therefore the community can gain more confidence in satellite observation use to solve real-world problems.

**Author response – RC3**

We thank the reviewer for the thoughtful comments provided. We respond to them below in the following manner: the comments are directly copied in black, our author responses in blue, and suggested new manuscript text is indicated in green. New line numbers in the revised manuscript are provided

**1.Regarding QA=1.0**

**• If the area does not have enough observations after applying QA = 1.0, it clearly indicates that the data are not sufficient for this test. I had suggested adding additional filters such as AOD and albedo, together with QA > 0.5, to help avoid possible artefacts. However, the author treated this approach as merely a philosophical choice to avoid artefacts, which is not correct.**

Our paper is attempting to quantify a way by which an objective analysis can be performed on retrieved XCH4 values and their uncertainty in tandem, so as to invert CH4 emissions with sufficient precision, that the inverted emissions are not merely an

artefact of the observed XCH4 noise. We fully agree that pixels with unusual albedo and aerosol interactions used for the inversion of XCH4 concentration are pixels which likely will have more uncertainty. What we have done is to use all of the data which has a QA>0.5, since this is what the community currently does as a standard. This was already mentioned in the previous version of the reply comments, please refer to the first response of comment to reviewer 3.

We specifically argue that AOD is insufficient, and claim that knowledge of the AOD at a minimum of two wavebands, merged with information of either SSA or AAOD at a minimum of two wavebands is in fact required to determine whether or not aerosols are substantially contributing to issues with XCH4 retrieval (i.e., Tiwari et al., 2023; Liu et al., 2024; Tiwari et al., 2025; Liu, J et al., 2024; Guan et al., 2025). For this reason, the version of the paper you reviewed in Figure S1 already included information about aerosols. In specific we demonstrated that the AAOD at 440nm, the AAOD at 880nm, and the ratio of the two AAODs (as observed from MISR over the region) following (Liu et al., 2025) can provide insight. Areas with a large AAOD ratio indicate there is more larger absorbing aerosol, which is more likely to impact the radiation wavebands which are used in the inversion of XCH4.

To further address your concerns, we have done additional analysis. We observe that the values of the ratio are somewhat larger over areas where we have filtered the emissions as being non-realistic [filtered points] (such as area A in Figure 4a). Furthermore, we observe that most of the grids where we have valid emissions [retained points] are located are in areas with very low AAOD ratios, as demonstrated in the new Figure S1g below. Therefore, our approach is successful in determining clearly that the few pixels impacted by aerosols in the region are removed using this process.

To further demonstrate the effectiveness of our method, we provide the PDF of the ratio of AAOD corresponding to the filtered points (Figure 5a), as well as the PDF of the ratio of AAOD corresponding to the retained points (Figure 5d). It can be clearly seen that the filtered emissions include some points with extremely high AAOD ratios, and most of the points with relatively high ratios of AAOD have been removed.

This result offers a physical explanation of why there is a substantial uncertainty in the XCH4 retrieved values, and strengthens our argument that 10% uncertainties are reasonable. What is also important is that our approach is capable of detecting such a non-linear uncertainty propagation, while standard emissions estimation approaches (Schneising et al., 2020; Veefkind et al., 2023; Hancock et al., 2025) in fact compute negative emissions, without realizing that the emissions are merely due to observational uncertainty.

