

## **Response to Referee #2**

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

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

### **General comments:**

The manuscript „Quantifying hard coal mines CH<sub>4</sub> emissions from TROPOMI and IASI observations using high-resolution CAMS forecast data and the wind-assigned anomaly method“ by Qiansi Tu et al., reports on a top-down approach to estimate methane emissions on the region scale. In their work the authors focus on the CH<sub>4</sub> emissions from hard coal mines in the Upper Silesian Coal Basin. Their emission estimation is based on applying a simple cone-plume-model and fitting the associated wind-assigned anomalies to enhancements in the XCH<sub>4</sub> data retrieved by satellite observations from TROPOMI and IASI over a period of three years. Simple, straight forward to apply approaches, as presented by the authors, to estimate CH<sub>4</sub> emissions on a local scale are highly relevant, especially in the light of recent COP 26 and the Global Methane Pledge that emerged from it.

The manuscript is well structured, but poorly written. The high amount of technical errors suggests that the authors have made an insufficient effort in proofreading, before submitting their manuscript to the journal. Nonetheless, I recommend the study as suitable for publication in ACP after the major revision has been addressed.

Thanks for pointing this out. We will do our best again to improve the language.

### **Specific comments:**

1. molec/s is a rather small-scaled unit for an observation-period of three years over a 100x100km region. Maybe kt/year is more suitable. Would also get rid of hard to read exponential nomenclature. Furthermore, in the introduction your use of units switches from kt/yr to TgCH<sub>4</sub>yr<sup>-1</sup>.

The units of molec./s and TgCH<sub>4</sub>yr<sup>-1</sup> are changed to kt/year as the referee recommended. The emission rate used in the cone-plume model has a unit of molec./s as typically used for remote sensing application for corresponding with column amounts (molec./area unit), so we would like to keep this unit in the text as well.

2. Sometimes it is not clear whether the CAMS GHG dataset or the CAMS emission inventory is being referred to (e.g. Lines 188 and 206; title of Fig. 5; caption Fig. 9; ...). This can be particular challenging, when the authors' emission estimates retrieved from the CAMS GHG dataset are compared to the CAMS emission inventory. I recommend using “CAMS-GLOB-ANT” throughout the text whenever referring to the emission inventory.

Thanks, we have changed “the CAMS emission inventory” to “CAMS-GLOB-ANT” to make it clearer.

- Line 21: “wind directions” Throughout the text, the division into wind regimes is designated differently. I think the designations wind regimes/segments/divisions are suitable, wind sectors/sections are not. Please adjust the text accordingly.

Thanks, the “wind sectors/sections” are changed to “wind regimes”.

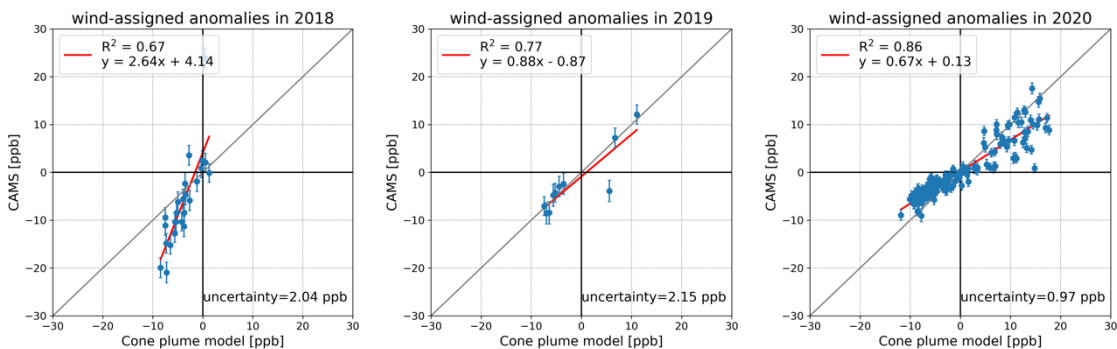
- Line 154: “This model is referred to as simple plume model”. A lot of plume models can be described as “simple”. I suggest referring to it as “cone plume model”. This would have the advantage that the designation is self-descriptive. If the authors want to stick with the term "simple plume model" for consistency with the earlier publication, that's fine with me.

The “cone plume model” is appropriate to represent the characteristic of our method. Changed accordingly.

- Line 162: To assume constant emission rates for three years is rather bold. According to the E-PERTR variations of a few percent are to be expected. Since the uncertainties in your estimate of emission rates are rather small, much of this might be due to interannual fluctuations. Please reconsider this statement and consider estimating emission rates for individual years.

The yearly emission strengths from CAMS-GLOB-ANT over the study area are 802.0 kt/year, 803.6 kt/year, and 807.4 kt/year in 2018, 2019, and 2020, respectively. These changes are 0.2% from 2018 to 2019, and 0.5% from 2019 to 2020, which is low and can be neglected.

The CAMS data are collocated to the TROPOMI+IASI data (see 11<sup>th</sup> comment), so the yearly amount of data is less and thus, poorer correlations of wind-assigned anomalies derived from the CAMS and the cone plume model are found:



The corresponding estimated emission rates are  $1597 \pm 92$  kt/year,  $689 \pm 118$  kt/year,  $512 \pm 12$  kt/year (i.e.,  $1.9E27 \pm 1.1E26$ ,  $8.2E26 \pm 1.4E26$ , and  $6.1E26 \pm 1.4E25$  molec./s) in 2018, 2019 and 2020, respectively. Higher uncertainties are found for the years 2018 and 2019. Therefore, using data for longer time periods results in better correlations and lower uncertainties.

6. In Eq. 2 you introduced  $\Delta XCH_4$  as the enhanced  $CH_4$  column. Later, especially in figures, you use  $\Delta XCH_4$  for the wind-assigned anomalies. Please introduce a distinctive notation for the wind-assigned anomalies to avoid confusion with the enhanced  $CH_4$ . See also the next comment for that.

We would like to thank the referee for pointing this out. The  $\Delta XCH_4$  in this study is only used to represent the enhanced  $XCH_4$ . The  $\Delta XCH_4$  used for the wind-assigned anomalies has been changed accordingly.

7. Line 163: Where does the empirical value of  $60^\circ$  come from? From your earlier publication I know that it comes from TROPOMI  $NO_2$  measurements, but this should be explained and cited here.

The sentence has been changed as the referee recommended:

“ $\alpha$  is the angle of the emission cone and has an empirical value of  $60^\circ$ , which has been derived from TROPOMI  $NO_2$  measurements (Tu et al., 2022)”

8. (1) Line 169-170: „For each wind sector, an averaged plume is computed and the difference of the two plumes are therefore the wind-assigned anomalies“. Above it is said that daily averaged plumes are calculated. Here, “averaged plume” refers to a plume averaged over all daily-plumes, which propagate NE/SW on daily-average. That means at that step you have two plumes for the entire three years. At each pixel your wind-assigned anomaly is calculated by  $\overline{XCH_4}_{SW}(i, j) - \overline{XCH_4}_{NE}(i, j)$ . Without checking your previous publication, I couldn't understand this. A few more equations would be helpful to explain your approach. Something like:  $\overline{XCH_4}_{SW/NE}(i, j) = \frac{1}{N_d} \sum_{d=1}^{N_d} XCH_{4,d}(i, j)$  with  $N_d$  = number of days and  $i, j$  SW/NE and wind-assigned anomaly =  $\overline{XCH_4}_{SW}(i, j) - \overline{XCH_4}_{NE}(i, j)$ .

(2) Please consider showing table A-1 here instead of the appendix.

(1) We have added the requested explanation in section 2.3 of the paper:

The daily plume from each point source (location at  $(i, j)$ ) is averaged over the daytime (8:00 UTC - 18:00 UTC):

$$\overline{XCH_4}_{(i,j)} = \frac{1}{11} \sum_{t=1}^{11} XCH_{4(i,j),t} \quad \text{Eq. 3}$$

these daily plumes are super-positioned over all point sources to obtain a daily plume ( $\overline{XCH_4}_{\text{daily}}$ ):

$$\overline{XCH_4}_{\text{daily}} = \sum_{s=1}^{N_s} \overline{XCH_4}_{(i,j),s} \quad \text{Eq. 4}$$

where  $N_s$  represents the number of the sources.

