Response to Referee #1

We would like to thank reviewer #1 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.

This paper estimates the methane (CH_4) emissions from one of the most outstanding CH_4 sources in Europe using a multi-platform of reference data sets (space-based observations, atmospheric simulations, emission inventory) and a novel, robust, simple approach. The paper provides new and interesting findings, and is written and structured well; therefore, I suggest it to be suitable for publication in ACP after specific and technical comments (listed below) are addressed.

1. Specific comments:

1.1 Title: The title suggests that the CH_4 emission quantification is jointly done using TROPOMI, IASI, and CAMS products. However, the CAMS data was mainly used as a validation tool of the wind-assigned anomaly method. Section 3.2 indeed shows and discusses briefly an example day using the CAMS and space-based observations, and Figure 8 summarizes the CH_4 emission rates using all different data sets, but this figure is not discussed in the text. I would recommend to change it to "Quantifying hard coal mines CH_4 emissions from TROPOMI and IASI observations, high-resolution CAMS forecast data and the wind-assigned anomaly method".

(1) We would like to thank the referee for pointing this out. Yes, the CAMS data were used to evaluate our method and helped to choose the most suitable wind information. However, the CAMS data (forecast and inventory) are not used in estimating the emission strengths from the TROPOMI and TROPOMI+IASI products. The high-resolution CAMS forecast data are considered as supporting information and thus, we would like to keep "using" in the title.

(2) We have added some discussion related to Figure 8 (see 3.4 below).

2. Section Data sets and method:

2.1. A subsection describing the USCB region would help the reader, for example, including the orography, the predominant wind regimes, etc. In addition, given that the COMet inventory is used in this paper, I would also recommend including a subsection providing some details about it.

Thanks for this comment. We have added related information to the introduction (in the 2nd and 3rd paragraph, respectively) as the referee suggested:

"The USCB is in the Silesian Upland, which is a plateau between 200 and 300 m above sea level with a predominant south-west wind. The USCB within Poland covers an area of over 5800 km^2, and to its south is the Tatra Mountain ridge with elevations larger than 2000 m a.s.l."

"A variety of state-of-art instruments, including in situ and remote sensing instruments on the ground and aboard five research aircraft, were deployed in order to provide independent observations of GHG emissions on local to regional scale and provide data for satellite validation."

2.2 Line 100: Include some reference and explanation about the expected uncertainties of the CAMS-GLOB-ANT inventories.

The reference has been added.

The CAMS inventories (anthropogenic and natural emissions) do not provide estimates of the uncertainties and a potential work on the uncertainty estimates might be available in the future (we acknowledge Dr. Claire Granier from Laboratoire d'Aerologie, Toulouse, France for providing this information).

2.3 Line 135-136: Include information about the TROPOMI overpass (time, frequency,...) similar to IASI.

The information about the TROPOMI overpass has been added:

"The instrument crosses the equator at about 13:30 local solar time at each orbit with a repeat cycle of 17 days. It observes a full swath (2600 km) per second with an orbit duration of 100 min."

2.4 Line 138: Include information about the number of quality-filtered TROPOMI dataset (and also for the combined TROPOMI+IASI product in the next paragraph). Is the space-based data set robust enough for CH_4 emission estimates?

(1) The number of data points in the quality-filter TROPOMI dataset is about 16,000 over three years. About 12000 data points are collected from the TROPOMI+IASI product. We have added this information to the text.

(2) TROPOMI XCH_4 data have been characterized by high spatial- and temporal-resolution with being in good agreement with TCCON (-3.4 ± 5.6 ppb) and GOSAT (-10.3 ± 16.8ppb) (Lorente et al., 2021). TROPOMI XCH_4 has been used in different studies to detect and estimate the CH_4 emissions from different sources, e.g., from coal mining (Varon et al., 2020), and from the oil and gas sector (Pandey et al., 2019; De Gouw et al., 2020). Moreover, in a previous study (Tu et al., 2022) the emission strength derived from TROPOMI was compared to one-day observations of ground-based FTIR instruments and both have the same order of magnitudes. Our result derived from TROPOMI products in this study is close to the CoMet inventory and results of other studies by using different methods. Thus, the space-based data set is robust enough for CH_4 emission estimates.