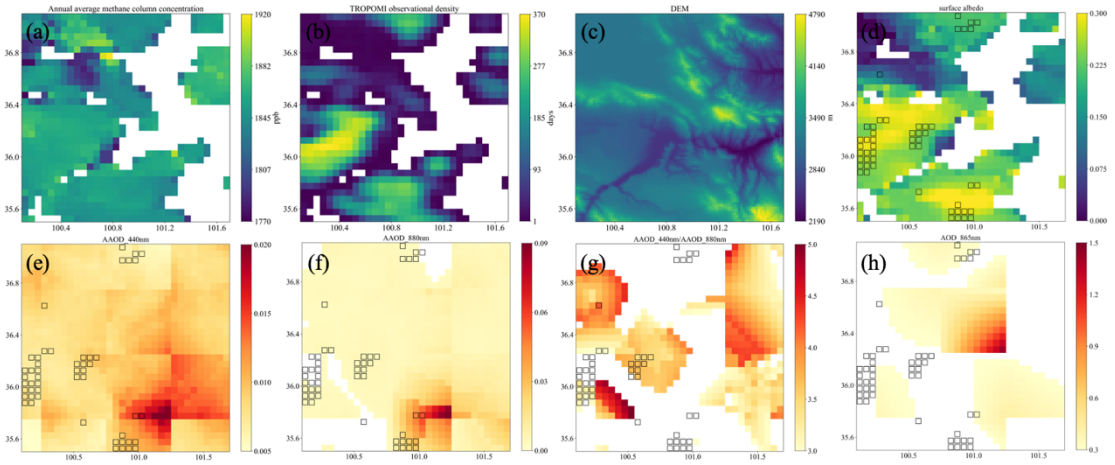


Figure S1: (a) Annual average methane column concentration of Waliguan region; (b)TROPOMI XCH4 observational density in Waliguan region; (c) Surface pressure in the Waliguan region; (d) The average surface albedo of Waliguan region; (e) The average AAOD_440nm of Waliguan region; (f) The average AAOD_880nm of Waliguan region; (g) The ratio of AAOD_440nm to AAOD_880nm; (h) The average AOD_865nm of Waliguan region. The black box represents the grid where our retained valid emissions are located

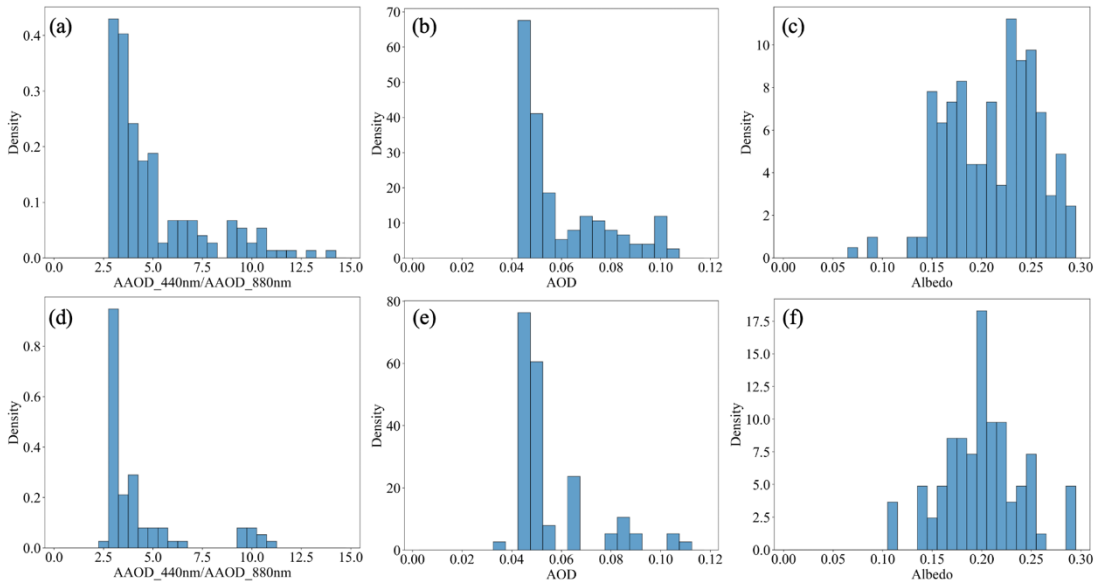


Figure 5: (a), (b), and (c) are the PDFs of the ratio of MISR AAOD observed at 443nm to AAOD observed at 865nm, MISR AOD observed at 865nm, and albedo corresponding to the location of the filtered emission (invalid emissions), respectively; while (d), (e), and (f) are the same respective values, but corresponding to the location of the retained emissions (valid emissions), respectively.