The wind distributions at different height levels (10 m, ~330 m, ~500 m) over the USCB region are presented in Figure 3. The wind speed increases with increasing altitude (see Table 1). The ERA5 wind is divided into two opposite wind regimes based on directions (e.g.,  $135^\circ$ - $315^\circ$  for SW and the rest for NE). For each wind regime, an averaged plume is computed:

$$\overline{XCH_4}_{4SW/NE} = \frac{1}{N_d} \sum_{d=1}^{N_d} \overline{XCH_4}_{daily,d} \quad \text{Eq. 5}$$

where  $N_d$  is the number of the days with SW wind or NE wind.

The difference between the two plumes is therefore the wind-assigned anomalies:

$$\text{wind - assigned anomalies} = \overline{XCH_4}_{4NE} - \overline{XCH_4}_{4SW} \quad \text{Eq. 6}$$

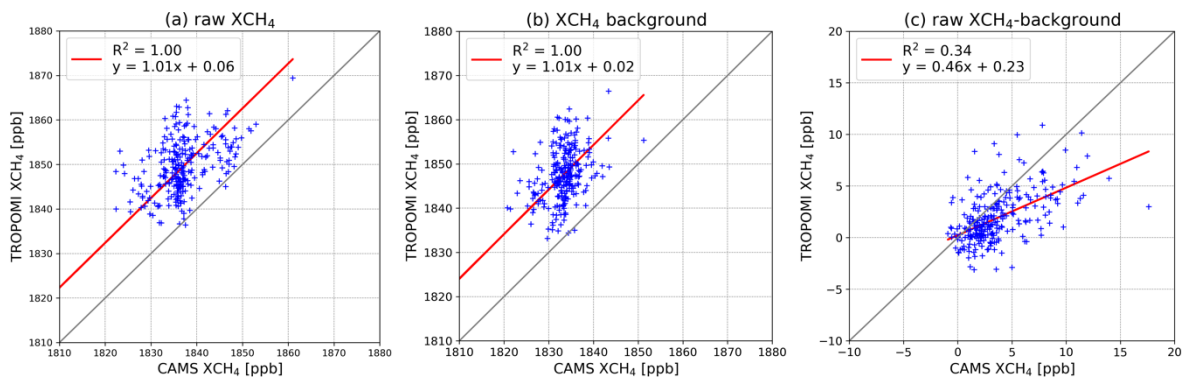
(2) The Table A-1 has been moved to section 2.3 as the referee recommended.

9. Line 175: Background removal is critical for correctly estimating the emission rate. From your earlier publication I know that the uncertainty in the background subtraction is included in the uncertainty of your enhanced  $XCH_4$  values. Please add a short statement about uncertainty in background subtraction in chapter 3.3. Moreover, I would be interested in how the background differs between CAMS and TROPOMI data.

A short statement about the uncertainty in background subtraction has been added:

“ $CH_4$  signal is weak compared to the background concentration which shows an increasing trend with obvious seasonality and strong day-to-day signals. It is necessary to remove the background signals before estimating the emission strengths. However, the imperfect elimination of the background introduces uncertainties, which can be determined by considering the deficits of the background model and the noise in the background (Tu et al., 2022). In this study, the uncertainties of the estimated strengths include the background uncertainties.”

The correlations of  $XCH_4$ , background, and the enhancement (raw  $XCH_4$  - background) between CAMS and TROPOMI are shown below:



10. Line 195: “The  $XCH_4$  anomalies (raw-background) and the wind-assigned anomalies are presented in Figure 5a and b, respectively”. Please change to  $XCH_4$  enhancement, to avoid risk of confusion to the wind-assigned anomalies. This would be more consistent as the term “enhanced” has already been used for background-free  $XCH_4$  in the context of Eq.2.

Thanks. These have been changed.

11. Line 196-197: I don't understand what you mean by: „Note, that the CAMS XCH<sub>4</sub> is coincided with TROPOMI XCH<sub>4</sub> for better comparison”. Were the CAMS data interpolated to the TROPOMI grid and accordingly the filtered TROPOMI grids are also missing for CAMS? But why are different grids missing for all plots displaying wind-assigned anomalies?

Thanks for pointing this out. There are nearly 400,000 data points for the CAMS XCH<sub>4</sub> during the study period over the study area due to its high spatial resolution ( $0.1^\circ \times 0.1^\circ$ ). It is hard to make the data processing program run for this high amount of data. Apart from that, the CAMS XCH<sub>4</sub> is used as an evaluation of our method in this study and to find the best ERA wind which is used for TROPOMI and TROPOMI+IASI data later. Thus, for each TROPOMI measurement, the nearest CAMS XCH<sub>4</sub> is selected as the co-located data, i.e., both data sets have the same amount of data (16,553). However, the TROPOMI data is further collocated with the IASI data (TXCH<sub>4</sub>), which results in a smaller data set (12,354). The different amounts of data lead to different grids missing.

To make the data sets to be consistent, we co-locate the CAMS to the TROPOMI+IASI data set. Figures (see 57<sup>th</sup> comment) and corresponding results are updated accordingly.

12. Line 213 ff: “To remove this influence, we calculate the tropospheric CAMS forecasts CH<sub>4</sub> (TXCH<sub>4</sub>) from the surface up to 7 km.” Why 7 km? The height of the tropopause surely changes over the course of the three-year observation period.

The following statement has been added to the paper:

"XCH<sub>4</sub> is affected by local surface emissions and a varying stratospheric contribution due to changes in the tropopause altitude (Liu et al, 2021; Schneider et al., 2021). This stratospheric contribution has to be taken into account, in order to be able to use XCH<sub>4</sub> for a reliable investigation of local surface CH<sub>4</sub> sources and sinks (Pandey et al., 2016). Our background removal method effectively accounts for the stratospheric contribution. To show this we apply the approach to CAMS forecasts of XCH<sub>4</sub> (which has a significant stratospheric contribution) and TXCH<sub>4</sub> (calculated from the CAMS forecast as the CH<sub>4</sub> averaged from surface to 7 km, which should have a very limited stratospheric contribution). The results are presented in Figure 5d-f. The CAMS TXCH<sub>4</sub> anomalies have similar distribution as CAMS XCH<sub>4</sub> anomalies, suggesting that our background removal approach reliably removes the stratospheric contribution."

13. Line 233-234: „In addition, the downwind plume is similar to the cone shaped plume in our simple plume model ...” What do you mean by similar? Just the spatial occurrence? As you use three different colormaps it is hard to judge by eye. It is clear, that the modelled cone-plumes result in XCH<sub>4</sub> enhancements which are smaller by a factor of two or even more, suggesting that the CAMS-GLOB-ANT emissions are too low.

Thanks, we have used the same colormaps for the first two figures (see 16<sup>th</sup> comment).

It is true that the XCH<sub>4</sub> enhancements derived from the modeled cone plumes are lower. This cone-plume model only considers a simple linear proportion of wind speed and emission strength. Huge

biases are expected in a simple day or in a short period. But these biases can be compensated over a long-term period.

The three kinds of XCH<sub>4</sub> enhancements on the example day have similar spatial patterns, which help to support the reasonable assumption of a cone-shape distribution. The corresponding sentence in the text has been changed.

14. Line 234: "... which implies our model assumption is reasonable." Either CAMS-GLOB-ANT has too small emissions, or the model generates a systematic bias. See comment above.

The sentence has been changed to:

"In addition, the spatial pattern of the downwind plume is similar to that of the cone-shaped plume, which implies our cone-shape assumption is reasonable."

15. Figure 5 correlation plots (c & f): I assume the gray line is the bisector. Please include your regression line. Please do so also for the other correlation plots in the manuscript.

The figures have been updated, see the 57<sup>th</sup> comment.

