Author response – Editor

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

This work has accomplished a lot and produced both interesting and important findings. However, there are still some outstanding issues of concern which need to be addressed. Looking forward to your updated version, which works to address the following issues:

1. More depth with respect to model uncertainties and measurement uncertainties.
2. Comparisons with external observational datasets (which do exist)
3. A deeper explanation of the realism of the results compared with on the ground reality
4. Analysis probing/discussing what weaknesses currently may need to be addressed by future works.

A major issue arises from the fact that your observational data has both (a) far higher spatial and temporal resolution than your model, yet (b) is still coarse when compared with surface observations-especially from large point sources like coal mines.

Therefore a deeper analysis (or least discussion) needs to be done which considers how much information and/or precision are lost due to the aggregation of the observations into the model environment, subsequent modeling and its uncertainties, and the final disaggregation and comparison with on-the-ground reality. For example, if your model is relying on an emissions dataset which has “zero emissions” on a grid which in reality has a “large source”, how is this uncertainty propagated through your system? I specifically raise this point because the reviewers have hinted that this is what may be occurring in actuality.

Overall, we have tried to address as much as possible the elements you mention, while keeping the focus of the study on the relative comparison of emissions obtained from the assimilation of satellite XCH4 datasets. We have added added references to relevant work and improved the discussions on several topics: the emission uncertainties and the ground reality, the need to bridge the gap between local studies and regional studies, the comparison with surface stations. We detail below these responses.

1. The question of uncertainties, for both model and measurements, is indeed a crucial question. In variational inversions, uncertainties are provided as inputs in the **B** and **R** matrix. The model error and the uncertainties on the a priori emissions is contained in the **B** matrix. The error is assumed to be 100% for CH4 emissions (see Section 2.4.2). Since the uncertainty is not provided in the a priori emission datasets, the errors have been chosen based on values from the intercomparison of CH4 inversions in Europe (Ioannidis et al., 2025), and from the study of Sénasi et al. (2021). The **R** matrix contains observation errors, and has been built with the individual retrieval errors provided in the satellite/surface observation datasets.

The uncertainties on posterior emissions are not directly accessible in our 4D-Var inversions. As discussed in the « Author response to referee #3 », this lack of uncertainty range intrinsically limits the statistical analysis of the consistency of posterior emissions. However, similar configurations of the CIF-CHIMERE inversion system have been used for all the inversions, thus we can

address the differences of the spatial and temporal CH4 emission distributions due to the differences in TROPOMI observation products.

Also, the uncertainty reduction (independent of the observation vector) can be estimated through our OSSEs: the ratio of the standard deviation of priors and the one of posterior emissions across the ensemble samples is an estimation of the uncertainty reduction. For the total budgets, it is estimated to 78% reduction for SRON and BLENDED, 74% for WFMD and 51% for surface stations. We do not use these values for setting error bars to our budget estimates, for two reasons: 1) the limited statistics of the ensemble (4 samples) and 2) the lack of proper uncertainty estimation for the prior (not provided with the emission inventories). We have added a discussion on this question in Section 3.4.1 and in the conclusion.

The propagation of the uncertainties in the model (such as your example of a large source not counted in the “zero emissions” pixel on the model grid), as well as the question of aggregation and disaggregation of the inputs/outputs of the model are crucial topics. However, addressing this complex issue would require a dedicated study, as the work presented in Zheng et al. (2025): we feel it goes beyond the scope of this manuscript. We study emission outputs at both pixel, country-scale and multi-country-scale (see Figure 10, Table S1): aggregating the results at larger scales smooth local errors that can arise from these problems. Our study insists on the fact that even at national scale, significant differences in CH4 emissions remain between the inversions based on the different TROPOMI products.

2./3. Comparisons with external datasets have been provided in several ways: comparison of the TROPOMI XCH4 observations with GOSAT observations (Section 2.1.3) and with TCCON observations (Annex C), comparison of the inversions and OSSEs of satellite observations with an inversion using independent surface observations (Sections 3.3, 3.4), evaluation of the CH4 simulated concentrations (prior and posterior) at these surface stations (Section 3.5).