The above content has been supplemented in the manuscript, corresponding to lines 288 to 294: “Next, we examine the impact of aerosol absorption based on MISR

AAOD at 443nm, 865nm, and the ratio of the two AAODs as given in (Figures S1e, f, g) following Liu, Z et al. (2025). Areas with a large AAOD ratio indicate the presence of larger-sized absorbing aerosol, which is more likely to impact the radiation wavebands used in the inversion of XCH4. We observe that the ratio is somewhat larger over areas with emissions we have filtered (such as area A in Figure 4a). Furthermore, we observe that most of the grids with valid emissions are located are in areas with very low AAOD ratios. The PDFs of the AAOD ratio for the invalid and valid emission points, shown in Figure 5a and d respectively. Therefore, our approach is successful in determining that the pixels more impacted by aerosols are in fact filtered."

**Figure 4(d) and 4(e) show high AOD in this area, which likely introduced bias in the data and resulted in negative emissions.**

We would like to clarify that the reviewer's comment is based on a misunderstanding. As described above, the presented plots are of AAOD, not AOD. AOD reflects the extinction, which changes the entirety of the stream of energy (Liu et al., 2024; Tiwari et al., 2023, 2025), while AAOD reflects the absorption, which affects the line-by-line radiance absorption of the XCH4 when inverted using Beer's Law, DOAS, or similar physics-based techniques (Kuhlmann et al., 2025; Tian et al., 2021).

We have added in specific data of the AOD (from MISR at 865nm). To display the areas with higher AOD more clearly, we have removed the values with AOD less than 0.3, as shown in Figure S1h, the AOD of the locations where our valid emissions are located is almost all less than 0.3. Figures 5b and e show the PDFs of AOD corresponding to the points of the invalid and valid emissions, respectively. Although the AOD in both Figure 5b and e is relatively small (all less than 0.11), the valid emissions have less AOD (MISR) than invalid emissions.

We also added TROPOMI AOD_SWIR as shown in Figure S2, The AOD at the location where our valid emissions are located remains very low, and the valid emissions have less medium level of AOD than invalid emissions as show in Figure S2a and b.

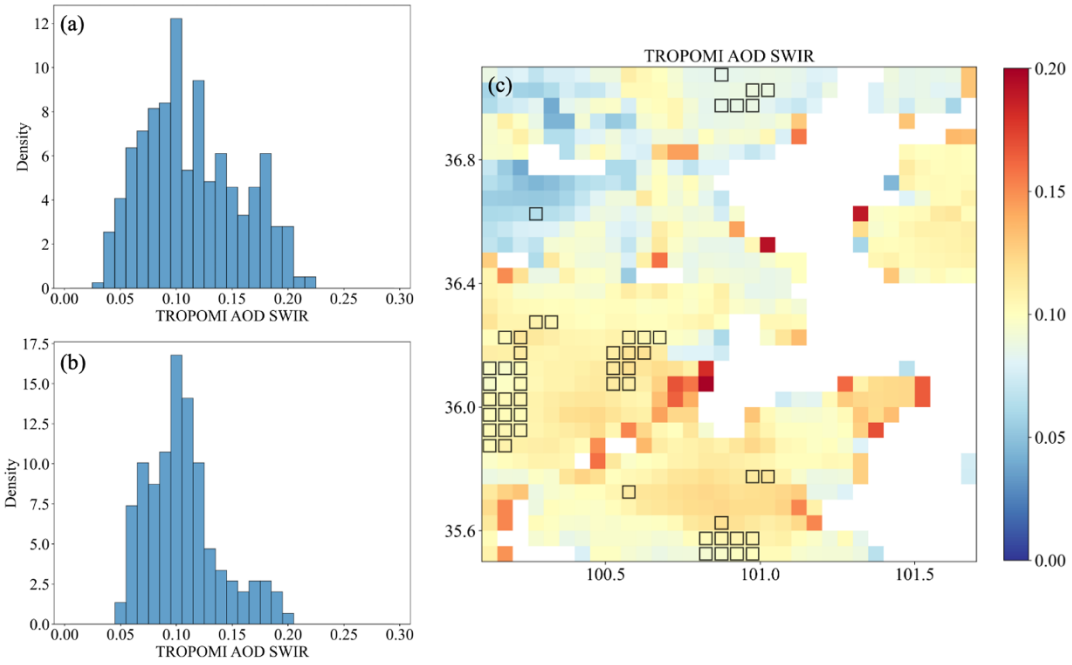


Figure S2: (a), (b), are the PDFs of the TROPOMI AOD_SWIR corresponding to the points of the invalid emission and valid emissions, respectively; (c) The average TROPOMI AOD_SWIR of Waliguan region. The black box represents the grid where our retained valid emissions are located