16. Figure 6: colormaps: Why a diverging colormap for wind speed? Is 4 m/s a representative mean value? If so, please indicate this in the caption, otherwise I would suggest a perceptually uniform sequential colormap. Also, please do not use the same colormaps for windspeed as for XCH<sub>4</sub> enhancements or wind-assigned anomalies.

Thanks, the figure and its caption have been updated.

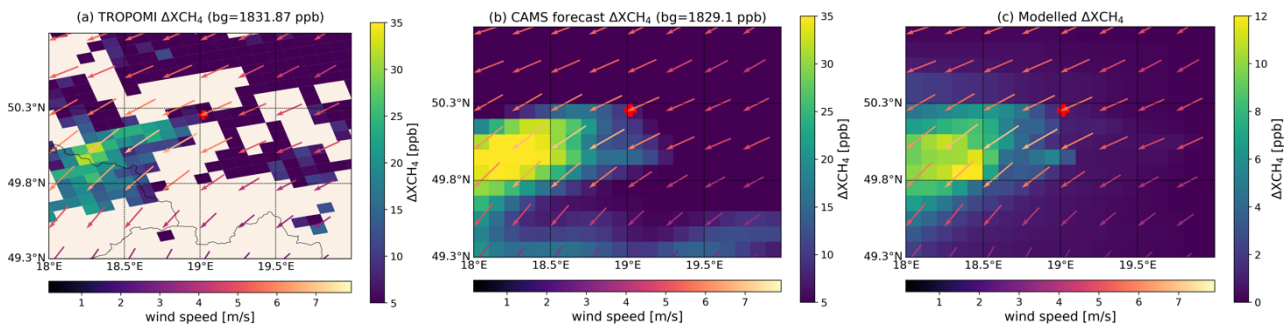


Figure 6:  $\Delta\text{XCH}_4$  together with the ERA5 wind at 12:00 UTC from (a): TROPOMI observations at 11:34 UTC, (b): CAMS forecast at 12:00 UTC, and (c): from the simple plume model (averaged over the daytime) based on the CAMS-GLOB-ANT inventory over the USCBA region on an example day (6 June 2018). The "bg" in the title of (a) and (b) represents the background, derived from the mean XCH<sub>4</sub> in the upwind region (50.3°-50.8° N, 19.5° -20.0° E).

17. Figure 6 colormaps: Why is TROPOMI transparent/shaded and the other two are not? Please have consistent colormaps for all plots. Especially the modeled plume has much smaller values than CAMS and TROPOMI. This becomes more difficult to see with the currently used colormaps.

The figures have been updated (see the 16<sup>th</sup> comment).

18. Figure 6: Do I understand correctly that CAMS forecast is at 12 UTC? Is TROPOMI also at 12 UTC? Is the modeled plume an average over 2018-06-06 or also at 12 UTC? Please clarify.

The CAMS forecast is at 12 UTC. The TROPOMI observation is around 11:34 UTC. The modeled plume is an average over 2018-06-06 (8:00 UTC – 18:00 UTC). This information has been added to the caption (see the 16<sup>th</sup> comment).

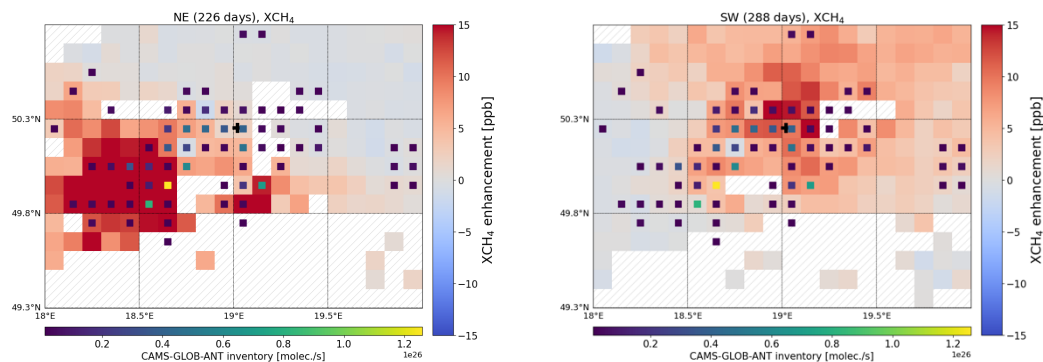
19. Figure 6: As you are showing snapshots of a specific time you should also give the respective value of the background term that has been subtracted. Either in the caption or the title.

The backgrounds for (a) and (b) have been added to the captions of the subfigures (see the 16<sup>th</sup> comment).

20. Figure 5 & 7: Why are the wind-assigned anomalies far more south-west than the CAMS forecasts/observations? Is the ERA5 wind wrong or is there a methodological error?

The CAMS data include both background and emissions from the sources. Compared to the background, the enhancements are tiny signals. Therefore, these background signals need to be removed before simulating the wind-assigned anomalies.

The CAMS enhancements (Figure5/7-a) contain all wind information (i.e., NE, SW, and rest). The wind-assigned anomalies are the difference between NE and SW. Moreover, there are more emission sources with higher emission rates in the SW direction of the study area. The plume, therefore, tends to cover the SW region. The enhancements for NE and SW wind are shown below:



21. Figure 5 & 7: I assume “CAMS emission (9.7E26 molec./s)” is the sum of all sources in the CAMS-GLOB-ANT inventory and used as an a priori value for the calculation of the wind-assigned anomalies, is this correct? Please clarify in the caption and do not repeat as a title for every single plot. Same for Figure 7.

The referee is right that we use the CAMS-GLOB-ANT inventory as the a priori values and locations of the sources for calculating the wind-assigned anomalies.

Figures have been updated (see the 57<sup>th</sup> comment).

22. Line 253: “The TROPOMI+IASI result has a slightly higher uncertainty than the TROPOMI result“. Please remove “slightly”. The uncertainty is more than a factor 4 higher.

Changed accordingly. We also removed “slightly” from the sentence on line 344 in the conclusion.

23. Line 263: „... the emission rate uncertainties of using  $XCH_4$  or  $TXCH_4$  are insignificant compared to the estimated emission rates.“ Please change “insignificant” to “small” or something equivalent. If uncertainties were insignificant, they would not need to be reported.

Changed accordingly.

24. Line 287: “Considering the height of the Planetary Boundary Layer (PBL), we use the ERA5 wind at 500 m above the ground (Figure 3c)” I don’t understand what you mean by that. The PBL height changes during the day and year. How is 500 m related to the PBL height? In your abstract you give the emission rates for a height of 330 m.

The enhancement in  $XCH_4$  that is being used to estimate the emission rates is composed of  $CH_4$  molecules that have been advected in different heights. To me it is unclear why a certain height should be more representative than the other (at least in the PBL). Shouldn’t an average wind speed over the entire vertical spread of the PBL be used. This would of course massively increase the uncertainty of your estimation. Please comment.

The PBL thickness over the study area ranges from 700 m to 1.5 km over the daytime in summer, which is about 1 km on average (Luther et al., 2019; Krautwurst et al., 2021). Winds typically increase above and decrease below the middle of the PBL. Thus, we chose the altitude (~500 m) in the center of PBL for an explicit averaging over the PBL thickness, and wind at 500 m is used to investigate the uncertainty of wind (see the 27<sup>th</sup> comment).

This sentence has been rephrased to:

“Assuming that the height of the Planetary Boundary Layer (PBL) is typically less than a kilometer, we use the ERA5 wind at 500 m above the ground (Figure 3c) for describing the transport of methane released in the study region.”

25. Figure 9: The grids that are shown are a superposition of the days on which, in the daily average, the wind blew in the respective narrow-regimes, right? If so, this should be explained once more in text in section 3.3.2. I don’t understand where all the missing data points originate from. If for example the cone-plumes are never advected into the narrow wind-regime at NW-SE, then, for the respective grid, the calculation is  $0-0=0$ , isn’t it? If so, you can of course filter these grids to make a distinction to cases where  $XCH_{4NW}$  and  $XCH_{4SE}$  are equal but not zero. If this is being done please explain it somewhere in the text.

