Author response – Editor

We thank the editor for their comments and feedbacks. We respond to them below: the comments are copied hereafter and shown in black, our author responses in blue.

You have done substantial work responding to the previous rounds of review. Two reviewers are satisfied. The other two reviewers acknowledge your efforts as well, yet raise important questions that need to be addressed before publication.

A tension revealed by your study, that inversions optimized for satellite observations do not always improve agreement with surface observations, is important. It is also, as you have noted note, not fully resolved. This is not a flaw; it is honest, and the community needs to understand. The reviewers have asked you to go further in exploring why this is the case and what it means for the usability of your products.

Please address the following points in your revision:

Model errors: The reviewers ask for greater clarity about how model errors (transport, boundary layer representation, tropopause height) may interact with retrieval biases. You discuss these separately, but the question is how they work in tandem. A more explicit discussion of this coupling, even if it is speculative, would help readers understand what inversions can deliver and what their limitations are.

A clarification on the model errors has been added as a response to RC3 and in the main text. It is a broad and complex topic, with a limited number of quantitative analyses. There are multiple links between retrieval errors and the atmospheric conditions as well as between the atmospheric transport model errors and the atmospheric conditions: correlation between retrieval and transport errors for a given pixel at a specific date; similarity of horizontal spatial patterns and temporal patterns of errors; impact of vertical transport on errors (particularly important for CH_4 is the representation of stratosphere-troposphere exchanges and the tropopause height). The high complexity and multiplicity of these relationships makes this topic extremely difficult to tackle, and even to discuss in a relevant way.

As an example, a specific air mass with relatively high aerosol and methane concentrations can generate high retrieval errors over its horizontal extent while the transport model could erroneously shifts this extent: in this case, it would be extremely difficult to build a relevant discussion on the coupling between the retrieval and atmospheric transport errors. How closely do the spatial patterns of transport model and retrieval errors follow the horizontal patterns of the atmospheric dynamics, and with which level of correlations between the two types of errors ?

The derivation of suitable statistics of the atmospheric transport model errors is already a highly complex topic. In practice, it is based on ensembles of simulations with varying models and input parameters (Locatelli et al., 2013, Szénási et al., 2021), or using the residual error method (Heald et al., 2004, Sheng et al., 2018, Chen et al., 2023). The results are generally summarized as statistics and the analyses hardly attempt at mapping the errors or to characterize them as a function of the dynamical patterns, so that we can hardly confront maps of transport errors with maps of retrieval errors for a practical inference of their correlation.

The derivation of spatial correlations in the model errors is therefore highly challenging, and these errors are usually (like in this study) modeled without correlations in the inversion configurations.

Here, **R** is diagonal and without model errors. Even if model errors were considered, the **R** components would be the sum of an observation error $\mathbf{R}_{obs}$ and a model error $\mathbf{R}_{mod}$, without correlation terms. These errors would be independent, thus adding an average model error would not change patterns in the outputs of the inversions, only reduce the fit to the observations. The analysis of the potential spatial coherence between the model errors and the retrieval errors would thus hardly be exploited in the current inversion configurations. This complex topic goes beyond the scope of our study.

Spatial and temporal validation at native scales: One reviewer's request for a grid-by-grid, time-by-time comparison is well-founded. You argue aggregation smooths local errors, but such smoothly also obscures the reliability of the retrievals at the scale needed for attribution. A representative analysis (for example, scatterplots for a few key regions or seasons) would help the community understand the magnitude of local mismatches and whether the spatial patterns are reasonable.

See responses to RC4.

Attribution: The goal of this work is to support mitigation efforts. If the spatial distribution is uncertain, even though the aggregates look good, what does this imply about using these inverse products for attribution of specific emissions sources or regions? A frank discussion, even just a rough quantification of the limits of confidence in attribution, would strengthen the paper's contribution.

See responses to RC4. A discussion to the previous point and this one have been added in the manuscript. Additional figures have also been added.

Seasonality: One reviewer asks whether the spring peak in satellite inversions appears in TCCON comparisons, and this is worth pursuing. Even a brief seasonal analysis for a few key sites (as suggested) would clarify whether this is real or an artifact.

The analysis has been performed for a few TCCON sites (see the response to RC3). The associated figures have been added in Supplementary material, and a discussion on the issue has been added in the text.

