Response to Referee #2

We would like to thank reviewer #2 again for taking the time to review the revised version of the manuscript. We appreciate the level of detail and the valuable suggestions which help us to further improve the manuscript.

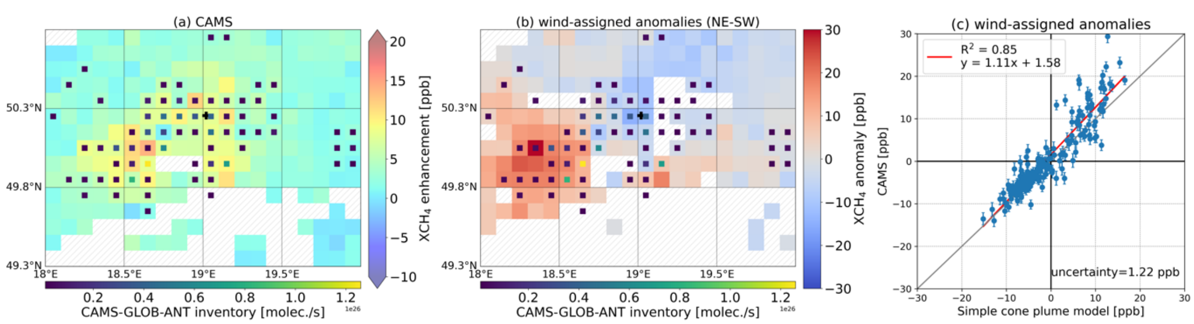
In this author's comment, all the points raised by the reviewer are copied here one by one and shown in blue color, along with the corresponding reply from the authors in black.

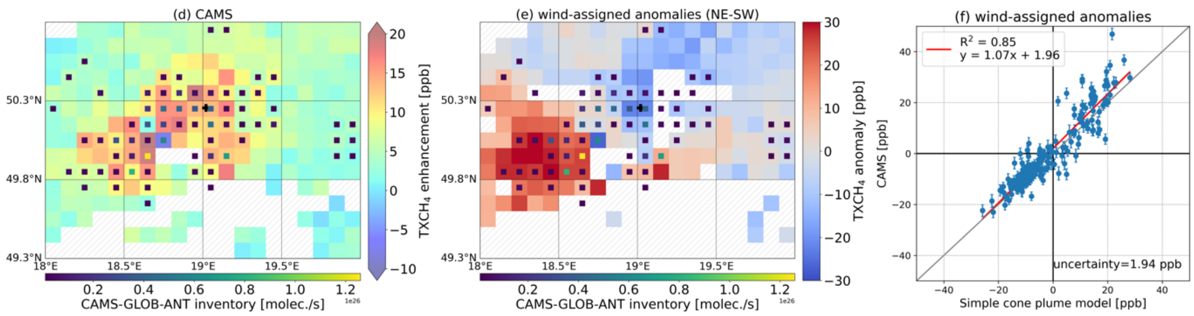
General comments:

The authors have made an impressive effort to improve the manuscript. The visual appearance of all figures has improved significantly. The text also shows significant improvements. Many stumbling blocks for the reader have been removed.
Nevertheless, some comments remain incompletely addressed. In particular, there are individual comments which the authors claim to have incorporated, but did not do so completely in the manuscript or did so incorrectly.

Concerning the improved Figures, I have to apologize. I've noticed that in my previous comments I've sometimes mixed up the terms "colormap" and "colorbar". At the current state I wonder why for some figures the colorbars cover different values. In general, it is obvious that $TXCH_4$ shows stronger enhancements than XCH_4. But for example, Figures 5 (a) and (b) should have the same range of values. (d) and (e) should also cover the same values. Otherwise, the visual comparison of the plots side by side creates a false impression. The same holds true for Figure 7, Figure A-2 and Figure A-3. If there is a reason to not use that same range of values in the colorbars, please indicate so in the respective captions. Like you did in Figure 6, where you're only want to compare spatial pattern.