If a wind direction dominates 60% of records for one day, i.e., if the wind direction belongs to one specific area ( $NW_{1/2}/SE_{1/2}$ ) more than 60 % of the daytime (08:00–18:00 UTC), then this predominant wind direction is selected for that day. When narrow wind regimes are used, the number of days with  $NW_{1/2}$  (or  $SE_{1/2}$ ) wind is much less than that with NW wind, i.e., fewer data points for  $NW_{1/2}$  (or  $SE_{1/2}$ ) regimes. Moreover, to eliminate the biases, we select the grids with more than 10 measurements. The fewer data points result in more missing grids here.

26. Line295 ff: „The final estimated emission strength is weighted by the number of the valid binning data in the plume maps under different wind regimes (i.e. 171 for narrow NE-SW and 26 for narrow NW-



SE, respectively).“ I do not understand the weighting. Are there 171+26 days in total? The emission rate of  $9.8E26$  molec./s from NE-SW regime is being weighted with 171 days and the  $14.0E26$  molec/s with 26 days. Result is then  $10E26$  molec/s which is given in line 303? If I understand correctly please insert the information that by “number of the valid binning data” you mean “number of days on which, on average, the wind blew in the respective wind-regime.”

We used the number of valid grids in the wind-assigned anomalies as the weighting, but this might be not fully accurate. The total days for  $NE_{1/2} - SW_{1/2}$  (115 days) and for  $NW_{1/2} - SE_{1/2}$  (71 days) should be used as the referee recommended.

The sentence has been rephrased as follows:

“The final estimated emission strength is weighted by the number of days on which, on average, the wind blew in the respective wind regime (i.e., 115 days for  $NE_{1/2} - SW_{1/2}$  and 71 days for  $NW_{1/2} - SE_{1/2}$ , respectively).”

Since the CAMS data are collocated to the TROPOMI+IASI TXCH<sub>4</sub> (see the 11<sup>th</sup> comment), the corresponding results in the paper are also changed:

“The estimated emission rate is about  $773 \pm 13$  kt/year ( $9.2E26 \pm 1.6E25$  molec./s) for the  $NE_{1/2} - SW_{1/2}$  field. This indicates that the effect of the segment in the wind field coverage is negligible when there are enough measurements. The use of  $NW_{1/2} - SE_{1/2}$  wind fields yields an emission strength of  $1176 \pm 109$  kt/year ( $1.4E27 \pm 1.3E26$  molec./s). The higher uncertainty is probably due to less measurements in these wind fields. The weighted rate is therefore about 927 kt/year ( $1.0E27$  molec./s), 13.4% higher than based on the wider NE-SW wind regime (Sec. 3.1).”

27. (1) Line 352 ff: “However, their speeds decrease by 19% at 10 m and increase by 32% at 500 m, which results in higher emission rates by -23% and 13 %, respectively.” How can that be? Wind-speed is linear in the calculation of  $\epsilon$ , isn't it? Accordingly, the emission rates should also be -19% & +32%. Please comment.

(2) Furthermore, “higher emission rates” is not correct for describing a decrease and an increase. Please rephrase.

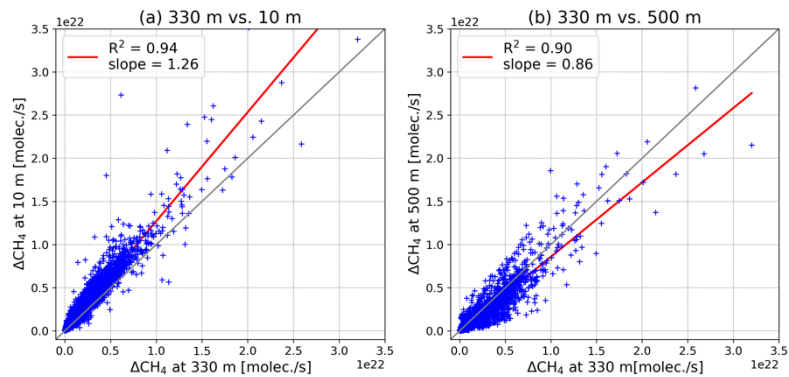
(1) We used TROPOMI data over a larger area before applying the coincidence criteria with the wind. Since we decided to collocate CAMS forecast data to the TROPOMI+IASI data set (see the 11<sup>th</sup> comment), the wind information is slightly changed (see Table 1 below). The weighted-average wind speed at each level is calculated based on the days at each wind regime. The wind speed reduces by 20% at 10 m, and increases by 32% at 500 m, compared to that at 330 m. The corresponding changes in the estimated emission strengths are -25% and 13%, respectively. These values and Table 1 have been updated in the paper.

Table 1: Number of days and the averaged wind speed ( $\pm$  standard deviation) per specific wind area in daytime (08:00 UTC – 18:00 UTC) at different vertical levels from November 2017 to December 2020 over the USCB region. The days for the three-year average coincide with the TROPOMI overpass days.

	NE / $>315^\circ$ or $<135^\circ$		SW / $135^\circ - 315^\circ$	
	Number of days in total (%)	Averaged wind speed $\pm$ standard deviation ( $\text{m s}^{-1}$ )	Number of days in total (%)	Averaged wind speed $\pm$ standard deviation ( $\text{m s}^{-1}$ )
10 m	39.1	$3.2 \pm 1.5$	56.9	$3.4 \pm 1.6$
~330 m (975 hPa)	38.7	$4.1 \pm 2.2$	56.9	$4.3 \pm 2.3$
~500 m (950 hPa)	38.7	$5.0 \pm 2.7$	57.3	$5.9 \pm 3.5$

“The wind speed is linear in the calculation of  $\epsilon$ , but the wind speeds do not all linearly change for each grid and for each time at different levels. This results in unequal changes between the wind speed and the enhanced columns, and later unequal changes in the estimated emission strength. In addition, the simple cone plume model introduces biases, i.e., the enhanced column in the downwind is set to zero when its location is out of the cone angle ( $60^\circ$ ). Slight changes in the wind directions might result in a huge difference in the enhanced columns.” This statement has been added to the text.

The figure below shows the correlation plots for the enhanced columns at 10 m and 500 m, compared to the ones at 330 m. The changes in the enhanced columns are -26% and 14%, which are similar to the changes in the estimated strengths.



(2) the sentence has been changed to:

“However, their speeds decrease by 20% at 10 m and increase by 32% at 500 m, which results in changes in the emission rates by -25% and 13 %, respectively.”

## Technical comments

28. Please consider perceptually uniform sequential colormaps, especially for figures 2, 4 and 6. Diverging colormaps are helpful in displaying differences, which in your case would only make them suitable for plotting wind-assigned anomalies and XCH<sub>4</sub> enhancements. If you stick to the red-blue diverging colormap for the anomalies consider hatching the grids with missing data. At the moment they are easily mistaken for value 0.

If you are using python to generate plots you might have a look here:

[https://matplotlib.org/2.0.2/examples/color/colormaps\\_reference.html](https://matplotlib.org/2.0.2/examples/color/colormaps_reference.html)

Thanks, your comment is helpful for improving the figures. We modified the figures and in the appendix we added a zoom into the area shown in Figure 4(b).