The above content has been supplemented in the manuscript, corresponding to lines 277 to 287: "Aerosols impact XCH4 in two different ways, through scattering and through absorption. First, aerosols increase radiative scattering, changing the entirety of the stream of energy (Kahn et al., 2023; Liu et al., 2024; Tiwari et al., 2023, 2025), while also absorbing radiation, affecting line-by-line radiance absorption used to invert XCH4 based on Beer's Law, DOAS, or similar physics-based techniques (Kuhlmann et al., 2025; Tian et al., 2021; Guan et al., 2025). We have included observations of AOD at a band as close to that retrieved from TROPOMI as possible (specifically observed by MISR at 865nm). As shown in Figure S1h, almost all of our valid emissions occur at locations where the 2019-2021 average AOD is less than 0.3. Figures 5b and e show the PDFs of AOD corresponding to the points in space and time of the invalid and valid emissions, respectively, while spatial plots (see Figure S2b and e) detail that while all grids are low, that those grids with valid emissions have lower AOD than grids with invalid emissions. Furthermore, we have analyzed the TROPOMI AOD_SWIR product (Figure S2), and found similarly that where our emissions are valid, the AOD remains both very low, as well as being smaller than the AOD on the invalid emissions locations, as show in Figures S2a and b."

These figures have been added to the manuscript and supplementary file, corresponding to Figure S5, Figure S1, and Figure S2.

**How can we trust these values? Additionally, the author mentions that this is a desert area. I requested albedo images, but they were not included in the revised version.**

We have included the albedo values. As shown in Figure S1d, our retained emissions do not occur at locations with either very low or very high surface albedo. Moreover, compared with the retained emissions, the surface albedo corresponding to the points where the filtered emissions are located are typically at the extreme ends of the albedo range. These invalid emissions have been effectively filtered out by our method.

The above content has been supplemented in the manuscript, corresponding to lines 295 to 297: "The surface albedo in this region as shown in Figure S1d. Our retained emissions do not occur at locations with either very low or very high surface albedo. Moreover, compared with the retained emissions (Figures 5c and f), the surface albedo corresponding to the filtered emissions are typically found closer to the extreme ends of the albedo range."

**In coastal, the methane remote sensing community has recognized both low and high albedo values when using QA > 0.5. How are such artefacts assigned in this study?**

In this work, we did not have emissions on the water region. We only have one emission grid on the coast, and the ground albedo on this grid is substantially larger than over the water region.

However, these factors alone cannot explain the total uncertainty, which may be influenced by other parameters, such as cloud, sensor and waveband resolution issues, etc. We assign reasonable uncertainty and use all data QA > 0.5, which we believe represents the most appropriate balance between data quality and coverage, and will support more future physically based research.

The above content has been supplemented in the manuscript, corresponding to lines 268 to 276: "The point of the filtering is to determine whether or not observational uncertainty or noise was the source of the retrieved emissions, or if the retrieved emissions were due to a physical signal. Some physical factors which contribute to signal observational uncertainty or noise which still exist in the QA>0.5 data include but do not fully filter for thin clouds and aerosol layers as well as moderate variations in water vapor, or medium-low albedo (Hu et al., 2016). This QA level also does not consider aerosol absorption or apply any constraints on co-absorbing gasses including but not limited to carbon monoxide. However, this work specifically analyzed a some of these drivers, and clearly determined that some of these drivers in fact contributed to those grids which were filtered (i.e., areas in which the emission derived from the signal

were negative or sufficiently small or large and positive to be considered noise)."