2.5 Line 144: Include reference for the improvement of IASI on the NWP systems.

The references below have been added to the text:

Collard, A. D.: Selection of IASI Channels for Use in Numerical Weather Prediction, ECMWF, https://www.ecmwf.int/node/8760, 2007.

Coopmann, O., Guidard, V., Fourrié, N., Josse, B., and Marécal, V.: Update of Infrared Atmospheric Sounding Interferometer (IASI) channel selection with correlated observation errors for numerical weather prediction (NWP), Atmos. Meas. Tech., 13, 2659–2680, https://doi.org/10.5194/amt-13-2659-2020, 2020.

2.6 Line 145: Is the statement about "different atmospheric trace gas profiles" referring to only CH_4 or to all the MUSICA products? If the latter, please consider including other references for completeness such as Schneider et al. (2022), Dieckmann et al. (2021) or García et al., (2018).

Thanks. The statement is a general introduction about IASI and adding other references as recommended by the referee is better.

Diekmann, C. J., Schneider, M., Ertl, B., Hase, F., García, O., Khosrawi, F., Sepúlveda, E., Knippertz, P., and Braesicke, P.: The global and multi-annual MUSICA IASI {H2O, δD} pair dataset, Earth Syst. Sci. Data, 13, 5273–5292, https://doi.org/10.5194/essd-13-5273-2021, 2021.

García, O. E., Schneider, M., Ertl, B., Sepúlveda, E., Borger, C., Diekmann, C., Wiegele, A., Hase, F., Barthlott, S., Blumenstock, T., Raffalski, U., Gómez-Peláez, A., Steinbacher, M., Ries, L., and de Frutos, A. M.: The MUSICA IASI CH_4 and N_2O products and their comparison to HIPPO, GAW and NDACC FTIR references, Atmos. Meas. Tech., 11, 4171–4215, https://doi.org/10.5194/amt-11-4171-2018, 2018.

Schneider, M., Ertl, B., Diekmann, C. J., Khosrawi, F., Weber, A., Hase, F., Höpfner, M., García, O. E., Sepúlveda, E., and Kinnison, D.: Design and description of the MUSICA IASI full retrieval product, Earth Syst. Sci. Data, 14, 709–742, https://doi.org/10.5194/essd-14-709-2022, 2022.

2.7 Line 151: Some information about the improvements/differences of the wind-assigned anomaly method with respect to other top-down approaches would help the reader to have a better idea of novelty and benefit of this method.

There are generally two kinds of methods to estimate the CH_4 emission strengths. The first method is based on the atmospheric transport model (e.g., GEOS-Chem), which is considered as a forward model to create the relationship between CH_4 and surface emissions (Zhang et al., 2020). The optimization is the inversion step to obtain the best fit between the observations and the model. This method is mostly used on regional to large scales. Another method is based on the conservation of mass (e.g., divergence), i.e., the sum of the emission and background equal to the observations. This divergence method was first used to estimate NO_2 emissions (Beirle et al., 2019) and later extended to estimate CH_4 emissions (Liu et al., 2021).

Our wind-assigned method is based on the theory of conservation of mass and uses a simple cone plume model, which is easy to apply than the other methods and the estimated emission strengths are reasonable compared with the ones from other studies. This information has been added to the text.

Beirle, S., Borger, C., Dörner, S., Li, A., Hu, Z., Liu, F., Wang, Y., & Wagner, T. (2019). Pinpointing nitrogen oxide emissions from space. Science Advances, 5(11). https://doi.org/10.1126/sciadv.aax9800.

Liu, M., van der A, R., van Weele, M., Eskes, H., Lu, X., Veefkind, P., et al. (2021). A new divergence method to quantify methane emissions using observations of Sentinel-5P TROPOMI. Geophysical Research Letters, 48, e2021GL094151. https://doi. org/10.1029/2021GL094151.