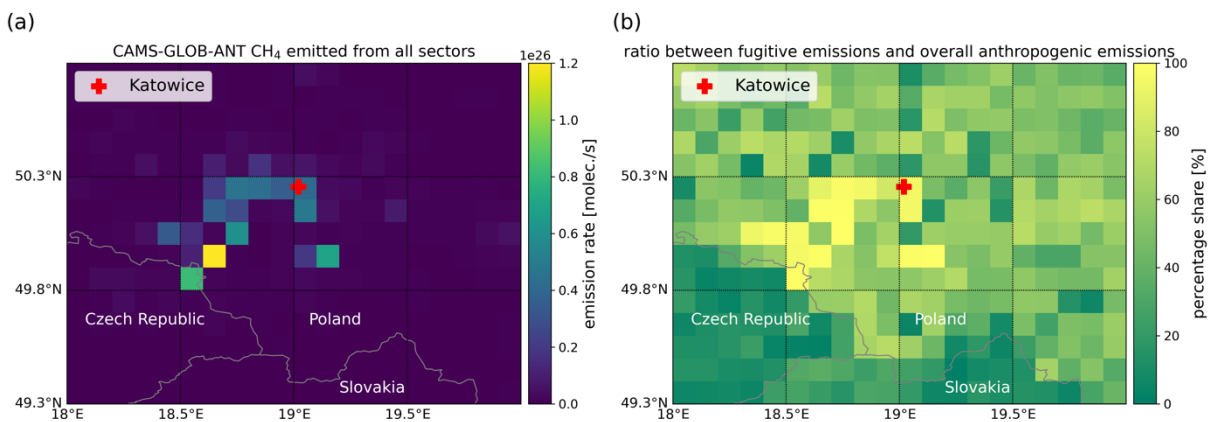


Figure 1: Spatial distribution of (a) the CAMS global anthropogenic emissions from all sectors and (b) percentage share of the fugitive emissions compared to the overall anthropogenic emissions over the USCB region on a  $0.1^\circ \times 0.1^\circ$  latitude/longitude grid. The fugitives are the dominant CH<sub>4</sub> sources.

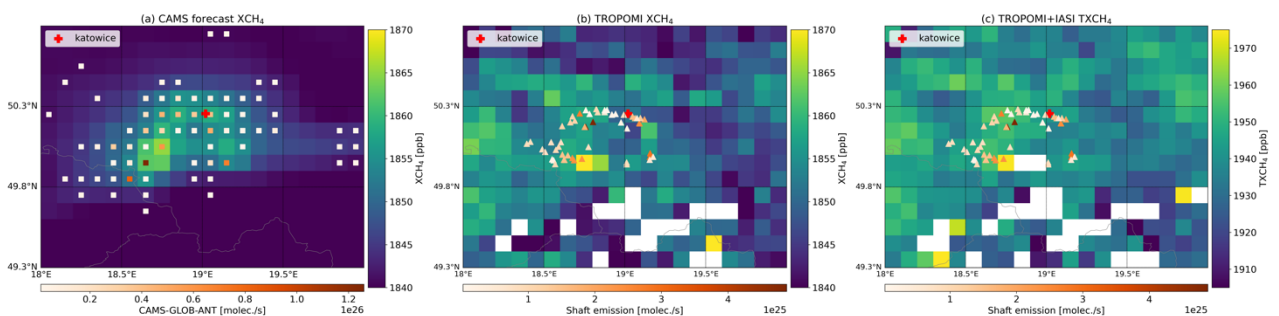


Figure 4: averaged (a) CAMS forecast XCH<sub>4</sub>, (b) TROPOMI XCH<sub>4</sub> and (c) TROPOMI+IASI TXCH<sub>4</sub> in the USCB region on a  $0.1^\circ \times 0.1^\circ$  latitude/longitude grid during November 2017-December 2020. The square and triangle symbols represent the locations of CAMS-GLOB-ANT sources (for a better viewing, only the emission strengths larger than  $1E24$  molec./s are shown here) and the active coal mine shafts from the CoMet inventory (Gałkowski et al., 2021), respectively. Different colors denote the amount of emission rates. The white grids represent no data from TROPOMI or the number of the points in the grid less than 5. A zoom version of panel (b) is shown in the appendix (Figure A-2). Note, a different colorbar has been used in panel (c).

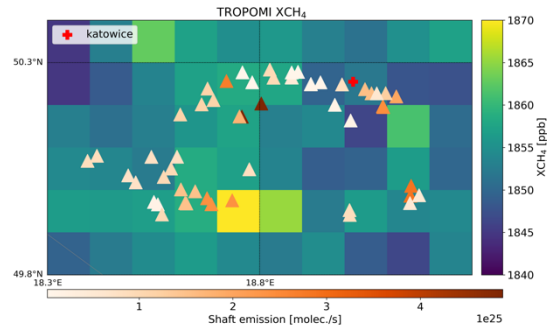


Figure A- 2: A zoomed version of Figure 4(b).

For Figure 6, see the 16th comment.

29. For many citations there is a dot missing after “et al”.

Thanks, changed accordingly.

30. For almost all figures the labeling is way too small. Please increase the font size corresponding to the text.

Thanks, the figures have been updated.

31. Line 12: „Intensive coal mining activities are in the Upper Silesian Coal Basin (USCB) in southern Poland, resulting in large amounts of methane (CH<sub>4</sub>) emissions.” Maybe shift the “are” in front of “resulting”.

Changed accordingly.

32. Line 13: “Annual CH<sub>4</sub> emission reached to 448 kt according to the European Pollutant Release and Transfer Register (E-PRTR, 2017).” Please remove the “to” or change to “... reached up to 448 kt ...”

Changed accordingly.

33. Line 14-15: “As a CH<sub>4</sub> emission hot spot in Europe, it is of importance to investigate its emission sources and accurate emission estimates”. Maybe insert “make” in front of “accurate emission estimates”

Changed accordingly.

34. Line 16: “column-averaged dry-air molar fraction observations of CH<sub>4</sub>”. Please change to “mole fraction observations”.

Changed accordingly.

35. Line 16-20: It is a rather long sentence. Maybe split up.

The sentence is split up into two sentences as the referee recommended:

“In this study, we use satellite-based column-averaged dry-air molar fraction observations of CH<sub>4</sub> (XCH<sub>4</sub>) from the TROPospheric Monitoring Instrument (TROPOMI) and tropospheric XCH<sub>4</sub> (TXCH<sub>4</sub>) from the Infrared Atmospheric Sounding Interferometer (IASI). In addition, the high-

resolution model forecast XCH<sub>4</sub> and TXCH<sub>4</sub> from the Copernicus Atmosphere Monitoring Service (CAMS) are used to estimate the CH<sub>4</sub> emission rate averaged over three years (November 2017 to December 2020) in the USCB region (49.3° - 50.8° N and 18° - 20° E).”

36. Line 27 ff: “... with using the Carbon dioxide and Methane (CoMet) inventory ...” What information is actually used from the CoMet inventory? As you report your emission estimates in the next sentence I assume that, here, you just take the locations of the shafts. Please be more specific, as the CoMet inventory also reports emission rates of individual shafts.

We used both the location and the proportion of the emission rate for each shaft in the total emissions, so as for the CAMS-GLOB-ANT. This information is added:

“Using the CAMS inventory (CAMS-GLOB-ANT) as the a priori knowledge (location and the proportion of the emission rate for each source in the total emissions) of the sources, together with ERA5 wind at 330 m, the wind-assigned XCH<sub>4</sub> anomalies for two opposite wind directions are calculated, which yields an estimated CH<sub>4</sub> emission of 815 kt/year ( $9.7E26 \pm 1.5E25$  molec./s) for CAMS XCH<sub>4</sub> and 798 kt/year ( $9.5E26 \pm 1.3E25$  molec./s) for CAMS TXCH<sub>4</sub>.”

37. Line 28: Not sure what is meant by “performed”? An inventory is not performed. How about “... from 2018”, “... covering the year 2018”, “issuing the year 2018” or something equivalent.

Thanks, the sentence has been changed to:

“This wind-assigned method is further applied to the TROPOMI XCH<sub>4</sub> and TROPOMI+IASI TXCH<sub>4</sub> with using data from the Carbon dioxide and Methane (CoMet) inventory derived for the year 2018.”

38. Line 34-35: “When using different wind coverage and different wind segmentation, an uncertainty of 4.2% and -2.1% is obtained, respectively”. How is an uncertainty negative? Maybe uncertainty is not the adequate word.

The referee is right that this word is not properly used. The “uncertainty” here meant the changes in emission strength when different wind information was used. The sentence has now been modified to:

“When using different wind coverage and different wind segmentation, the estimated emission strengths change by 4.2% and -2.1%, respectively.”