I recognize that some of these questions may not have complete answers. You are working at the frontier of what is possible when interacting with such a complex system. The goal is not to resolve everything, but to be transparent about what we know, what we don't know, and what this means for end users of your products.

Thank you again for your careful and thoughtful work. I look forward to seeing the revised manuscript.

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Author response – RC3

We thank the reviewer for their comments and feedbacks on the manuscript. We respond to them below: the comments are copied hereafter and shown in black, our author responses in blue.

General comments:

The study looks at the use of 3 different Tropomi XCH4 retrieval products in regional methane flux inversion for Europe. Authors embarked on solving a set of complex problems involving the biases of the retrieval products, transport model errors and uncertainties of the inverse model results. They try to trace down the flux differences to the drivers of XCH4 biases in the respective products, ending up in a complex analysis, involving use of ML approach following Balasus et al. 2023. As the reviewers of the original manuscript mentioned, the resulting report was not easy to read and extract the essential results. The problem was not fully resolved in the revised manuscript, but as noted by review reports, the study does contain a lot of useful analysis and is very much worthy of publication.

Detailed comments:

Authors faced number of complications to the analysis origination in several problems among which only some could be addressed.For clarity and readers' convenience it would be useful to state what is investigated in detail (eg contribution of aerosols and other variables to L2 biases), and what is not, such as transport model errors and biases. Among those, one can count on contribution of the tropopause height variation impacting observed XCH4 (eg Saad et al 2014) that could be miscalculated by CHIMERE.

References: Saad, K. M., et al: Derivation of tropospheric methane from TCCON CH4 and HF total column observations, Atmos. Meas. Tech., 7, 2907–2918, https://doi.org/10.5194/amt-7-2907-2014, 2014.

Following your recommendation, a list of the problems that are addressed in the study has been added. It is first mentioned in the introduction of the manuscript, and further detailed in Section 3.3. It is finally discussed in the conclusion.

The main objective of this study is to assess the consistency of the emissions estimated from inversions using 3 TROPOMI products, and how the differences in the products impact the spatial and temporal patterns of the emission estimates. We provide the following analyses:

- We compare the results of the inversions using the 3 TROPOMI products. We investigate the differences in the posterior emissions by describing the differences between products (coverage, XCH4 differences, observation errors, column averaging kernel, prior CH4 profile), and evaluating the contributions of aerosols, albedo, striping patterns and other retrieval variables to the XCH4 differences.
- Using OSSEs, we try to disentangle these drivers and quantify the impact of the differences in coverage, observation errors and XCH4 differences on the emissions. We also evaluate the impact of the background optimization on the posterior emission results, as part of the inversion system.
- We compare the amplitude of the differences with external evaluation (surface station measurements, TCCON observations).

Some limits of the analyses are highlighted as potential causes of discrepancy, but not deeply investigated:

- Albedo and aerosol-related biases on the estimated emissions are discussed qualitatively (paragraph « SRON-WFMD XCH4 differences » of section 3.4.2 partially addresses this issue as SRON and WFMD treat differently such biases) but not quantitatively estimated. It is suggested as a next step.
- A detailed study of the transport model errors (miscalculation of the tropopause height, representation of the boundary layer, transport errors at meso-scale...) and on how they interact with the errors related to the retrievals is not performed, as it is a complex topic that goes beyond the scope of the study (see response to the editor).

Fig. 4 Caption. Here one can use “Column averaging kernel” instead of simply “Averaging kernel”

The change has been done, in the figure caption and in the text.

Fig 10c Inverse modeling points to difference between Tropomi and surface data inversion in spring, could it be also found in TCCON comparison? Suggest adding seasonally resolving version of Fig C (for some key sites could be be OK) The domain average XCH4 seasonality on Fig 2a, does show same difference between products, which is not reflected in fluxes, as products with different seasonality happen to produce similar fluxes.

The peak of high CH4 fluxes seen in Fig. 10c in the results of the 3 inversions using TROPOMI data cannot be seen in the corresponding XCH4 observations (Fig. 2a), as mentioned in Section 3.3. Investigations were carried out to identify the cause of the positive corrections in Western and Central Europe associated with this peak, regarding the observations and the background optimization. Yet, it has not been fully clarified.

To compare the spring variations of XCH4 observations and TCCON total column measurements, a seasonal version of Fig. C1 has been added in the supplementary material (Figures S5, S6). A short discussion has been added in the corresponding Annex C, where the comparison TROPOMI/TCCON is made. The figure is shown below (Fig. 1) and includes the sites of Garmisch, Karlsruhe, Orléans, Paris and Sodankyla, where they are a sufficient number of co-located observations (except in JFM for Sodankyla).