Our study focuses on the relative comparison of TROPOMI satellite products, not on the absolute validation of emissions inferred from TROPOMI inversions. The latter has been analysed in the validation of global (Yu et al., 2023 ; East et al., 2025) or regional (Liang et al., 2023 ; Ioannidis et al., 2025) inversions with one TROPOMI product or the other, but few inter-comparisons of all the TROPOMI products had been performed : an issue that we address in this article.

Local studies provide valuable information on the emissions, and the comparison with regional/global studies is important. Integrating data from hotspots and high emission local areas in regional and global studies is a perspective for future work, and is made possible by satellites as TROPOMI. This is also why it is necessary to have a clear idea of the biases of the regional studies performed with satellite data, the issue that we tackle in this article.

For this study, we have made new comparisons in Poland and Romania as suggested by Referee #4 (see answer to question 5). However, for reasons mentioned in the response and as the manuscript is already quite long, we do not incluse these additional comparisons to surface datasets in the main text. The deeper analyses of the comparisons is mentionned in the text as future work.

4. In the conclusion of the manuscript, as well as in the ends of “Results” sub-sections, we discuss the limits of this analysis and future work :

- Regarding TROPOMI observations: we used the XCH4 datasets with as low post-processing as possible. Further understanding of the biases in the derived CH4 emissions requires corrections of the albedo and aerosols biases highlighted in Section 3.1.
- The characterization of uncertainties related to the background (lateral and top boundary conditions, stratosphere) and its role in the optimization process (Section 3.3). This has been the object of a recent study (Nesser et al., 2025).
- The gap between satellite-based and surface-based inversions, in terms of space-time distribution of the increments and of simulated CH4 measurements (Sections 3.3, 3.5).

5. Regarding the aggregation of observational data with both a higher space-time resolution that the model and a coarser resolution than surface data, two elements can be mentionned.

First, we have performed inversions with super-observations, aggregating observations at the spatial resolution of the model (0.5°x0.5°) and with hourly resolution. The posterior emissions and the simulated observations (prior and posterior) were close (<1% difference), while the patterns of the spatial and temporal distributions remain unchanged. For this reason, we chose to use the entire dataset to remain as general as possible, as "a TROPOMI user would do".

Secondly, the coarser resolution than surface data limits the analyses at local scale: this is why we perform analyses at both the pixel and the country-scale. The objective is not to focus on local point sources, but to include the emissions at larger scales to build country-scale and regional-scale budgets, a complementary approach to local studies.

References:

East, J.D., Jacob, D.J., Jervis, D. et al. Worldwide inference of national methane emissions by inversion of satellite observations with UNFCCC prior estimates. Nat Commun 16, 11004 (2025). https://doi.org/10.1038/s41467-025-67122-8

Ioannidis, E., Meesters, A., Steiner, M., Brunner, D., Reum, F., Pison, I., Berchet, A., Thompson, R., Sollum, E., Koch, F.-T., Gerbig, C., Wang, F., Maksyutov, S., Tsuruta, A., Tenkanen, M., Aalto, T., Monteil, G., Lin, H., Ren, G., Scholze, M., and Houweling, S.: An inter-comparison of inverse models for estimating European CH4 emissions, Earth Syst. Sci. Data Discuss. [preprint], https://doi.org/10.5194/essd-2025-235, in review, 2025.

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Nesser, H., Bowman, K. W., Thill, M. D., Varon, D. J., Randles, C. A., Tewari, A., Cardoso-Saldaña, F. J., Reidy, E., Maasakkers, J. D., and Jacob, D. J.: Predicting and correcting the influence of boundary conditions in regional inverse analyses, Geosci. Model Dev., 18, 9279–9291, https://doi.org/10.5194/gmd-18-9279-2025, 2025.

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Author response – RC 4

We thank the reviewer for their thoughtful comments. We respond to them below: the comments are copied hereafter and shown in black, our author responses in blue; suggested new manuscript text is indicated in green.