39. Line 40-42: This sentence is hard to read. In my opinion the word “and” is used too often. I think in “... and waste disposal ...” you can remove it.

The sentence has been changed as the referee recommended:

“Methane sources induced by anthropogenic activities include fossil fuel production and use (e.g., coal mining, gas/oil extraction), waste disposal, and agriculture, which in total accounts for about 60% of the total CH<sub>4</sub> emissions (Saunio et al., 2020).”

40. Line 43: “,... to the atmosphere CH<sub>4</sub> level are still ...”. This seems off. Maybe change to “atmospheric” or “atmosphere’s”

Changed accordingly.

41. Line 75: "... data sets provide a large coverage and long-term XCH<sub>4</sub>/TXCH<sub>4</sub> observations, which helps to better estimate CH<sub>4</sub> emission ..." I guess it should be "help", not "helps".

Changed accordingly.

42. Eq. 1: The square root should also include the numerator. The calculation of the standard deviation is trivial. If you want you can remove the equation.

The equation has been removed as the referee recommended.

43. Line 111: "... emissions from ships with a magnitude of 19 are much lower ...". What do you mean by "magnitude"? Do you mean the "count" of ships?

It should be the "orders of magnitude of the emissions". The sentence has been changed to:

"The emissions from the sectors "agriculture soils" and "solvents" are zeros. The CH<sub>4</sub> emitted from ships has 19 orders of magnitude, which are much lower than the other sectors".

44. Line 113: "Compared to its high amount, the seasonal variations of the fugitives sector can be ignored." Sounds off to me. Maybe avoid "high amount" when referring to emission rates. I suggest "The seasonal emission variations of the fugitive sector are minor and can be ignored" or similar.

The sentence has been changed as the referee recommended:

"The seasonal variations of the fugitives sector are minor and can be ignored."

45. Line 113 – 119: From here on, the paragraph is no longer stringent to me. What the authors are basically saying is that the fugitive sector is dominant in the USCB. As the fugitive sector has minor seasonal variations they do not consider them. I suggest the following restructuring from line 111 onwards: "Thus, these three sectors are not shown here. The sources from agriculture livestock ( $1.7E25 \pm 4.0E25$  molec./s) amount only 4% of the total emissions in this region. The dominant CH<sub>4</sub> sources in this region are fugitive sources from from energy production and distribution (e.g. fuel use). With a mean value of  $7.9E26$  molec./s and a standard deviation of  $2.2E25$  molec./s they account for 82% of the anthropogenic CH<sub>4</sub> emissions in the CAMS-GLOB-ANT inventory ( $9.7E26$  molec./s in total). This becomes particular visible in the spatially overlapping distribution within the USCB (see Figure 2). The seasonal emission variations of the fugitive sector are minor and can be ignored. Therefore, we apply the three-year mean of total emissions at grids with significant emissions without considering seasonal variations in the simple plume model (see Sect. 2.3)"

We would like to thank the referee for rephrasing the sentences. The sentences are changed as the referee recommended.

46. Figure 1: The coloring is highly unfortunate. In the legend the "Fugitives" is listed last and easily mistaken for "Off road transportation". Please list "Fugitives" first and change the color for "Off road transportation".

The figure is updated to:

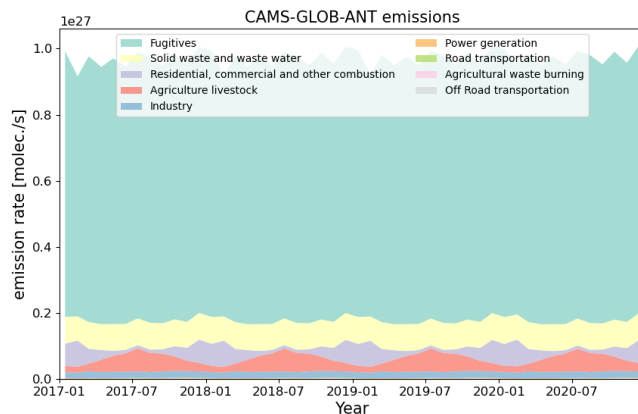


Figure 2: Stacked area plot for different sectors of the monthly averaged CAMS global anthropogenic emissions ( $>1E20$  molec./s) in the USCB region for 2017-2020 (<https://permalink.aeris-data.fr/CAMS-GLOB-ANT>, last access: 22 December 2021. Granier et al., 2019).

47. Figure 2: I assume that the barely visible gray lines are the borders to the Czech Republic and Slovakia. Please increase the resolution of the basemap so that the borders can be recognized as such. For a better orientation you might consider inserting country abbreviations.

At first glance, the two heatmaps look identical, which is of course the point being made here. However, I'm a bit unsure about the gain of information when two nearly identical images are shown side by side. Perhaps a heatmap of the percentage shares of fugitive emissions compared to overall anthr. emissions would be better. Please comment.

Thanks, the figure in terms of percentage share helps readers to better understand that fugitive emissions are the dominant sources. The figures have been updated (see the 28<sup>th</sup> comment)

48. Line 146: Comma before “which”

Changed accordingly.

49. Line 147: “... it is able ...” What does “it” refer to? I guess it refers to the combined product, which you introduce as such only in the following sentence. I suggest “... we are able to ...”.

Changed accordingly.

50. Line 154: You reference the figure 2 from a previous publication. In my opinion it would be beneficial to actually show the figure again.

The figure has been added in the Appendix.

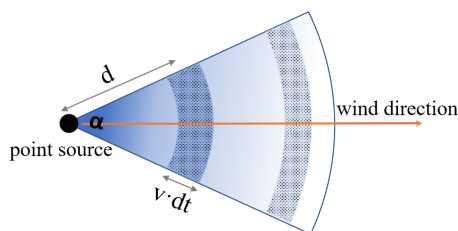


Figure A- 1: Sketch of the simple plume model used to explain the CH<sub>4</sub> emission estimation method. The methane at the point source is distributed along the wind direction (wind speed:  $v$ ) in the cone-shaped area with an opening angle of  $\alpha$ . The point source emits the methane at an emission rate of  $\epsilon$ . We assumed the methane molecules are evenly distributed in the dotted area A, and the distance from area A to the point source is  $d$ . Therefore, the emitted methane in  $dt$  time period equals to the amount of methane in the area A. It yields the equation  $\epsilon \times dt \approx \Delta column \times \frac{\alpha}{\pi} \times \pi \times d \times v \times dt$ . This figure is adopted from Tu et al. (2022).

51. Line 159 and Eq. 2: The indices  $i$  of  $(x_i, y_i)$  should be subscripted.

Changed accordingly.

52. Figure 3: The individual plots in Figure 3 will separately be referred to with a, b and c (e.g. Line 288). Please add a numbering to the plot or change reference in the text to left, middle and right.

Changed accordingly.

53. Figure 4: Please increase the size of the squares for the CAMS-GLOB-ANT sources. The color of the sources is so difficult to distinguish. An increase in the size of triangles for the CoMet sources would also be beneficial, although this might be more difficult as the triangles overlap. If possible, please improve the visibility. If not, you might consider providing a zoom to the shafts in a separate subsection, which was suggested by Referee#1.

Changed accordingly. See the 28<sup>th</sup> comment.

54. Figure 4: In the caption it says “during November 2017-December 2020”. Does this mean the displayed XCH<sub>4</sub>/TXCH<sub>4</sub> data are an average of this period? If so, please indicate this in the caption. Otherwise, please specify the displayed day.

The data in the figure represent the average. The information has been added in the caption as the referee recommended (see the 28<sup>th</sup> comment).

55. Figure 4: I assume that the white grids are missing data. Please indicate this in the caption. Moreover, the color choice is unfortunate, as it is missing values are difficult to distinguish from the mid-range values in the colorbar. Please see my earlier comment regarding colormaps.

The figure is updated (see the 28<sup>th</sup> comment).

The sentence has been added in the caption:

“The white grids represent no data from TROPOMI or the number of the points in the grid is less than 10.”