Zhang, Y., Gautam, R., Pandey, S., Omara, M., Maasakkers, J. D., Sadavarte, P., Lyon, D., Nesser, H., Sulprizio, M., P., Varon, D., Zhang, R., Houweling, S., Zavala-Araiza, D., Alvarez, R. A., Lorente, A., Hamburg, S. P., Aben, I., Jacob, D.: Quantifying methane emissions from the largest oil-producing basin in the United States from space. Science Advances, 6(17), eaaz5120. https://doi.org/10.1126/sciadv.aaz5120, 2020.

2.8 Line 176: Describe slightly the results obtained (first validation of the wind-anomaly method) in Madrid experiment (Tu et al., 2021) to highlight the robustness and reliability of the method.

Thank you for this important point. We have added more information in section 2.3:

"This method was firstly used to estimate the CH_4 emission from landfills in Madrid, Spain based on nearly three-year space-borne XCH_4 data, and different opening angles were investigated to obtain an empirical value (60º) (Tu et al., 2022). The CH_4 emission strengths derived from satellite products have the same orders of magnitude as the ones from single-day observations by ground-based instruments, showing that this method works properly."

3. Results and Discussion:

3.1 Line 205: During the COMet campaign, high-resolved aircraft profiles were performed allowing CH_4 emission rates to be estimated (e.g. Fiehn et al., (2020), Kostinek et al. (2021)). Have the authors analyzed the aircraft dataset to corroborate that the wind fields at 300 m are the optimal option? As discussed in the "Uncertainty analysis", the vertical wind shear is the most critical factor to estimate the CH_4 emission rates.

We did not analyze the short-term aircraft dataset in this study. Our method is based on a long-term dataset, i.e., the CAMS XCH_4 and wind-assigned method to find out that the estimated emission strength fits best with the CAMS-GLOB-ANT inventory by using wind information at 330 m. There might have high biases for only using a short-term period of data. Moreover, the wind speed at 330 m is more or less an average of the ones at 10 m and 500 m.

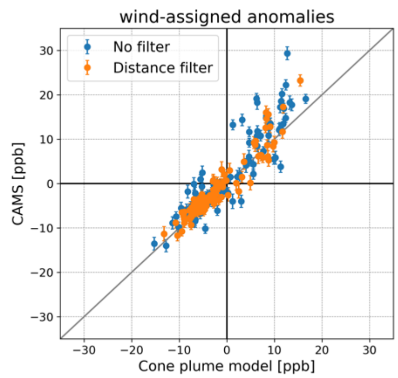
3.2 Line 206-209: Please provide more details about this statement (ie, the small changes in wind could not be properly captured by ERA wind fields). What would the net effect of ruling out these pixels be in the total estimations?

We consider the wind changes (speed and direction) over daytime and these effects are averaged based on the time scale and super-positioned for all emission sources. The enhanced column is proportional to distance, and it is set to zero only when the distance is zero, i.e., the points locate exactly in the emission sources' places. The distance-related filter ("the points whose distances to the nearest dominant sources are less than 10 km") is not applied in calculating the enhanced columns and in estimating the emission strengths. The previous correlation plots in the manuscript were distance-related filtered, which might mislead the readers. This sentence is removed, and correlation plots have been updated.

3.3 Figure 5: Why is there more scatter in the positive anomalies?

These positive anomalies represent the values in the SW area (i.e., the downwind region of the NE wind), where more emission sources are located than in the NE area. The enhanced CH_4 columns (Eq.

2) are proportional to the distance, and thus, the positions that are near the emission sources can be easily affected by the sources. Although we removed the points whose distances to the nearest dominant sources are less than 10 km in the previous correlation plots, the points might be affected by other sources, which probably results in more scatters in the positive anomalies. The figure below shows that most scatters are related to the points that are near the emission sources.