Thanks for the comment to further improve the figures. Fig. 5 (a) and (d) (and other related figures) represent the overall enhancements (raw XCH_4/$TXCH_4$ - background), whereas Fig. 5 (b) and (e) represent the difference between the enhancements for NE wind and for SW wind fields. We think that there is no need to use the diverging colormap in (a) and (d), otherwise it might bring some misleading information here. Thus, a different colormap is used for (a) and (d) (comment from SC 16 is also considered here). For either enhancement figures or wind-assigned anomalies figures, the colorbar covers the same range of values for XCH_4 and $TXCH_4$, i.e., (a) and (d) use the same colormap and colorbar, so do (b) and (e).





Comments of Referee #1:

- 1.1: I strongly agree with Referee #1. The CAMS data was used solely for the validation of the wind-assigned anomaly method. The term "using" suggests that the CAMS data was actually used for your emission estimates, which they where not. I would even go one step further than Referee #1 and not include the CAMS data in the title at all: "Quantifying hard coal mines CH_4 emissions from TROPOMI and IASI observations using the wind-assigned anomaly method". However, this is only a recommendation. If the authors feel differently I suggest discussing it with the editor.

Thanks for this suggestion. We agree and have changed the title accordingly.

- 2.1: for the sake of completeness, a study on measurements on HALO should also be cited, as HALO was the flagship of the campaign. I recommend Galkowski et al. 2021 (https://doi.org/10.5194/amt-14-1525-2021) on in situ observations on HALO and Wolff et al. 2021 (https://doi.org/10.5194/amt-14-2717-2021) on airborne lidar observations. Also, I want to raise your attention to Andersen et al. 2021 (https://doi.org/10.1016/j.aeaoa.2021.100135) on UAV based emission estimates in the USCB and Luther et al. 2022 https://doi.org/10.5194/acp-22-5859-2022. If it seems fitting to you, you could include these two publications at an appropriate location of your manuscript. But of course, only as an option for you.

Thanks for recommending these related references.

Andersen et al., 2021 has been already cited in the beginning of the introduction, but its method and result were not introduced. A sentence has been added to the text:

> "Active AirCore system aboard an unmanned aerial vehicle (UAV) was used to measure CH_4 downwind of a single ventilation shaft and emission rates ranging from 0.5 to 14.5 kt/year based on a mass balance approach and ranging from 1.1 to 9.0 kt/year based on an inverse Gaussian method were estimated (Andersen et al., 2021)."

Luther et al., 2022 was also already cited as an ACPD version in the manuscript ("A recent study (Luther et al., 2021) displays a larger emission rate of 414 – 790 kt/year based on a network of four portable FTS instruments (EM27/SUN) during the CoMet campaign."). We have noticed that the paper has been accepted and thus, we have updated the citation.

The other two studies have been also cited:

"For example, Gałkowski et al. (2021) present results of in situ GHG measurements obtained over nine research flights of the German research Aircraft HALO (High Altitude and LOng Range Research Aircraft) acting as the airborne flagship of the CoMet campaign, together with simultaneous flaks measurements for isotopic composition of CH_4. A new lidar CHARM-F (CO_2 and CH_4 Atmospheric Remote Monitoring Flugzeug) was also onboard HALO and its measurements were investigated to determine CO_2 emission rates from the power plant (Wolff et al., 2021)."

Specific comments:

- SC 0 (new comment):
  - In the abstract in lines 21-27 validation results are given. I'm in big favor of giving results in the abstract, but only the main results, i.e. the emission estimates based on the satellite observations. Here, it is sufficient to simply state, that the wind-assigned anomaly method is validated using CAMS forecast data, showing good agreement to the CAMS-GLOB-ANT inventory. You don't have to give numbers. For the reader the results of the validation distract from the main results, which are supposed to be the highlight in the abstract.