Figure 1 indicates a low seasonal dependency in the differences, apart from Sodankylä. In spring (AMJ), the TROPOMI-TCCON difference is in average always positive (except for BLENDED in Paris, slightly negative), which indicates that TROPOMI XCH4 estimates are higher than the ones of the TCCON stations. However, this is also the case in winter (JFM), whereas the fluxes from inversions are not peaking in February and March 2019. A more detailed look at monthly resolution in Figure 2 shows high positive XCH4 difference in April in Karlsruhe and Sodankylä, but no clear evidence of a peak of the TROPOMI-TCCON difference in the April-May period. In Sodankylä, the TROPOMI-TCCON difference is positive during spring 2019, and negative in autumn 2019, consistent with the findings of Lindqvist et al. (2024).

Therefore, the comparison between TROPOMI and TCCON suggests positive difference in winter and spring, thus one would expect higher fluxes from TROPOMI-based inversions in comparison to a TCCON-based inversion. But the sping peak cannot be clearly identified in this comparison, suggesting the peak’s origin is partly related to the inversion system in itself.

References:

Lindqvist H, et al. Evaluation of Sentinel-5P TROPOMI Methane Observations at Northern High Latitudes. Remote Sensing. 2024; 16(16):2979. https://doi.org/10.3390/rs16162979