Lines 298 to 308: "These factors alone cannot explain the total uncertainty, which may be influenced by other parameters, such as cloud, sensor and waveband resolution issues, etc. This result offers a physical explanation of why there is a substantial uncertainty in the XCH4 retrieved values. What is also important is that our approach is capable of detecting such a non-linear uncertainty propagation, while standard emissions estimation approaches (Schneising et al., 2020; Veefkind et al., 2023; Hancock et al., 2025) in fact compute negative emissions, without realizing that the emissions are merely due to observational uncertainty. To be clear, this approach herein is further validated, since applying the uncertainty in general does actually capture a subset of physical driving factors which are expected to lead to greater retrieval noise."

**2.Ground-based measurement comparison with TROPOMI total columns**

**• Although the station is located at high altitude and represents free-tropospheric concentrations (considered as WMP background concentration), surface measurements cannot be directly compared with total column measurements, nor should their differences be treated as errors in the TROPOMI concentration. If the goal is to compare a ground-based station with total column measurements, air core measurements should be performed, as discussed in this paper: https://amt.copernicus.org/articles/10/2163/2017/**

We have compared the surface observations with air core observations as given in Figure 2 of Tao et al., (2024), which actually overlap on a day with TROPOMI and surface data, and are located close to our region of interest.

Tao et al (2024) used AirCore-measured CH4 profile compare to the spatially and temporally nearest satellite-measured (L2) and simulated CH4 profiles and found that the satellite significantly underestimated the CH4 enhancement. This difference is consistent with what is shown in our Figure 1.

We have found that the air core vertical observations and uncertainty bound match very well with the surface observations that we are using. This again supports that based on measurements, a 10% uncertainty applied to TROPOMI over the region of interest is reasonable.

The above content has been supplemented in the manuscript, corresponding to lines 217 to 220: "Observed vertical CH4 profiles were made by Tao et al. (2024) Figure 2 using AirCore at the same time and very close to where this work's satellite observations were analyzed. The results compare closely with the surface observations

made at the WMO station in Waliguan, validating that the surface observations in this region are a reasonable representation of the column average values, as shown in Figure 1."

**3. Method application and uncertainty quantification**

**• The method applied in this study requires proper uncertainty quantification, which is currently lacking. Important aspects such as wind fields,**

We believe that this statement is consistent with the methodology as explained on line 104 where we have used multiple different wind fields, possibly being mis-understood. We used daily wind data across the range from 550 to 750hPa. Our approach is a model-free analysis which actively calculates first order wind-based advection and pressure-based advection, as well as first order diffusion, similar to WRF, GEOS, and other regional and global scale models. We likely believe that the reviewer may not have realized that we do not use wind linearly, as we have stated in Eq (2), although many satellite based approximation of emissions in fact do just apply wind as a linear correction factor. Our calculation is based on the gradient of XCH4 as well as the gradient of the wind, which means that the propagation of white noise is due to the nonlinear propagation associated with the gradient term, not the linear wind field, as described in Eq (2). Due to the significant terrain changes in this area, we select the wind at the corresponding altitude level based on the altitude of each grid. However, we have already demonstrated in previous work for $NO_x$, CO, BC and CH4 emissions in polluted regions (Qin et al., 2023; Lu et al., 2025; Li et al., 2023; Li et al., 2025; Hu et al., 2024) that while this issue is relevant, that it is less impactful than using the gradient term correctly, and in the case of $NO_x$, the issue of retrieval error. This paper is the first attempt to bring the issue of retrieval error into the retrieval of CH4 emissions.

**the choice of mean values instead of medians,**

In the previous reply, we have explained the use of the median to replace the spatial filtering in the first step. Please refer to the fifth response to reviewer 3 in the previous version of the reply comments. In fact, we have always retained all of the data and can produce many different statistics, please see https://figshare.com/s/058f7f73953264e0d439.