Thanks for this comment. The abstract related with the CAMS data has been shortened according to referee's comment:

> "The wind-assigned anomaly method is first validated using CAMS forecast data (XCH_4 and $TXCH_4$), showing a good agreement to the CAMS-GLOB-ANT inventory. It indicates that the wind-assigned method works well."

  - The same applies for the last paragraph of the abstract (i.e. lines 37-43). The sensitivity analysis of wind speed is a method for determining a contribution to the uncertainty in the emission estimates. The results of this analysis should be reflected in the given uncertainty of the emission estimates. For the abstract it is sufficient to state that a sensitivity analysis of wind speed for different altitudes has been made.
    This is part of your chosen approach and should be stated before giving the main results.

The last paragraph is changed to (comments from SC 24/27 are considered as well):

> "Uncertainties from different error sources (background removal and noise in the data, vertical wind shear, wind field segmentation, and angle of the emission cone) are approximate14.8% for TROPOMI XCH_4 and 11.4% for TROPOMI+IASI $TXCH_4$. These results suggest that our wind-assigned method is quite robust and might also serve as a simple method to estimate CH_4 or CO_2 emissions for other regions."

- SC 3: In line 201 it now says "wind regime sector". In Table 1 it says "wind area". Please recheck the manuscript, if all your changes are applied to your will.

Changed accordingly.

- SC 4: It still says "simple plume model" in the title of subsection 2.3, in lines 131, 185, 277, 400, captions of Fig. 6 and A-1. Please review your entire manuscript for consistency and use only "cone plume model" or "simple cone plume model".

Thanks. The "simple cone plume model" is used and all the related texts have been changed accordingly. The xlabel of the correlation plots in Fig. 5, 7, 9-11 and Fig. A-4 have been updated as well.

- SC 5: If there are such high spreads and uncertainties in your estimates for the individual years, I don't understand how you come up with such low uncertainties in your estimate of the three-year period. Please comment this in the scope of SC 24/27 below.

The high uncertainties in the first two years mainly come from high uncertainties in the elimination of the background due to the small amount of data. The same situation is met in Sect. 3.3.2 and Fig. 9 (d), when small amounts of data had to be used to derive the estimates for $NW_{1/4}$-$SE_{1/4}$ wind. This high level of uncertainty is significantly reduced if larger data sets are available.

- SC 6: Eq. 6. I'm a bit baffled by the mix of equation and free text. I have to admit that I'm not sure if that's formally allowed or not. You might think of a variable for "wind-assigned anomaly". Something like "δXCH_4" or similar. I realize that you would only need this variable at this point so this is only a recommendation. You could also wait for a comment from the type setting of the journal.

Thanks for the suggestion. A variable for the wind-assigned anomaly would be indeed nice, but finding the right name is not so easy, thus we would like to leave it as it is for the time being and wait for comments from the typesetting.

- SC 12: I'm afraid you didn't add your statement to the manuscript. At least in the "track-changed" it is unchanged (see 256 ff). Moreover, you still haven't answered the question. How did you come up with exactly 7 km? Why not 6 or 8 km?

Sorry for this mistake. The statement has been added to the manuscript (Sect. 3.1).