56. Figure 4: For a better comparison please consider using an identical colormap for a) and b). The TROPOMI & IASI data product has of course higher values. If the colorbar consists of the same colors, please indicate the shift in values in the caption.

The figure has been updated (see the 28<sup>th</sup> comment).

57. Figure 5 & 7: If a diverging colormap is being used, please center the colorbar to the value 0. Please use the same colormap for all four plots.



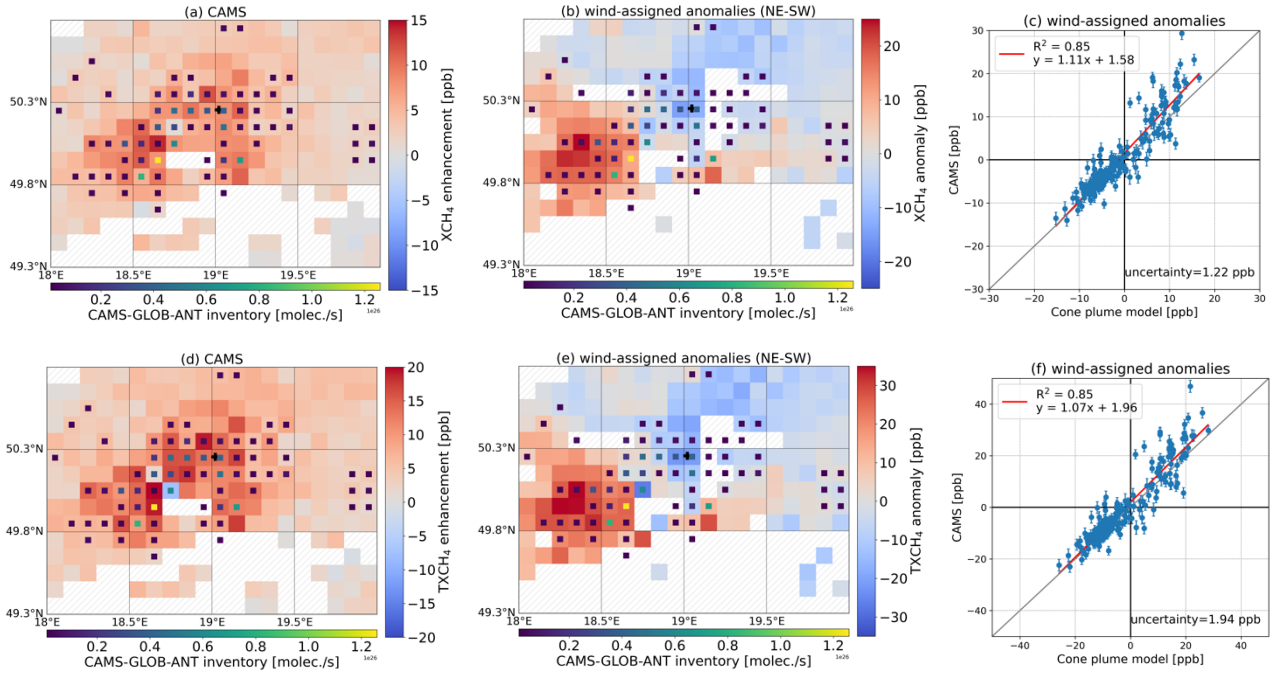


Figure 5: (a)-(c): CAMS XCH<sub>4</sub> enhancement anomalies (XCH<sub>4</sub>-background), the wind-assigned anomalies (NE-SW), and correlation plot of the wind-assigned anomalies between CAMS and the cone plume model with using the CAMS-GLOB-ANT inventory (9.7E26 molec./s in total) and ERA5 wind at 330 m during November 2017-December 2020 over the USCB region. (d)-(f): the same as for the upper panel but for CAMS TXCH<sub>4</sub> (colorbars in (d) and (e) are different from that for XCH<sub>4</sub>). The square symbols represent the locations of the CAMS-GLOB-ANT (>1E24 molec./s) inventory and different colors denote the amount of emission rates. The hatched areas in (a)-(b) and (d)-(e) represent no data in these grids. The uncertainties in (c) and (f) represent the mean error bars, i.e., error propagation of the background uncertainty and the CAMS standard deviation.

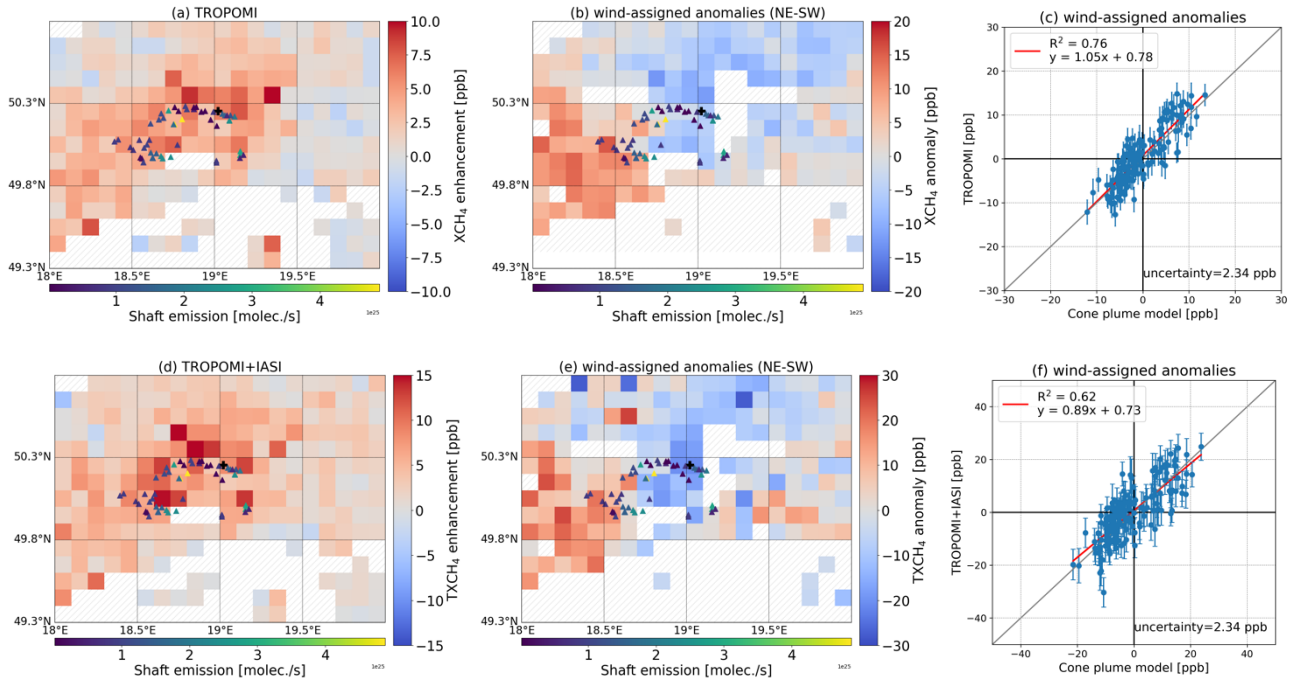


Figure 7: Similar to Figure 5, but for (a-c) TROPOMI XCH<sub>4</sub> and (d-f) TROPOMI+IASI TXCH<sub>4</sub>. The a priori knowledge of sources are based on the CoMet inventory (Gałkowski et al., 2021). The triangle symbols represent the locations of the active coal mine shafts and different colors denote the amount of emission rates.

58. Figure 5 & 7: Please avoid the term “anomalies” if you are not referring to wind-assigned anomalies. Rather use “enhancement” as suggested in an earlier comment

Changed accordingly. See the 57<sup>th</sup> comment.

59. Figure 5 & 7: Please do not repeat the identical title for multiple plots in the figure. I Suggest to name the lines on the left with [XCH<sub>4</sub>, TXCH<sub>4</sub>].T. Name the columns with [CAM5, modelled (cone-plumes + ERA5), correlation plot]. Instead of “modelled (cone-plumes + ERA5)” you could of course choose a term of your choice. Something like “wind-assigned anomalies (SW- NE)” or similar would be fine too.

