## **General comments:**

The manuscript "Quantifying hard coal mines CH4 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 CH4 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 XCH4 data retrieved by satellite observations form TROPOMI and IASI over a period of three years. Simple, straight forward to apply approaches, as presented by the authors, to estimate CH4 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.

## **Specific comments:**

- 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 TgCH4yr-1.
- 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.
- 3. 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.
- 4. 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.
- 5. 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.
- 6. In Eq. 2 you introduced ΔXCH<sub>4</sub> as the enhanced CH<sub>4</sub> column. Later, especially in figures, you use ΔXCH<sub>4</sub> for the wind-assigned anomalies. Please introduce a distinctive notation for the wind-assigned anomalies to avoid confusion with the enhanced CH<sub>4</sub>. See also the next comment for that.
- Line 163: Where does the empirical value of 60° come from? From your earlier publication I know that it comes from TROPOMI NO<sub>2</sub> measurements, but this should be explained and cited here.

- 8. 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:  $\overline{XCH_4}_{SW/NE}(i,j) = \frac{1}{N_d} \sum_{d}^{N_d} XCH_{4,d}(i,j)$  with N<sub>d</sub> = number of days and i,j  $\in$  SW/NE and wind-assigned anomaly =  $\overline{XCH_4}_{SW}(i,j) \overline{XCH_4}_{NE}(i,j)$ . Please consider showing table A-1 here instead of the appendix.
- 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 XCH4 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.
- 10. Line 195: "The XCH4 anomalies (raw-background) and the wind-assigned anomalies are presented in Figure 5a and b, respectively". Please change to XCH4 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 XCH4 in the context of Eq.2.
- 11. Line 196-197: I don't understand what you mean by: "Note, that the CAMS XCH4 is <u>coincided</u> with TROPOMI XCH4 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?
- 12. Line 213 ff: "To remove this influence, we calculate the tropospheric CAMS forecasts CH4 (TXCH4) 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.
- 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 XCH4 enhancements which are smaller by a factor of two or even more, suggesting that the CAMS-GLOB-ANT emissions are too low.
- 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.
- 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.
- 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 XCH4 enhancements or wind-assigned anomalies.
- 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.
- 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.

- 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.
- 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?
- 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.
- 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.
- 23. Line 263: "... the emission rate uncertainties of using XCH4 or TXCH4 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.
- 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<sub>4</sub> that is being used to estimate the emission rates is composed of CH<sub>4</sub> 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.

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 coneplumes 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.

- 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."
- 27. 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  $\varepsilon$ , isn't it? Accordingly, the emission rates should also be -19% & +32%. Please comment.

Furthermore, "higher emission rates" is not correct for describing a decrease and an increase. Please rephrase.

## **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 XCH4 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
- 29. For many citations there is a dot missing after "et al".
- 30. For almost all figures the labeling is way too small. Please increase the font size corresponding to the text.
- 31. Line 12: "Intensive coal mining activities are in the Upper Silesian Coal Basin (USCB) in southern Poland, resulting in large amounts of methane (CH4) emissions." Maybe shift the "are" in front of "resulting".
- 32. Line 13: "Annual CH4 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 ..."
- 33. Line 14-15: "As a CH₄ 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"
- 34. Line 16: "column-averaged dry-air molar fraction observations of CH4". Please change to "mole fraction observations".
- 35. Line 16-20: It is a rather long sentence. Maybe split up.
- 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.
- 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.
- 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.
- 39. Line 40-42: This sentence is hard to read. In my opinion the word "and" is used to often. I think in "... and waste disposal ..." you can remove it.
- 40. Line 43: "... to the atmosphere CH4 level are still ...". This seems off. Maybe change to "atmospheric" or "atmosphere's"
- 41. Line 75: "... data sets provide a large coverage and long-term XCH4/TXCH4 observations, which helps to better estimate CH4 emission ..." I guess it should be "help", not "helps".
- 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.
- 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?
- 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.

- 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 ± 4.0E25 molec./s) amount only 4% of the total emissions in this region. The dominant CH4 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 CH4 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)"
- 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".
- 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.

- 48. Line 146: Comma before "which"
- 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 ...".
- 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.
- 51. Line 159 and Eq. 2: The indices i of (xi, yi) should be subscripted.
- 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.
- 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.
- 54. Figure 4: In the caption it says "during November 2017-December 2020". Does this mean the displayed XCH4/TXCH4 data are an average of this period? If so, please indicate this in the caption. Otherwise, please specify the displayed day.
- 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.

- 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.
- 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.
- 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
- 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 [XCH4, TXCH4].T. Name the columns with [CAMS, 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.
- 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 XCH4 enhancements (i.e. XCH4 background), the middle plots are displaying wind-assigned anomalies. Please correct the colorbar labels.
- 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.
- 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  $\Delta$ XCH4 is not consistent.
- 63. Line 216: "9.1E24 ± 1.2E24 molec./s" I guess there is a typo in the exponent.
- 64. Line 227: "Figure 6 illustrates the enhance XCH4 (raw XCH4-background in the upwind) distribution ..." Please correct "enhance" to either "enhanced XCH4" or "XCH4 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.
- 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.
- 66. Figure 8: Please be precise in the labeling of the horizontal lines, i.e. "Total Emission (CAMS-GLOB-ANT", "Total Emission (CoMet inventory)".
- 67. Figure 8: Please remove the shaded background and instead add a legend: "a priori: squares CAMS-GLOB-ANT, triangles CoMet inventory", or something similar. If plotted among each other triangles and squares are easier to compare.
- 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.
- 69. Line 280: "Here we investigate the wind uncertainties ...". Please insert a comma after "here"
- 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.
- 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 \_narrow is always subscripted consequently, as is being done in the caption of Fig. 9, or, alternatively, the subscripts  $SW_{1/2}$  or  $SW_{1/4}$  could be used to specify whether the wind field is divided into halves or quarters.
- 72. Figure 9 and text in section 3.3.2: Isn't it "SW-NE" instead of "NE-SW"?

- 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.
- 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.
- 75. Line 335: "To investigate the CH4 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.
- 76. Line 340: "... and reasonablely compared to the CoMet inventory (6.6E26 molec./s)" Please change "reasonablely" to "reasonable"
- 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.
- 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.
- 79. WMO Reference from Line 40 is missing.