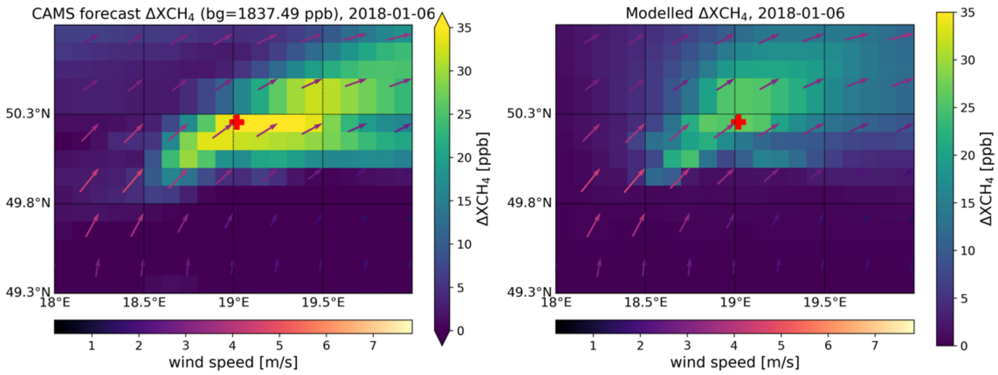
We use 6 km (sorry, 7 km was a typo here) which is adopted from the study by Schneider et al. (2021), where the TROPOMI+IASI $TXCH_4$ products are the partial columns up to 6 km a.s.l. The results derived from the two data sets are then comparable. For TROPOMI+IASI $TXCH_4$ products, ground surface to 6 km a.s.l. has been chosen, because it represents the layer for which the DOFS (degree of freedom of signal, i.e., the sum of the diagonal elements of the averaging kernel) of the combined profile product is generally very close to 1.0, meaning that the combined satellite product is well sensitive to variation of these partial column amounts

- SC 13/14: "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." If you do not explain how these biases come about, it is hard to understand why they are compensated over a long period.

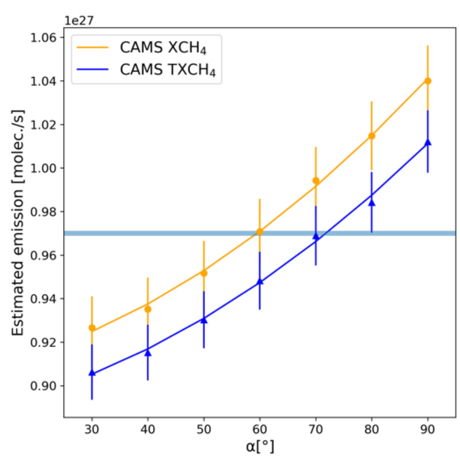
  I do not understand why you expect such huge biases. Most plume models (e.g. Gaussian plume model) are recursively linear with wind speed and linear with emissions rate. Usually, in plume models some parameter accounts for turbulence. The only possible representation of turbulence in your model is in the angle α=60°. In some cases, this angle will be too small, in some cases it will be too large. This effect might be canceled out, as you suggested, over the long observation period. But it seems as your cone-model is either showing lower XCH_4 enhancements, or supposedly too narrow plumes. As your overall goal is to be representing for the overall observation period this is no show-stopper, but please openly discuss/explain the limits of the cone plume model to the reader.
  In your former publication you showed plots of the cone plumes for different values of α. While you derived 60° as the best fit to the N_2O plumes for Madrid, I'm not convinced that 60° is necessarily the optimal fit for the USCB, too. At the very least should investigate to what

quantitative extent variations in α impose uncertainties on your emission estimate. As far as I can see this has not yet been considered in your uncertainty analysis Sect. 3.3.

Thank the referee for pointing it out. The assuming opening cone α can be either too small or too large in different single days. For example, the figures below show enhancements from CAMS XCH_4 and the simple cone plume model on January 6, 2018. The angle α is overestimated, which results in lower values compared to the CAMS XCH_4 enhancements. This effect can be canceled out over the long-term observations.



The estimated emissions over the study period show a positive correlation to the α values (see figure below, the blue horizontal line represent the total value of the CAMS-GLOG-ANT inventory). The emission rate derived from CAMS XCH_4 fits the best to the CAMS-GLOB-ANT inventory for α=60°. This finding supports our empirical choice for α (over a long-term period).