**and the impact of different filtering procedures should be included to ensure robustness and reliability of the results.**

We have displayed the effects of the different filtering procedures clearly in Figures 3. This includes grid boxes versus none, filtering steps, etc. As shown in Figure S1,

Through the spatial filtering in the first step, we have filtered out invalid emissions from areas that are significantly affected by aerosols, albedo, and other factors. In the previous version, we have conducted a sensitivity analysis on the selection of the threshold for the first step of spatial filtering. After the first step of filtering, based on the theory of nonlinear propagation of satellite observation uncertainty in the gradient term, we carry out the second step of temporal filtering. We have already provided a detailed explanation in our previous response on how uncertainty is transferred to the emission results through the gradient term (refer to the fourth comment to reviewer 3 in the previous version of the reply comments).

**4. Comparison with existing estimates**

**• I strongly suggest comparing the results with bottom-up emission estimates and inversion-based estimates over the Permian Basin. This would provide valuable context and help validate the findings.**

In the previous version of the response, we have already provided the bottom-up emission inventory of the Waliguan region and found that there were sources missing in the inventory (EDGAR). The bottom-up emission inventory (EDGAR) of the Permian Basin is shown in Figure S6a, and Figure S6b shows the difference between our emission results and EDGAR, with the difference ranging from -285 to 3830 kg/h/grid, and the 95th percentiles of the differences is 2920 kg/h/grid. The EDGAR emission inventory is significantly lower than our results, in part due to many emissions grids missing from their dataset. We also compared the differences between the inventory of Cusworth et al. and EDGAR, as shown in Figure S6c. The difference range is from -3240 to 8440 kg/h/grid, and the 95th percentiles of the differences is 2450 kg/h/grid, which is close to the difference range between our results and EDGAR.

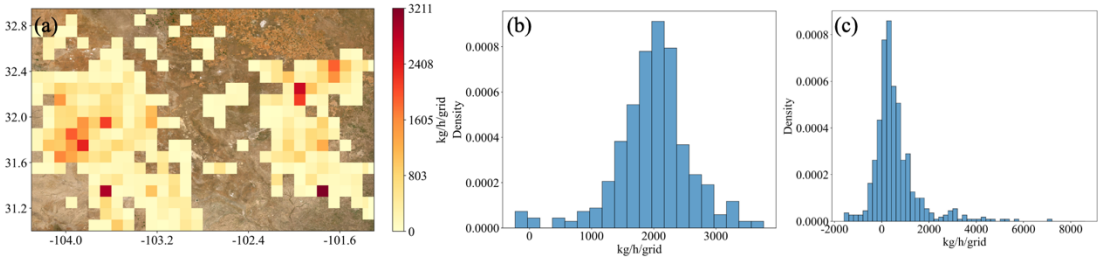


Figure S6: (a) The average annual emission flux of all sectors of EDGAR; (b) The difference between our retained valid emissions and EDGAR in 2019; (c) The difference between Cusworth et al., and EDGAR in 2019.

The above content has been supplemented in the manuscript, corresponding to lines 410 to 418: "To better compare with other inventories many of which use emissions in terms of mass per time per grid, we first convert emissions fluxes (μg/m²/s) into

emission rates over each entire TROPOMI grid (kg/h/grid). The EDGAR emission inventory over the Permian Basin is shown in Figure S6a, and Figure S6b shows the difference between our emission results and EDGAR, with the grid-by-grid ranging from -285 to 3830 kg/h/grid, and the 95th percentiles of the difference is 2920 kg/h/grid. The EDGAR emission inventory is significantly lower than our results, in part due to many emissions grids missing from their dataset, although their grid with the highest emission is still lower than our result, indicating that our approach does not have a high bias. We also compared the differences between the inventory of Cusworth et al. (2021) and EDGAR, as shown in Figure S6c. The difference range is from -3240 to 8440 kg/h/grid, and the 95th percentiles of the difference is 2450 kg/h/grid, which is close to the difference range between our results and EDGAR." The figure has been added to the supplementary file, corresponding to Figure S6

**5. Clarification on comparison with Cusworth et al. (2021)**

**• In Line 575, the manuscript states:**

**"It's clear that our results are larger, which is consistent with the observations made by aircraft having scan widths of 3 and 4.5 km, which is always smaller than our grid resolution of 0.05° × 0.05° (~5 km). This means that even if their scan crossed the center of our grid, our grid would still contain information outside of their scan width, and if the scan only crossed a small amount of our grid, then the grid would contain far more information."**