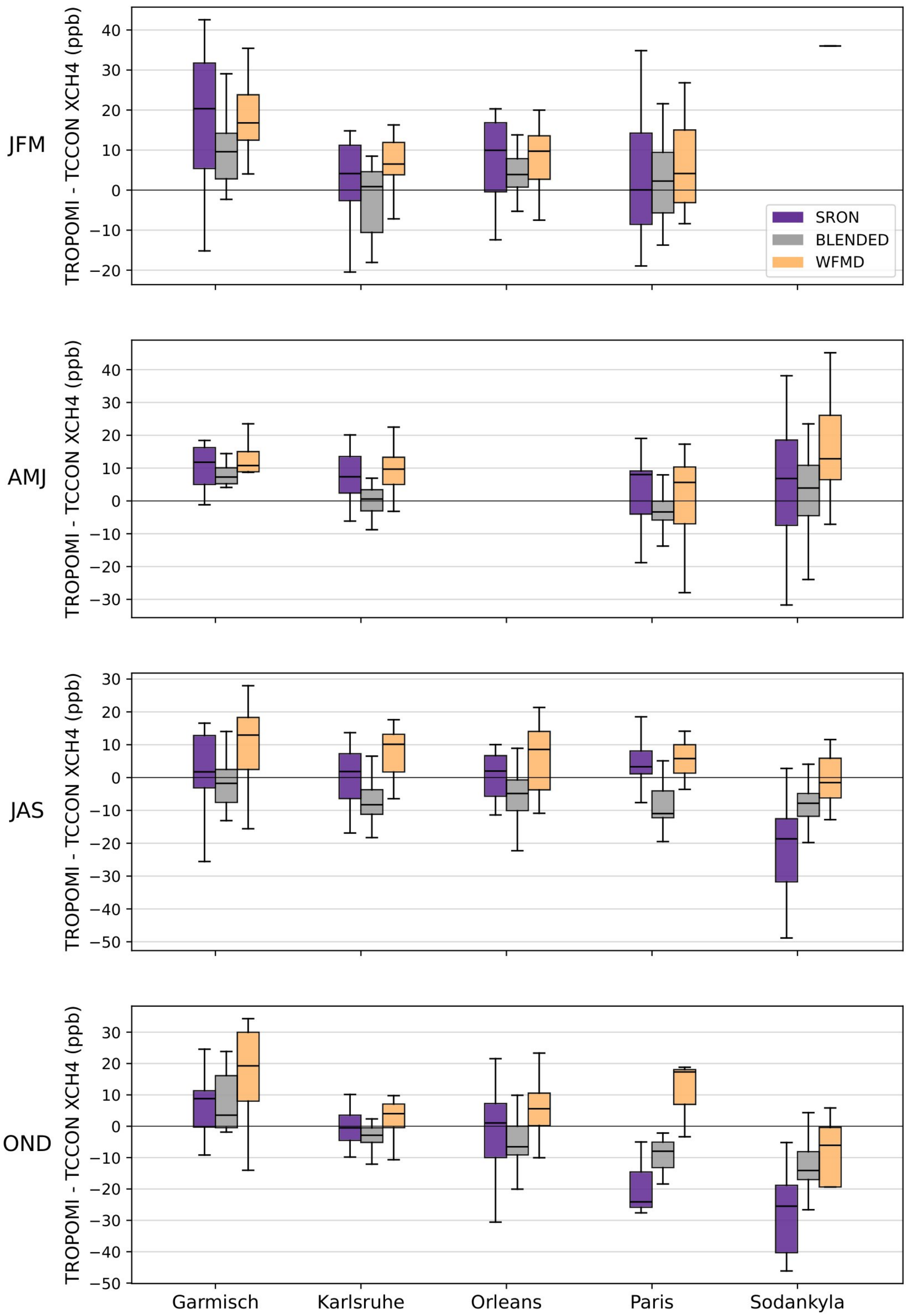

Figure 1: Similar as Fig. C1, but seasonally-resolved. Median and quartiles of the daily averaged differences between TROPOMI and TCCON XCH4 (ppb) for 5 TCCON stations, for all seasons in 2019.

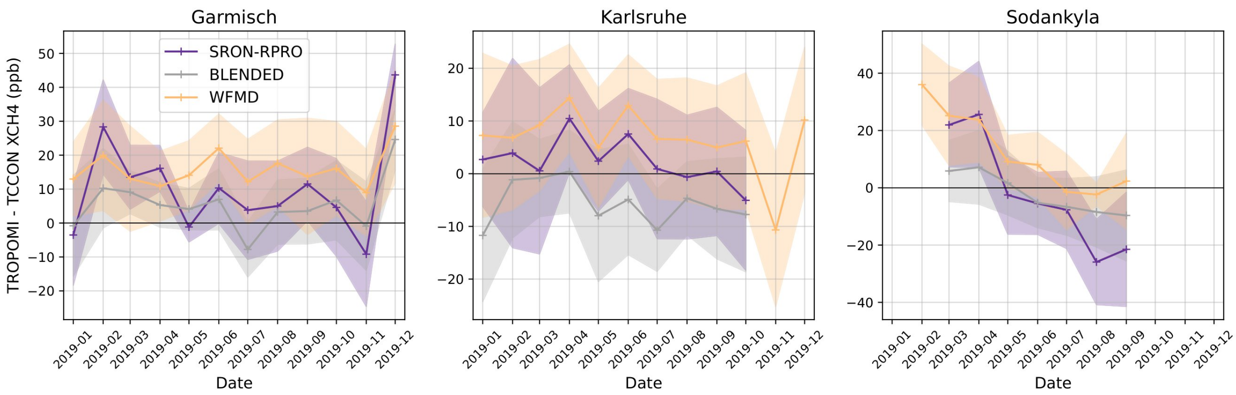

Figure 2: Time series of the monthly averaged differences between TROPOMI and TCCON XCH4 (ppb), for 3 stations. The shaded area shows the standard deviation within each month.

Author response – RC4

We thank the author for their comments and for the discussion. We respond to them below: the comments are copied hereafter and shown in black, our author responses in blue.

This study provides a valuable reference for regional emission quantification driven by satellite data by comparing the inversion results of three TROPOMI products. The existing work is relatively complete, and the previously asked questions have been addressed reasonably well. However, there are some outstanding points which must be considered further.

1. The only way to get a national scale or regional scale budget is to sum all of the independent sources together. In your case, your model has a resolution of 0.5 x 0.5.You need to compare day-by-day, 0.5 x 0.5 grids with existing known emissions from medium and large surface sources.

We disagree with this statement. If the only way to get a national scale or regional scale budget were to sum all of the independent sources together, all CH4 atmospheric inverse modelling studies and efforts to develop new CH4 satellite missions should focus on point source studies with very high resolution images of the plume downwind to these sources. The local analysis is definitely the most sensible for large point sources with well identified plumes in the satellite images. However, the inverse modelling community has also shown capabilities to solve for the aggregation of strong localized sources and diffuse sources (which do not generate clear patterns locally) over large scales with moderate resolution observations (e.g. from GOSAT and now TROPOMI), and the analysis of moderate but smooth variations across continents and countries (see the introduction of the article for regional and global studies of CH4 emission mapping). The integration of CH4 signal through atmospheric mixing often provides good capabilities to monitor budgets of emissions over large regions while limiting those for attributing such an overall emission budgets at finer spatial scales, and in particular at the model spatial resolution, and this phenomenon is exacerbated by the limitations of the spatial and temporal coverage of the satellite data. The statistical inversion noise (combining the observation and model errors) enhance the difficulties to retrieve robust emission estimates at the horizontal resolution of the transport model.