We have added a subsection to the Sect.3.3 and discussed the impact of a suboptimal choice for α. The choice of the cone angle introduces small uncertainties to the averaged results. The subsection below has been added in the manuscript:

3.3.4 Investigation of different choices for angle of the emission cone

The angle ($\alpha = 60°$) used in the simple cone plume model is an empirical value which affects the deduced emission strengths. Figure 11 shows the results when α is decreased or increased by 10°. Changes in the spatial distributions of wind-assigned anomalies and in the correlations derived from

CAMS and the simple cone plume model are nearly negligible when using different angles in the model. The estimated emissions are 789 ± 16 kt/a (9.5E26 ± 1.9E25 molec./s) for $\alpha = 50°$ and 832 ± 17 kt/a (9.9E26 ± 2.0E25 molec./s) for $\alpha = 70°$, which are 3% lower and 2% higher than that with using the empirical angle ($\alpha = 60°$).

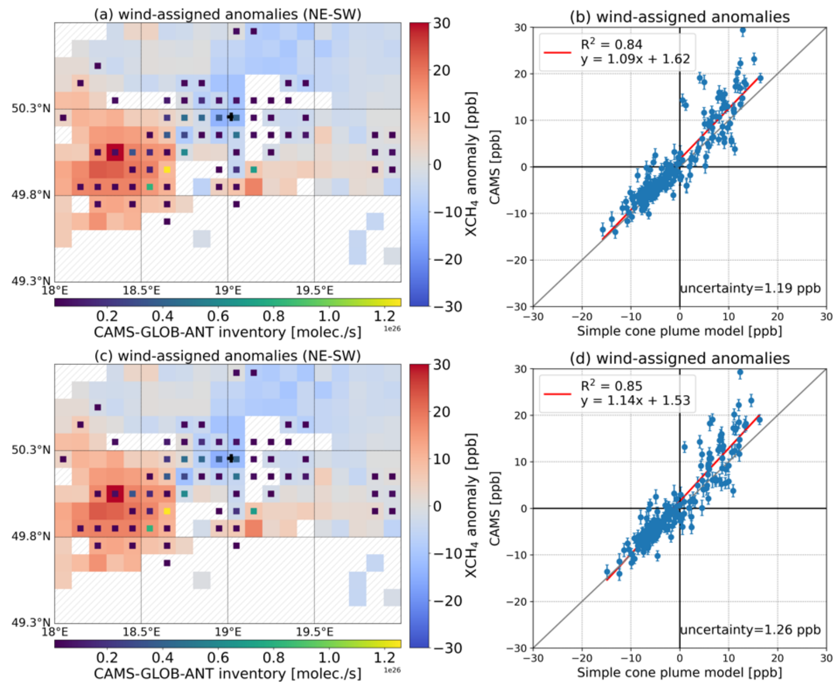


Figure 11: Similar figures to Figure 5b-c. Results are derived from CAMS XCH_4, CAMS emission inventory and ERA5 wind at 330 m for (a)-(b) $\alpha = 50°$, and (c)-(d) $\alpha = 70°$.

- SC 16 Figure 6: Why do the colorbars start with the value 5 and not 0? If the colorbars are extended (i.e. $\Delta XCH_4<5$ is the same color as $\Delta XCH_4=5$), an extension arrow at the colorbar should indicate so. The same applies for the upper end of the colorbar. But, as mentioned in the general comment above, I strongly recommend using the same range of values for all colorbars. If the spatial pattern in (c) would not be recognizable anymore you can leave the colorbar as it is now., but at least start your colorbars in (a) and (b) with 0.

  Also indicate the different colorbars in the caption. Currently it seems like the modelled plumes fit perfectly to the CAMS forecast and the TROPOMI data. Which they do only in spatial appearance, not concerning the magnitude of XCH_4 enhancement.

Thanks for this noting. Fig. 6 (a) and (b) have been updated. For Fig. (c), we mentioned that different colorbars are used. Note, the enhancement is always positive based on the simple cone plume model (c) and thus, no extension is applied to the colorbar.

