Response to Referee #3

We would like to thank the reviewer #3 for taking the time to review this manuscript and for providing valuable and constructive feedback that helped us to further improve the manuscript.

In this author comment all the points raised by the reviewer are copied here one-by-one and shown in bold text, along with the corresponding reply from the authors in plain text.

Major comments

1. Reconsider the structure of the introduction. For the moment, I cannot capture what is the current status of the CH4 emission research from the ground-based and space-based measurements, what are the key issues, and what the authors will do in this work to solve/improve the issues. For example, I would like to suggest moving some texts in Section 2.1 and Section 2.2 to the introduction. Only keep the data description/technical part in Section 2.

Thank you for the comments. We followed your suggestions by adding more information to introduce the ground-based and space-based measurements. Therefore, we moved the respective text parts from Section 2.1 and 2.2 to the introduction and added additional references.

2. In the abstract: "As CH4 emission strength we estimate $7.4\times10^{25} \pm 6.4\times10^{24}$ molec s-1 from the TROPOMI XCH4 data and $7.1\times10^{25} \pm 1.0\times10^{25}$ molec s-1 from the TROPOMI+IASI merged TXCH4 data." Why the uncertainty derived from the TROPOMI+IASI TXCH4 data is larger than that from the TROPOMI data? I thought that the advantage of using TROPOMI+IASI is to obtain more information in the troposphere so that users can reduce the uncertainty.

It is not possible to have the $TXCH_4$ by either the TROPOMI or IASI product individually. The synergetic combination of TROPOMI and IASI enables us to detect tropospheric CH_4 independently from the upper tropospheric/stratospheric CH_4. Because the TROPOMI+IASI TXCH4 product is not influenced by the varying tropopause, it is more sensitive to the tropospheric variations than the XCH_4 data. On the other hand, the merged product has a larger noise error: (1) because the vertical distribution of CH_4 is in general much more difficult to measure than the total column of CH_4 and (2) because we derive the vertical distribution by considering two independent measurements, each with its own noise error. With the current data availability, we estimate that the emission rates obtained from the $TXCH_4$ data have a slightly higher uncertainty than the emission rates we obtain from the XCH_4 data. This might change for a larger number of data points (e.g. by using data from more years or by applying the method to IASI and TROPOMI successors on the upcoming METOP-SG satellite, which offers much more collocated observations).

However, we would like to point out that in our study using $TXCH_4$ data in addition to XCH_4 data nicely documents the robustness of the method. Important for a correct estimation of the

emission is the correct removal of the methane background signal. For XCH_4 the stratospheric and the tropospheric background have to be removed. For $TXCH_4$ only the tropospheric background has to be removed. In our study we use XCH_4 and $TXCH_4$ data sets. Figure 7 shows the rather different background signal of XCH_4 and $TXCH_4$. Despite this difference we estimate very similar emission rates from both data sets (the emission rate uncertainties using XCH_4 or $TXCH_4$ are insignificant compared to the estimated emission rates). This proves that our method gives robust results even when using data with rather different background signals.

Minor/technical comments:

P2line36. It is confused with "that this". Please reword this sentence" That this strength is lower than the one derived from the satellite observations is a plausible result."

corrected.

P2line45: "are to" -> "are"

corrected.

P2line49: 'while' -> 'in which'

corrected.

P2line49: What does '~55% uncertainty' mean? Please clarify it in the text.

clarified. It means that in the global uncertainty share the landfills own 55%.

P2line53: 'space borne' -> 'space-borne'. As you use both ground-based and space-based measurements in the study, why do you only highlight the space-based data here?

corrected. Thanks for pointing it out. The ground-based data should be also highlighted here.

P2line62: 'TCCON' for the first time, please write down the full name

corrected. The full name is "Total Carbon Column Observing Network" and it has been added to the text.

P3line70: 'column-average' -> 'column-averaged'

corrected.

P3line81: 'The Bruker EM27/SUN' -> 'A Bruker EM27/SUN'

Here we mention the specific instrument and we think "the" is a proper word.

P4line115: 'we apply a strict quality control' - > 'we apply strict quality control'

corrected.

P4line124: 'particular' ->'particularly'

corrected.

P5line128: 'such synergetic' ->'such a synergetic'

corrected.

P5line146: 'emssion' ->'emission'

corrected.

P7line171: ‘each individual landfill’ -> ‘each landfill’

Here we would like to emphasize that each landfill is considered as an independent source. Therefore, we use “each individual” here.

P7line186: ‘in Madrid area’ -> ‘in the Madrid area’

corrected.

P7line191: ‘which brings error’ -> ‘which brings an error’

corrected.

P8line202: ‘Due to its coarser spatial resolution the TROPOMI XCH4’ -> add a ‘,’ after resolution

corrected.

P8line212: ‘This value fits well to’ -> ‘This value fits well with’

corrected.

P9line228: ‘as attenuated signal’ -> ‘as an attenuated signal’

corrected.

P9line229: ‘a time period of’ -> ‘a temporal window of’

corrected.

P10line237:’during some years after sealing’ -> ‘for years after sealing’

corrected.

P10line243: ‘To better representing’ -> ‘To better represent’

corrected.

Pllline276: ‘fusiform-shape plumes’ - >’ fusiform-shaped plumes’

corrected.

P12line283:’ is due to noise of’ -> ‘is due to the noise of’

corrected.

P12line295:’ CH4 has a relatively longer lifetime than NO2’ -> ‘CH4 has a longer lifetime as compared to NO2’

corrected.