This study compares the emissions estimated from variational inversions in 2019 over Europe with a resolution of 0.5˚ assimilating three TROPOMI products of dry-column methane mole fractions (XCH4), which is a valuable and an important work. The results shows that the largest contributions to XCH4 differences may be attributed to aerosol scattering and sensitivity to albedo. And find that seasonal emissions are highly correlated across the inversions. Besides, this study also used the Observing System Simulation Experiments (OSSEs) to disentangle the drivers of differences between the posterior emissions, and reveal that observation density and errors, averaging kernels and prior profiles play a key role in the inversion's capacity to constrain the emissions. Overall, the insights of this study are valuable, and the conclusions drawn are helpful to other studies. I recommend publication in ACP after addressing the following general and specific comments.

General comments

1. Line 178, "average concentrations decrease slightly from January to May, reaching a minimum in April/May", does this pattern have anything to do with the monthly observations? Since from figure 1(a), the observation count decreased from April, and reaching a minimum in May. From Figure 2(a), monthly XCH4 reach a peak in November, while from figure 1(a), there is a minimum observation count in November, how does the observation (sampling difference) affect the results?

The sampling difference indeed affects the temporal and spatial averages of XCH4. The peak in November is partially explained by the decrease in observation count for this month, and especially in the low observation count in the northern latitudes where XCH4 is lower.

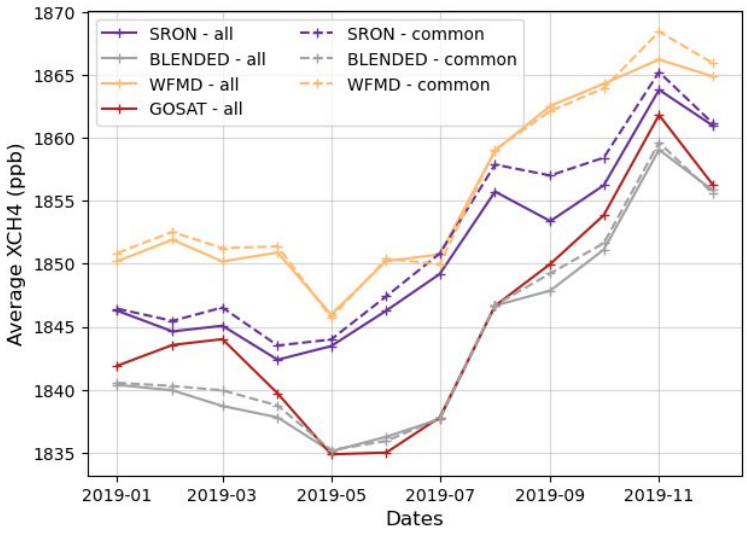

Figure – Similar to Figure 2 (a), including the TROPOMI products restricted to common observations (dashed lines).

To disentangle the effect of the differences in XCH4 variations and in the sampling variations, we compare the TROPOMI products restricted to the common observations across the 3 products (see the remark L.176 on the average values). The time variations are compared in the figure below, they slightly differ between « all observations » and « common observations » but overall the shapes remain similar. A remark has been added in the manuscript to clarify this point.

2. As can be seen from Figure 10(c), only the satellite has a seasonal change, but there is no obvious change between the ground observation and the priori, and the change trend of the surface observation from March to June is not the same as the change trend of the satellite. How can the accuracy of the satellite results be verified if both surface observations and prior comparisons are inconsistent with the satellite results? How to verify the effectiveness of the product?

In this study, the inversion assimilating independent surface observations (from the ICOS, WDCGG, NOAA ESRL and EBAS databases, see Section 2.1.2) is used for comparison. The TROPOMI-based and surface-based emissions show higher emissions in winter and a minimum in summer: satellite and surface inversions are rather consistent, but they are not consistent with the prior. However, the satellite-based inversions show a peak in April and May that is not seen neither in the surface-based posterior emissions nor in the prior emissions. As mentionned L.465, the origin of this peak has been investigated but we provide only partial explanations.