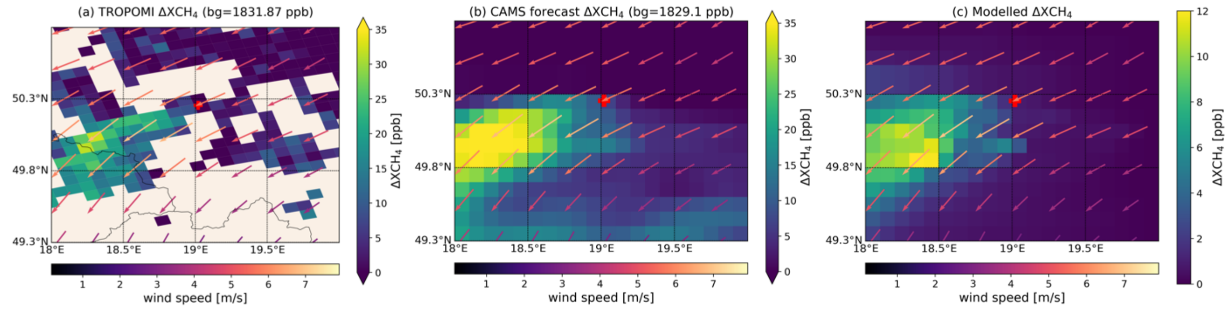


Figure 6: ΔXCH_4 together with the ERA5 wind at 12:00 UTC from (a): TROPOMI observations at 11:34 UTC, (b): CAMS forecast at 12:00 UTC, and (c): from the simple cone plume model (averaged over the daytime) based on the CAMS-GLOB-ANT inventory over the USCB region on an example day (6 June 2018). The "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). <u>Note, a different colorbar has been used in panel (c) for improved recognizability.</u>

- SC 17: Sorry, in my first review I mixed the terms „colormap" and „colorbars" by mistake. Here, I was actually referring to the colorbars and the different range of values covered by it. As mentioned in SC 16 this is ok, if you mention in the text why you did so.

Thanks, no problem – please see the reply above.

- SC 20: I actually think that these two plots are of high explanatory value. Please consider including them in the manuscript (optionally). Especially, because I now realized that I misunderstood, that by NE/SW in the title of your figures you mean wind coming from NE/SW and not plumes propagating in NE/SW-direction. My bad, but if you want to make sure that this will not be misunderstood by the reader, you could include these two plots in the manuscript.

Thanks. As the referee suggested, these two figures provide additional supporting information for readers to better understand the applied methods. They have therefore been added in the appendix.

- SC 24/27: In the abstract you give your emission estimate with 479±4 kt/yr and 437±18 kt/year. Then you share the results from your sensitivity analysis regarding wind speed, separately. The uncertainties given your sensitivity analysis should be included in the uncertainties of your emission estimates. The CH_4 exhaust from the ventilation shafts is released at a height of approx. 20 m. Propagating downwind it will be carried upwards by convective eddies and thereby distributed in the entire boundary layer. Have a look into the video supplement of https://amt.copernicus.org/articles/14/2717/2021/#section11 or the Figure A2. While the highest concentrations of CH_4 will, for sure, be advected in the middle of boundary layer, or even closer to the ground, you'll need to include the vertical variations of wind speed in your emission estimate uncertainties in some way!

The referee is correct in assuming that we did not consider the vertical variations of wind speed. Instead, we use wind speed and direction at a certain (hopefully) representative altitude to derive the emissions. For this reason, we discussed the sensitivity resulting from using the wind field at different levels (10 m and 500 m). Please note that other current methods for quantifying emissions also use the simplification of a 2-dimensional transport in the planetary boundary layer, e.g., Liu et al. (2021). We are investigating the possibility of extending our method by incorporating a 3-dimensional plume dispersion model.

(Liu, M., van der A, R., van Weele, M., Eskes, H., Lu, X., Veefkind, P., de Laat, J., Kong, H., Wang, J., Sun, J., Ding, J., Zhao, Y. and Weng, H.: A New Divergence Method to Quantify Methane Emissions Using Observations

of Sentinel-5P TROPOMI, Geophys. Res. Lett., 48(18), e2021GL094151, doi:https://doi.org/10.1029/2021GL094151, 2021.)