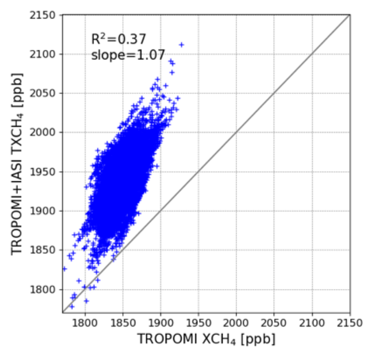
3.4 Line 224: As mentioned before, section 3.2 shows and discusses briefly an example day using the CAMS and space-based observations, but the emission rates using the whole data set is not included and discussed. If I understand well, the analysis was done because Figure 8 summarizes the CH_4 emission rates using all different data sets for the discussion of effect of wind at different levels, but this figure is not discussed in the text (neither in section 3.2 and section 3.3). Including this information in the text would help to compare the results with COMet inventories (discarding the influence of space-based observations uncertainties).

Thank you for this important comment. The following paragraph has been added to the paper:

"Figure 8 summarizes the estimated emission strengths derived from different products based on different a priori knowledge of inventories and wind information at different altitudes (for specific values see Table A- 1). Different a priori inventories result in 16%-32% changes in strength at different altitudes, which is generally smaller than the 47% difference in the total amount of inventories (9.7E26 for CAMS-GLOB-ANT and 6.6E26 molec./s for CoMet inventory). This is probably due to the different locations of sources and different proportions of each emission source in the total strengths in the two inventories. When using the CAMS-GLOB-ANT inventory, CH_4 emission rates derived from CAMS XCH_4 and $TXCH_4$ are ~37% and ~56% higher than those derived from TROPOMI XCH_4 and IASI+TROPOMI $TXCH_4$, respectively. This difference is mainly due to the difference between the CAMS forecast and satellite products. The strength increases with respect to the increasing wind speed at higher altitude. Whereas the increment is not always proportional to the wind speed, i.e., less increase in the strength with respect to the wind speed at higher altitude (see Sect. 3.3.1)."

3.5 Line 247: There is a significant change of slope for the combined $TXCH_4$ product (Figure 7 f). Do the authors have some explanation for this?

The different slope for modeling emission strength derived from $TXCH_4$ products is mainly due to the difference between XCH_4 and $TXCH_4$. XCH_4 is the ratio of the total column of CH_4 and the total column of dry air, whereas the $TXCH_4$ is the ratio of the total column of CH_4 in the troposphere and the column of the tropospheric dry air. Mixing ratios of CH_4 decrease in the stratosphere, resulting in higher absolute values of $TXCH_4$ than XCH_4, with a slope of 1.07 (see figure below). The modeled ΔXCH_4 and $\Delta TXCH_4$ in Figure 7(c) and (f) are the same product and thus, a lower slope is expected in fitting the TROPOMI+IASI $\Delta TXCH_4$ to the model ones. The ratio (1.07) of $TXCH_4$ and XCH_4 is close to the ratio (1.05/0.89=1.18) of the slopes in Figure 7 (c) and (f), which further supports the explanation above.



4. Technical comment:

4.1 Line 19 and line 76: Include the period covered by this study in the abstract and introduction.

Corrected, thanks.

4.2 Line 70: Include acronym for tropospheric XCH_4 ($TXCH_4$).

Thanks, this has been added.

4.3 Line 89: Consider plural for “aerosol”.

corrected, thanks.

4.4 Figure 1: The colours used for “Off Road transportation” and “Fugitives” are quite similar and make it hard to distinguish them only by looking at the plot. The final full stop is missing.

The figure has been updated.

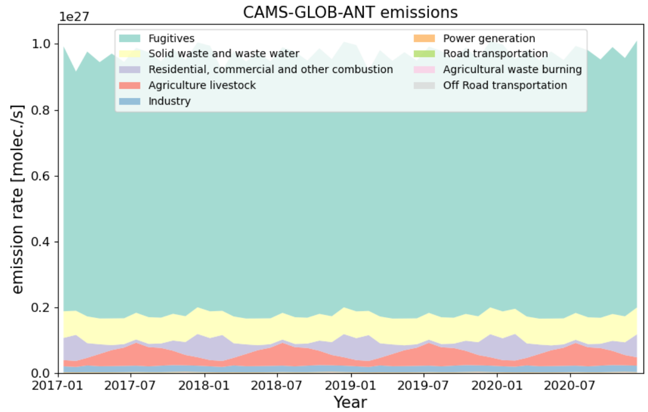


Figure 1: Stacked area plot for different sectors of the monthly averaged CAMS global anthropogenic emissions (>1E20 molec./s) in the USCB region for 2017-2020 (https://permalink.aeris-data.fr/CAMS-GLOB-ANT, last access: 22 December 2021. Granier et al., 2019).