Also, sources with individual facility-level methane emissions greater than approximately 10 t/h are estimated to contribute 8 %–12 % of total global oil and gas production methane emissions (Lauvaux et al., 2022), but have larger contributions locally (Cusworth et al., 2026). Point sources estimates are thus so far limited in their potential to assess national to regional scale budgets, with current observing high resolution capabilities (e.g., Tanager, GHGSat, Emit, EnMAP); dedicated studies are ongoing to find ways to upscale point source estimates to larger scales in a relevant way but these are the work of the upcoming years for the community and will likely justify a large number of dedicated publications. Inversions focus on the smaller emission sources, that account for a large proportion of anthropogenic emissions (Williams et al., 2025), as well as diffuse anthropogenic and biogenic sources. In particular, it makes sense for Europe which has only a few basins with large localized sources (e.g., cf CAMS Hotspot Explorer; https://apps.atmosphere.copernicus.eu/methane-explorer/), whereas most CH4 signals are diffuse and coming from various sources.

For these reasons, both point sources and regional/global inversions are relevant and complementary in their use of the information content of satellite observations. Our publication focuses on the use of satellite images in regional/global inversions and assess limitations specific to regional/global inversions, that are not necessarily critical in hotspot studies. Indeed, retrieval differences will have a lesser impact when studying plumes with very strong signals, whereas in inversions, gradients are smaller and even small observation deviation critically impact inversion results, which is the point of our study.

There is on-going effort to bridge the gap between local point-source estimates to regional and meso-scale inversions (Naus et al., 2023, Visser et al., 2025). The study of Santaren et al. (2021) explores how « atmospheric inversions may have the potential to exploit spatial variations in XCO2 fields to quantify regional to national budgets of more diffuse sources or of all types of sources, even when the overlapping of several plumes prevents the quantification of the emissions from individual cities and point sources ». This is still a large open topic, but goes beyond the scope of this article.

Regarding the comparison with known emissions, gridded inventories such as the one we use as a prior estimate of the anthropogenic emissions (EDGARv8) represent state of the art compilation of “medium and large sources”, ingesting as much local information as possible for such a compilation and mapping efforts. This often results in the application of average emission factors and uncertain data on local activites when not only a rough spatial disaggregation of national totals with activity proxies. Local gridded inventories would bring more robust information on local sources but would suffer from the same types of limitations, and (to our knowledge) there is no systematic robust estimate of local sources based on observations over areas that would exceed the scale of our model grid cells (approximately 50km x 50km). Therefore, we hardly see a better confrontation to other emission estimates at our inversion scale than that to gridded inventories such as EDGAR (used in this study) or other similar inventories, which is done in the comparison between prior and posterior emissions.

2. A scatterplot of grid-by-grid, time-by-time is required.If this can be analyzed country-by-country or emissions type-by-type, then this is better, but not required. We need this information to understand if the retrievals are real at the retrieval level, or if the averages make sense for the wrong reasons. The combination of 1 and 2 will ensure that your day-by-day, grid-by-grid emissions are reasonable.

We provide scatter plots of posterior emissions against prior emissions and other posteriors, in the matrix of Figure 3. Each scatter point represent a given date and pixel. This can be used to understand the distributions relatively to others. A second plot is made by aggregating the country yearly budgets and comparing posterior and prior budgets (Figure 4). It shows better consistency at the country scale, yet some differences remain: highlighting the fact that results from the inversions of the TROPOMI products are not fully consistent even on aggregated scales. This is exactly the point of our study to express reserves on using TROPOMI data as such in regional inversion, as has already been published in various papers, including very recently in ACP (e.g., Hancock et al., 2025). We recommend further investigation and caution on regional inversions.

These plots have been added in the Supplementary material. References to the figures and the corresponding discussion has been added to the main text. We have also clarified the scope of the study, and the relevance and the limits of the attribution of posterior emissions to budgets across the different scales.