In your uncertainty analysis (Sect. 3.3) you analyze three sources of uncertainty. Within the analysis the emission results vary strongly from the validation emission estimates given in the abstract and conclusion. Considering this, I'm confused how the uncertainty in your overall emission estimate can be so small. Please state the relative contributions of the sources of uncertainty to the overall uncertainty, including uncertainties induced by the selection of cone opening angle α (see SC 13/14)

The previous uncertainties in the emission estimate are determined by considering the deficits of the background model due to the imperfect elimination of the background. The uncertainties related to the noise in the data set (e.g., the noise in the satellite observations) were not included. In the newly revised manuscript, we have updated the uncertainty values to include noise uncertainties based on the error propagation. The sentence concerning the uncertainties of the emission results has been added to Sec. 2.3:

> "The uncertainties (± values) in the emission estimate are determined by considering the deficits of the background model due to the imperfect elimination of the background and the noise in the data set."

Meanwhile, further error sources as discussed in Sect. 3.3 are included in the complete uncertainty budget (as collected in Table A- 2) for the specification of the total uncertainty (in percentage) on the emissions we deduced (as the reader would expect). The following text has been added to Sect. 3.3:

> "The changes in the estimated emission rates for different products due to different error sources are summarized in Table A- 2. Based on the error propagation, the total uncertainty in the estimated emission rates from the different error sources (background removal and noise in the data, vertical wind shear at 500 m, wind field segmentation, and opening angle $\alpha = 70°$) is approximately 14.7% for CAMS XCH_4, 14.8% for TROPOMI XCH_4 and 11.4% for TROPOMI+IASI $TXCH_4$. Note that, the use of narrowed angular wind regimes is not a preferable way due to few amounts of data in narrowed wind regimes and thus, is not considered an error source. In addition, the 500 m wind shear was used as a contribution to the budget, as the 10 m wind is not expected to be representative of the PBL."

Table A- 2: Changes in the estimated emission rates for different products when using different input data or under different situations compared to their results using the default setting (wind at 330 m, NE-SW wind segmentation, α =60º, CAMS-GLOB-ANT for CAMS XCH_4 and CoMet inventory for TROPOMI XCH_4 and TROPOMI+IASI $TXCH_4$).

	CAMS XCH_4	TROPOMI XCH_4	TROPOMI+IASI $TXCH_4$
Background removal & noise in the data	2.1%	3.6%	6.1%
Vertical wind shear (500 m)	13.4%	6.8%	5.8%
wind field segmentation (N-S)	-5.2%	12.7%	7.7%
angle of the emission cone (α=70º)	2.1%	0.07%	-0.02%
Total:	**14.7%**	**14.8%**	**11.4%**

Technical comments:

- TC 50: In the caption it still says "simple plume model" although the authors have confirmed to switch to the term "plume cone model". Please watch out for consistency.

Sorry for this mistake. Changed accordingly.

- TC 51: In Eq. 2 the indices "i" are still not subscripted. Actually, d and ΔCH_4 are functions of the location. So x_i, y_i should not be subscripted, only the index "i":

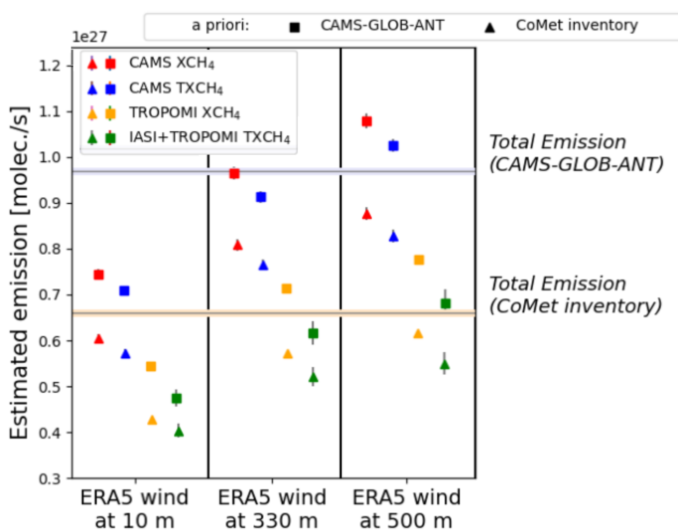
$$\Delta \mathrm{CH}_4(x_i, y_i) = \frac{\varepsilon}{v \cdot d(x_i, y_i) \cdot \alpha}$$