**Instead of only stating that the results are consistent, please quantify the differences between your results and those of Cusworth et al. (2021). For example, provide a plot of the standard deviation or relative differences to show how your estimates compare with theirs. This would strengthen the credibility of the comparison.**

Thank you very much for your suggestion. The construction of this model is based on the mass conservation equation. By fitting the ground observation data as physical constraints with the TROPOMI XCH4 data, a total of 900 sets of fitting coefficients were obtained. Note that these fitting coefficients are physical variables, specifically representing the time and weight scales of diffusion and advection. Based on these coefficients, 900 emission values can be estimated for each grid point. We take the average of the 450 values within the 20th to 80th percentile range as the final emission estimate for this point, and the uncertainty range is defined as the 20th and 80th percentiles of the emission value sequence. This range is meant to widely sample more standard observed conditions. Unfortunately, Cusworth did not provide high spatial and temporal resolution wind, pressure, and concentration data, or we could choose this range with more certainty (i.e. Lu, F et al., 2025).

According to the above logic, we have redrawn the scatter plots of our results and the observed emissions of Cusworth et al. The red dots in the scatter plot of Figure 8b represent the points where our emission results overlap with Cusworth (2021) in both time and space, with the R is 0.8, indicating a good match, and the MAE is 660 kg/h/grid. The black dots and blue dots respectively represent our effective emissions and the emission estimates of Cusworth et al. at other times in the grid where the red dots are located. We found that the methane emissions from these emission facilities varied significantly over time.

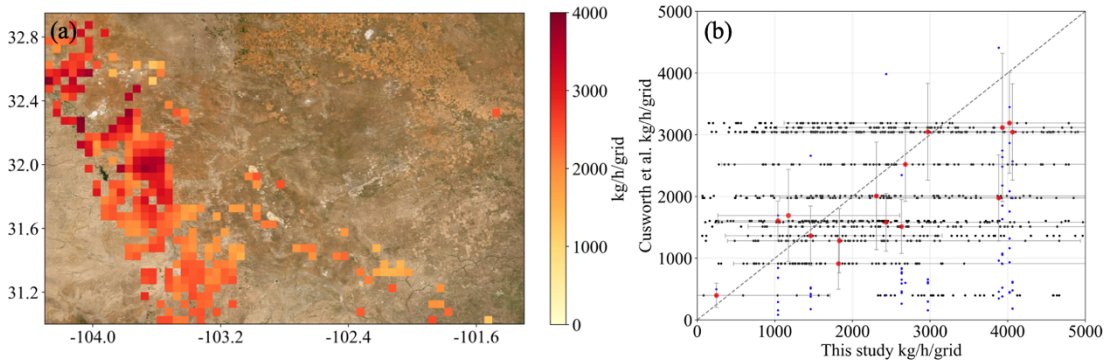


Figure 8: (a) Annual mean CH4 emissions (kg/h/grid) on grids which overlap spatially with an emission observed by observed by Cusworth et al. (2021); (b) The red dots in the scatter plot represent the points where our retained valid emissions overlap with those of Cusworth et al. (2021) in both space and time and the difference is within 2 times. The black and blue dots denote emission estimates from our study and Cusworth et al. (2021) respectively, which share spatial overlap but differ temporally. The gray error bars represent the associated uncertainties. Backgrounds of (a) is from Esri World Imagery.

The above content has been supplemented in the manuscript, corresponding to lines 435 to 440: "The red dots in the scatter plot of Figure 8b represent the points where our emission results overlap with Cusworth et al. (2021) in both time and space, with R=0.8 and MAE=660 kg/h/grid, both indicating a reasonable agreement. The black dots and blue dots respectively represent our effective emissions and the emission estimates of Cusworth et al. (2021) on the same grid, but during other days when Cusworth et al. (2021) does not have observations but TROPOMI does. We found that the methane emissions from these emission facilities varied significantly over time, indicating that TROPOMI allows for a better understanding of how emissions dynamics may change over time."

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