3. A specific analysis of this should be done in space and time, to ensure that the retrievals are robust over all times of the year. Please check.

The space and time analysis is presented through the seasonal maps of Figures S1, S2 (in the Supplementary material) as well as with the time series of Figure 10c. Given the fact that yearly and country-scale emission totals show already substantial differences across the inversions, we consider that it is not relevant for the conclusions of this study to show detailed analyses at both higher temporal and spatial resolutions at the same time. Given the large number of plots that would be required, we prefer not to extend even more the (already long) article.

4. If they are not reasonable in space and time, and their sum is reasonable, you still cannot do attribution. This is the point of such an inventory. We want to know from a quantitative perspective, if attribution is possible or not. Based on the uncertainty of the emissions from 1-3, please quantify how reliable the product is in terms of attribution.

In our study, we compare posterior estimates of the emissions to the a-priori information coming from bottom-up (BU) inventories. The assimilation of satellite observations of XCH4 provide complementary signal for the mapping of emissions at a 0.5° resolution, as mentioned in the previous responses. In this process, aggregating spatially and temporally the emissions decrease the uncertainty on these emissions.

Previous studies have assessed the finest time and space resolution at which information can be retrieved from the available atmospheric data into the emission space (e.g., Pison et al., 2018). Similarly, studies with OSSEs on point sources (Naus et al., 2023) showed that « daily estimated (point source) emissions have large errors (50%), but these errors mostly average out on monthly and annual scales of 25 and 15%, respectively ». Following this idea, the attribution of emissions can be difficult at the pixel-scale, but still relevant at coarser scales (country, region). This is what we do in the study: we discuss the consistency of top-down emission results at different scales, using different products retrieved from the same L0 measurements. Our work shows a lack of consistency of the emissions at country-scale, therefore comparing products at a finer (i.e. pixel-) scale, and attributing emissions at a finer scale, are beyond the scope of our study and not relevant.

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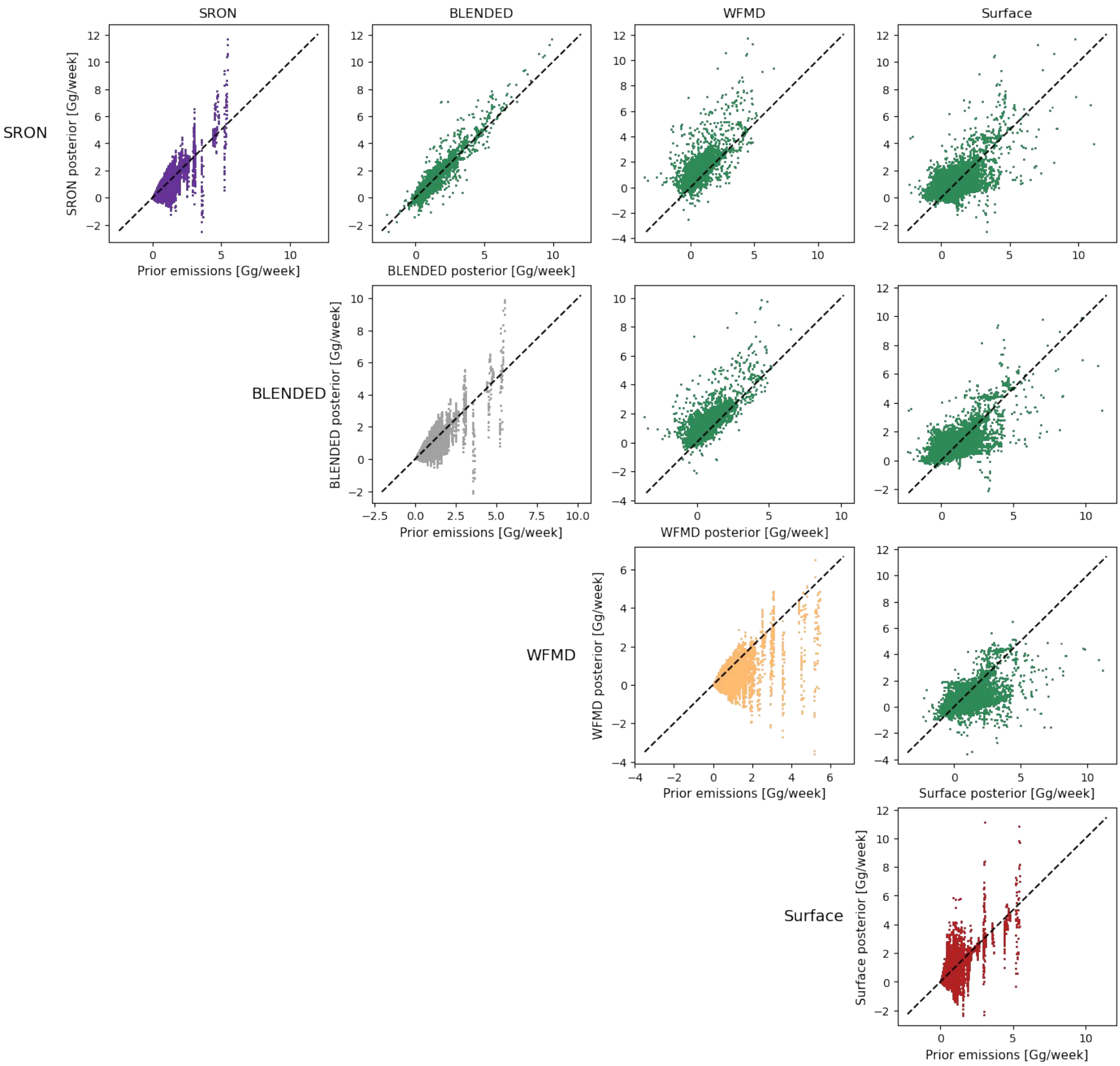