Thank you for clarifying it again. Changed accordingly.

- TC 57: caption Fig. 5: "colorbars in (d) and (e) are different from that for XCH_4". Actually, all four colorbars cover different values. To me it makes sense to have different colorbars for XCH_4 and $TXCH_4$, as $TXCH_4$ generally shows higher enhancements. But why different colorbars for (a) & (b)? And why different colorbars for (d) & (e)? See general comment.

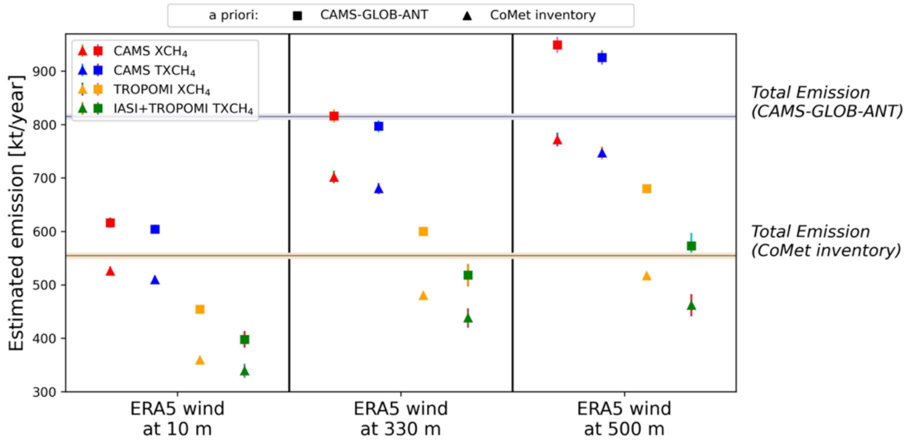
Please see our reply to the general comment above.

- TC 67: My apologies, by my phrasing "... among each other ..." it was not clear what I actually meant. I thought of something like that:



So that triangles are actually vertically aligned with the squares. In this plot the reader should become aware that CAMS-GLOB-ANT emissions are always higher than from the CoMet inventory. By plotting them vertically aligned this becomes more obvious.

Many thanks for helping with the improvement of the figure. The figure has been updated:



- TC 71: "We use $NE_{1/2}$ for 0°-90°, $SW_{1/2}$ for 180°-270°, $NW_{1/2}$ for 270°-360°, and $SE_{1/2}$ for 90°-180°". My recommendation was to use the subscript "1/2" everywhere when the wind field is divided into two halves (i.e. everywhere before Sect. 3.3.2). The subscript "1/4" was supposed to be used, when the field is divided into quarters (previously designated by "_narrow"). This indexing is of course only necessary when talking about the narrowed angular wind regimes in Sect. 3.3.2. So, if you don't want to include an index in the manuscript before this Sect. it's fine by me. But in Sect. 3.3.2 it should be "1/4".

We use NE (or SW) covering a range of 180º in our pervious analysis. The narrow fields in Sect. 3.3.2 represent half of the predefined NE (or SW) range, and thus, a subscript of "1/2" is used here. This definition seems to bring some misunderstanding. We changed the "1/2" to "1/4" as the referee recommended. The corresponding text and the caption in Figure 9 (a) and (c) have been changed.