Changed accordingly. See the 57<sup>th</sup> comment.

60. Figure 5 & 7: The colorbar-label for the left plots (a & d) and the middle plots (b & e) are currently the same. The left plots are displaying XCH<sub>4</sub> enhancements (i.e. XCH<sub>4</sub> – background), the middle plots are displaying wind-assigned anomalies. Please correct the colorbar labels.

Changed accordingly. See the 57<sup>th</sup> comment.

61. Figure 5 & 7 caption and title of middle plots: “... the wind-assigned anomalies (NE-SW) ...” Shouldn’t it be “SW-NE”? Otherwise the positive values should be in the NE.

The NE wind results in higher values in SW region, vice versa. Therefore, NE-SW results in positive values in SW region. See the figures in 20<sup>th</sup> comment.

62. Figure 5 & 7 correlation plots (c & f): Please remove the title. The information is already given in the axis’ labels. Also, as mentioned before, the use of ΔXCH<sub>4</sub> is not consistent.

Changed accordingly. See the 57<sup>th</sup> comment.

63. Line 216: “9.1E24 ± 1.2E24 molec./s” I guess there is a typo in the exponent.

Thanks, it should be 9.1E26 ± 1.2E24 molec./s (i.e., 798 ± 11 kt/year). Changed accordingly.

64. Line 227: "Figure 6 illustrates the enhance XCH<sub>4</sub> (raw XCH<sub>4</sub>-background in the upwind) distribution ..." Please correct “enhance” to either “enhanced XCH<sub>4</sub>” or “XCH<sub>4</sub> enhancement”. Why is “in the upwind” specified? From the explanation in the appendix of your earlier publication the background determination is not limited to the upwind.

The “enhance” has been changed to “enhanced”.

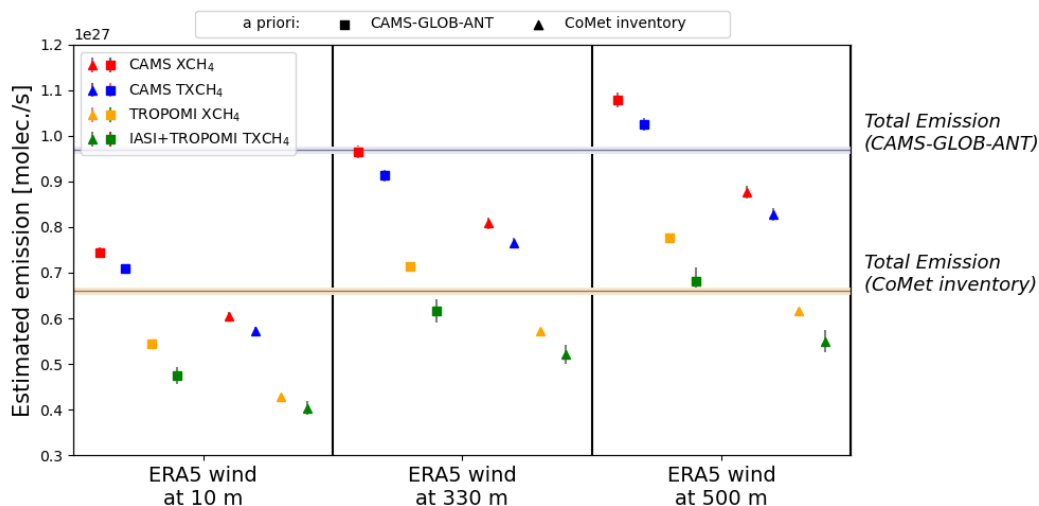
The referee is right that we determine the background is not limited to the upwind, but based on long-term observations, from which the seasonal cycle, linear increase, etc. are removed. Here, the enhancements from three different datasets on a single day are shown as examples, and thus, we use the XCH<sub>4</sub> in the upwind as the background.

65. Line 244: "... anomalies show high amounts around the areas ..." To me "amounts" sounds off. Please consider something like "high concentrations", "high methane content" or something similar.

The "high concentrations" is used, as recommended by the referee.

66. Figure 8: Please be precise in the labeling of the horizontal lines, i.e. "Total Emission (CAM5- GLOB-ANT)", "Total Emission (CoMet inventory)".

We would like to thank the referee for these comments to improve the figure. Changed accordingly.



67. Figure 8: Please remove the shaded background and instead add a legend: "a priori: squares CAM5-GLOB-ANT, triangles CoMet inventory", or something similar. If plotted among each other triangles and squares are easier to compare.

Changed accordingly. See the 66<sup>th</sup> comment.

68. Figure 8: The error bars are very small, as you mention in the caption. Nevertheless, please use either a uniform color, such as black or gray, or simply the color of the respective marker. At the moment it seems like they change colors randomly.

Changed accordingly. See the 66<sup>th</sup> comment.

69. Line 280: „Here we investigate the wind uncertainties ...“. Please insert a comma after “here”

Changed accordingly.

70. Line 284: "Compared to the wind at 330 m, the wind distributions are similar ..." Please specify, in the whole text, that you are referring to the distribution of wind directions.

Changed accordingly.

71. Section 3.3.2: Since the designation SW and NE were used previously and now SW and NE are still used for narrow, the text is a bit confusing. Either <sub>narrow</sub> is always subscripted consequently, as is being done in the caption of Fig. 9, or, alternatively, the subscripts SW<sub>1/2</sub> or SW<sub>1/4</sub> could be used to specify whether the wind field is divided into halves or quarters.

Using the subscripts as the referee recommended, is a better way to make the text clearer. We use NE<sub>1/2</sub> for 0°-90°, SW<sub>1/2</sub> for 180°-270°, NW<sub>1/2</sub> for 270°-360°, and SE<sub>1/2</sub> for 90°-180°.

72. Figure 9 and text in section 3.3.2: Isn't it "SW-NE" instead of "NE-SW"?

It should be NE-SW (see the 61<sup>st</sup> comment).

73. Line 310: "The wind category here is based on its predominant wind fields over the USCB region ...". Please change "its" to "the" or rephrase.

The sentence has been changed as the referee recommended:

"The wind category here is based on the predominant wind fields over the USCB region and is divided into two opposite sectors (SW and NE)."

74. Line 311: "To investigate its uncertainty, we apply another kind of segmentation." What does the "its" refer to? Please change to "To investigate the effect of the segmentation on the uncertainty in the emission rate estimation, we additionally apply another kind of segmentation" or similar.

The sentence has been changed as the referee recommended:

"To investigate the effect of the segmentation on the uncertainty in the emission rate estimation, we additionally apply another kind of segmentation: N (<90° or >270°) and S (90° - 270°) categories."

75. Line 335: "To investigate the CH<sub>4</sub> emissions from this hot spot, the CoMet campaign was performed in 2018. Locations and emission rates of the ventilation shafts of the coal mine used in this study are based on this inventory". "This" probably refers to the CoMet campaign. A campaign is not an inventory. Please rephrase.

The second sentence has been changed as the referee recommended:

"Locations and emission rates of the ventilation shafts of the coal mine used in this study are based on this campaign."

76. Line 340: "... and reasonably compared to the CoMet inventory (6.6E26 molec./s)" Please change "reasonably" to "reasonable"

Changed accordingly.

77. Line 343: "... up to 5.68E26 molec./s derived from one flight (Kostinek et al.(2021)). Similar 2D anomalies and plumes are also observed ..." Similar to Kostinek et al.? Otherwise, please separate into two paragraphs to make it clear that you are now writing about plumes/anomalies and no longer about total emission estimates.

Changed accordingly.

78. Table A-2: Instead of "CAMs emission" & "shafts emission" I think it would be better to use "CAMs-GLOB-ANT" and "CoMet inventory" according to the caption. In the left column you could label the line as "prior emission sources" or similar.

Thanks, changed accordingly.

79. WMO Reference from Line 40 is missing.

The reference has been added.