Figure 3: Scatter plots of emissions in the different inversions. The diagonal plots show posterior against prior emissions. The non-diagonal plots show a comparison of the posterior emissions. Each scatter point represents a pixel of 0.5°x0.5° and one week, i.e. One element of the control vector. The dashed line is the 1:1 line.

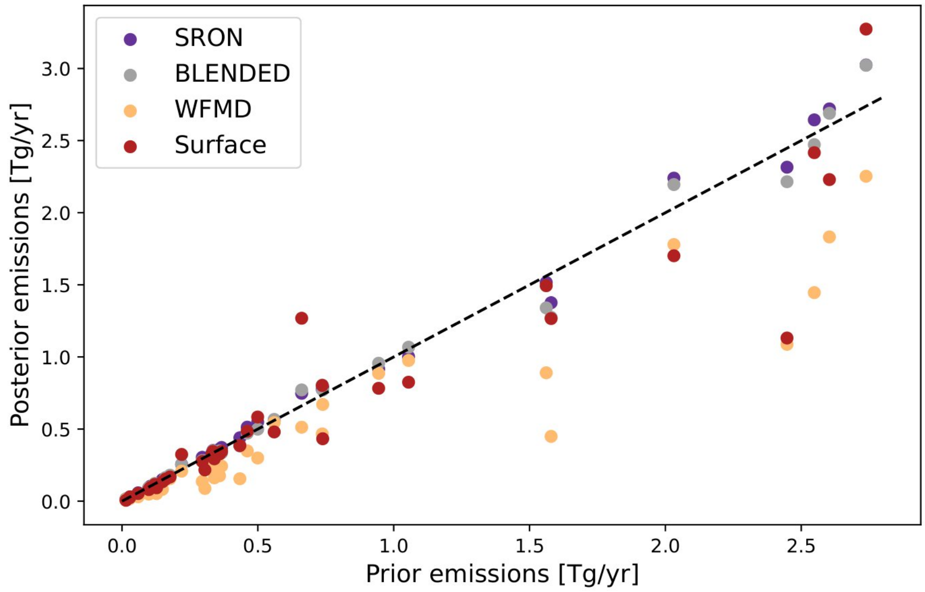

Figure 4: Scatter plot of the yearly and country averaged CH4 prior and posterior emissions, across the different inversions. Each scatter point represents a country.

5. Please follow-up on the response to the fifth question of RC4 for Response-Version 3.

(1) Is the scaling factor for all 8 grids the same across each different posterior? Or is the total amount scaled differently?

If we understand correctly, you ask whether the relative contributions of the 8 pixels in the area vary across posteriors, or if it remains constant and only the total of the emissions varies.