4.5 Line 154: Please consider moving the description of the ERA wind model to line 164.

This has been done as the referee recommended.

4.6 Line 166: 08:00 UTC or 09:00 UTC as in the CAMS products description. Why do not use the CAMS products starting at 08:00 UTC?

The daily wind-assigned plume from each emission source is averaged over daytime (8:00 UTC – 18:00 UTC), i.e., we considered the wind changes over the day. The different single-source-resolved plumes from all emission sources are super-positioned to a total daily plume. We then fit the different daily plumes to the CAMS XCH_4. Because the daytime average emissions are calculated, we then use the daily averaged CAMS XCH_4 as well. However, the CAMS XCH_4 has a temporal resolution of 3h, starting from 00:00 UTC. Therefore, the CAMS XCH_4 at 9:00, 12:00, 15:00, and 18:00 UTC are used to calculate the daily average, and their standard deviations are considered as uncertainties.

4.7 Line 180: Correct “500 m” and “three-year average” in the figure caption.

Thanks, corrected.

4.8 Figure 3: Correct “TROPOMI” in the figure caption.

The typo in Figure 4 has been changed accordingly.

4.9 Figure 5: Include the meaning of the error bars in the figure caption (is the STD given by Eq 1?).

The information has been added.

4.10 Figure 6: To be consistent with the other figures, please consider modifying this figure accordingly (coloured bars, labels (a, b, c), “modelled” in the title of third subplot, definition of first subplot,...)

Thanks, the figures have been modified.

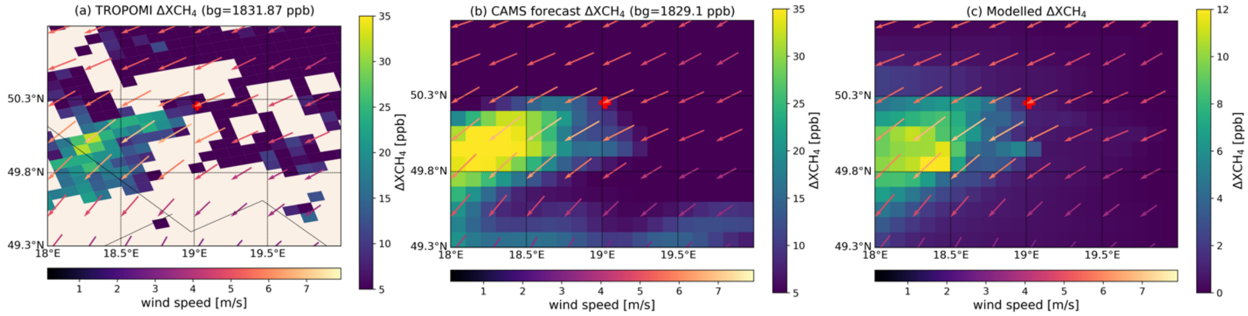


Figure 6: ΔXCH_4 together with the ERA5 wind at 12:00 UTC from (a): TROPOMI observations at 11:34 UTC, (b): CAMS forecast at 12:00 UTC, and (c): from the simple plume model (averaged over the daytime) based on the CAMS-GLOB-ANT inventory over the USCB region on an example day (6 June 2018). The "bg" in the title of (a) and (b) represents the average background, derived from the mean XCH_4 in the upwind region (50.3º-50.8º N, 19.5º -20.0º E).

4.11 Figure 7: Correct "TXCH4" in subplot (e).

Thanks, corrected.

4.12 Figure 8: Correct "wind" in the x-label for 300 m. Correct "300 m, 500 m".

Changed accordingly.

4.13 Line 284: Correct Figure A-1 to plain text.

Changed accordingly.