P16line333: ‘yields emission rates of close to’ -> ‘yields emission rates close to’

corrected.

P18line374: ‘derived from source location’ -> ‘derived from the source location’

corrected.

P19line396: ‘are indeed representative for’ -> ‘are indeed representative of’

corrected.

P19line397: 'CH4 has a long lifetime' -> 'CH4 is a long-lived gas'

corrected.

P20line423: 'As outlook, this methodology…' -> 'This methodology…'

corrected.

P25 A-3: "personal communication of Omaira García" Why do you write this? If I understand correctly, Omaira García is one of the co-author.

Thank you for pointing it out. We will remove this text.

P26 Eq8. I would like to see a table or an expansion for the x_BG vector

$\boldsymbol{x_{BG}}$ is a vector, consisting of the coefficients for each component of the background model. The components of the background model are described in line 513-516 and more detailed information is below.

P26 Eq 8. How do you calculate the K_BG? Perturbation? If yes, then how do you choose the perturbation size? How does the size affect the result?

$\mathbf{K_{BG}}$ is a matrix, where each row represents an individual satellite observation and each column a component of the background model. The first component of the background model is a constant, thus the entries in the first column of $\mathbf{K_{BG}}$ are 1.0 for each row (for each observation). The second component of the background model represents the linear of CH_4, thus the second column of $\mathbf{K_{BG}}$ is the time (t) when the respective observation has been made (each row might have a different observation time). The next six columns of $\mathbf{K_{BG}}$ are for the seasonal cycle and the entries are $\sin(2\pi t/365)$ and $\cos(2\pi t/365)$ for the $\frac{1}{\text{year}}$ frequency, $\sin(4\pi t/365)$ and $\cos(4\pi t/365)$ for the $\frac{2}{\text{year}}$ frequency, and $\sin(6\pi t/365)$ and $\cos(6\pi t/365)$ for the $\frac{3}{\text{year}}$ frequency). Then for fitting the daily anomaly there are further columns, where each column represents data from a single day. Rows (i.e. observations) that represent this day have entry 1.0, all others have entry 0.0. For fitting the horizontal anomaly (which is constant in time) there are columns, where each column represents a horizontal location (latitude × longitude resolution of 0.1° × 0.135°). Each row (i.e. observation) that represents this location has entry 1.0 and all other rows have entry 0.0.

P26line510: The authors mentioned that K_BG* is the same as K_BG, but set to zero for observations where the wind data suggest a significant impact of the CH4 plume on the satellite data. What do you mean by "a significant impact"?

We calculate the plume signal according to Sect. 2.3, i.e. for each observation we have a

theoretical plume signal. All observations for which we expect a plume signal being beyond the 75% percentile of all plume signal values is defined to be significantly affected by the plume. The 75% percentile is chosen empirically. It ensures that the background calculation is not significantly affected by the plume and that there are still sufficient observations available for estimating the background in a robust manner.

P26line513: I guess the authors is talking about x_BG instead of K_BG?

Here we refer to $\mathbf{K_{BG}}$. Maybe we clarify this by writing: "$\mathbf{K_{BG}}$ is a Jacobian matrix where each row represents an individual satellite observation and each column a component of the background model. The background model considers a smooth background, which is …". This has been added to the revised manuscript.

P26. Eq9: It is also confusing me that you wrote y_BG in Eq.8 but y in Eq.9 without any explanation in the text.

$\mathbf{K_{BG}}^*$ (and thus $\mathbf{G_{BG}}$) is set to zero whenever y_{plume}=0. This means for Eq. 9 we can replace y_{BG} by y, i.e. use y in Eq. 9 instead of y_{BG}, which one would expect from Eq. 8. Actually, y_{BG} is what we want to estimate (see Eq. 12). We see the point of the referee and will add after Eq. 9 the following explanation: "Because $\mathbf{K_{BG}}^*$ (and thus $\mathbf{G_{BG}}$) is set to zero whenever y_{plume}=0, we can use in Eq. 9 y instead of y_{BG}."

P26 line 519: How do you create the Sy,n and Sa matrices? If I understand correctly, these are key parameters for your y_BG calculation.

Although the authors said that" The matrix $\boldsymbol{S}$y,n stands for the noise covariance of the satellite data ", where are the noise of the satellite measurements come from? from the satellite L2 data?

For Sa "with a very low constraint value for the coefficient determining the constant and higher constraint values for the other coefficients". What do you mean 'a very low'? What are the diagonal values for each retrieval parameter?

Thanks for these important comments – we should have been more explicit on these points.

Both $\mathbf{S}_{y,n}$ and $\boldsymbol{S_a}$ are diagonal matrixes. For $\mathbf{S}_{y,n}$ the noise comes from the satellite measurements, i.e. the XCH_4 precision of the satellite data.

The diagonal values of $\boldsymbol{S_a}$ are (20 ppb)^2 for the coefficients representing the seasonal cycle, the daily anomalies and the horizontal anomalies. The diagonal value of $\boldsymbol{S_a}$ that represent the constant CH_4 values are set to (10000 ppb)^2 and the diagonal value of $\boldsymbol{S_a}$ that represents the linear temporal increase is set to (1 ppb/day)^2.

Concerning the uncertainty treatment (Eq. 11, 14, 20, 21) we improved the related text. In addition, please note that in Eq. 12, 13, 14 and 20 there were typos, where $\mathbf{K}$ should be $\mathbf{K_{BG}}$. This has been corrected in the revised manuscript.