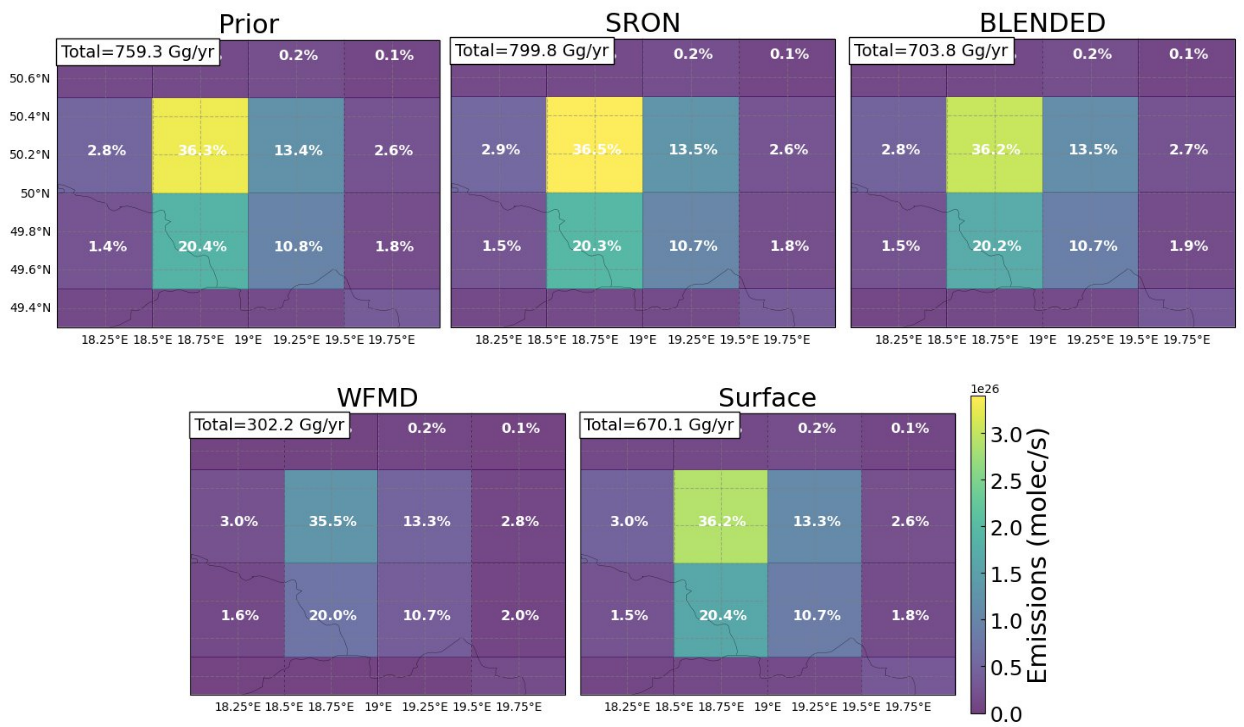

Figure 5: Maps of prior and posterior CH4 emissions for the 4 inversions, over the USCB region studied in Tu et al. (2022), in 2019. Emissions are in molec/s. The contribution of each pixel to the total is written in white.

Figure 5 shows the values of the total prior/posterior emissions and their relative contribution for each concerned grid cells (white figures, in % of the total). The changes in the relative contributions are very small, it is mainly the total over the region that is scaled. It is consistent with what has been said before: the inversions are not designed to attribute very local emissions, but to monitor emission budgets over large regions.

(2) how spatially does this map compare with Tu et al., 2022?

Following what has been said in the response to the first 4 questions, it is not relevant to compare results of our posterior emissions to the results of Tu et al. (2022) at the pixel-level. Given the coarse resolution at this scale, we do not expect the inversion to attribute correctly the emissions from one point-source to the other. Emissions can be compared at the scale of the entire basin to an inventory of the emissions in the area, but not at the scale of point-sources.

Also, as mentioned in the previous response, the study of Tu et al. (2022) has been performed in the year 2021, and extrapolating the emissions to another year (2019 in our case) may introduce errors; indeed emissions in Eastern European countries often vary significantly from one year to the next one, as reported in Romania by Stravapoulou et al. (2023) or Kulhmann et al. (2025). Moreover, the prior estimates of emissions that we use (anthropogenic emissions from EDGAR v8) are far higher than the estimates of Tu et al. As we perform Bayesian inversions, that computes the most probable state given the a-priori knowledge, it is not relevant to compare too deeply the results in the case of large differences in the priors. What we can say is that the total emissions are decreased to values closer to the estimates of Tu et al.

References:

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