The main focus of this article is on relative differences between products, thus the comparison of the results of surface and satellite inversions has neot been extensively analysed. It has been pinpointed as a challenge for future work, to reconcile the “satellite perspective” and the “surface perspective”. Indeed, surface and satellite observations have intrinsic differences (in-situ vs. remote sensing, localized vs. global, uncertainties...). These differences lead to different constraints on emissions in the inversions. The consistency of satellite-based and surface-based inversions is necessary to evaluate the realism of posterior CH4 emissions.

This challenge is also highlighted by the comparison of prior/posterior observations at the surface stations in Section 3.5 (see comment #6). The analysis should be extended to a larger set of prior emission and observation datasets (other satellite/surface datasets). The use of ground-based remote sensing observations (such as TCCON or COCCON) could also provide valuable data to bridge the gap between surface and satellite inversions. We have improved the discussion in the manuscript, but an extensive dedicated study would go beyond the scope of the article.

3. Figure S3 reveals substantial spatial variability which on a pixel-by-pixel basis is a lot larger than the changes on a country-by-country basis given in Table S1. This needs to be explained more deeply. Some of these changes look to be 100% or more. Why has the optimization not captured these very well? What impacts could this have for new or emerging emissions sources? Is the optimization weighted individually pixel-by-pixel? Could there be some issues in the a priori spatial temporal distribution, and therefore the model error is much larger than currently considered?

Figure S3 shows the absolute increments for the OSSE scenarios, while Table S1 shows increments on total country emissions for the 4 inversions. These two figures should not be compared : synthetic pseudo-observations are assimilated in the OSSEs, while real observations are assimilated in the inversions. Moreover, increments are shown as absolute values in Figure S3 to capture their intensity. But country emission budgets in the inversions are the sum of positive

and negative increments, thus smoothing high pixel-to-pixel variations. So I'm not sure I understand the objective of your first question.

The optimization is weighted individually pixel-by-pixel, with spatial and temporal correlations of errors (see Section 2.4.2). The model error is set to 100% for CH4 emissions. Uncertainties are not provided in the a priori inventories, so the errors have been chosen based on values from the intercomparison of CH4 inversions in Europe (Ioannidis et al., 2025), and Szénasi et al. (2021).

References:

Ioannidis, E., Meesters, A., Steiner, M., Brunner, D., Reum, F., Pison, I., Berchet, A., Thompson, R., Sollum, E., Koch, F.-T., Gerbig, C., Wang, F., Maksyutov, S., Tsuruta, A., Tenkanen, M., Aalto, T., Monteil, G., Lin, H., Ren, G., Scholze, M., and Houweling, S.: An inter-comparison of inverse models for estimating European CH4 emissions, Earth Syst. Sci. Data Discuss. [preprint], https://doi.org/10.5194/essd-2025-235, in review, 2025.

Szénási, B., Berchet, A., Broquet, G., Segers, A., van der Gon, H. D., Krol, M., Hullegie, J. J., Kiesow, A., Günther, D., Petrescu, A. M. R., Saunois, M., Bousquet, P., and Pison, I.: A pragmatic protocol for characterising errors in atmospheric inversions of methane emissions over Europe, Tellus B: Chemical and Physical Meteorology, 73, 1–23, https://doi.org/10.1080/16000889.2021.1914989, 2021.

4. Figure S1 reveals an observation count. However, I am still not sure about how many days' worth of observations there are in each grid. For example, if a model-sized grid has one TROPOMI pixel each day for 10 days, and another model-sized grid has ten TROPOMI pixels on a single day, then would both cases count as having the same observation count? This map is confusing because it is merging together the spatial and temporal counts. In reality, the value of these two different cases would be very extensive. They would have substantial differences in terms of wind, topography, aerosols, clouds, surface reflectance, and other issues. I am not sure if I can believe the model uncertainty until this can be disentangled.

This map has been though to give the reader an idea of the variations of the spatial distribution of the observation count along the year, and how they compare between products. Indeed, in both cases you mention, the 10 observations would be counted the same way : space and time counts cannot be fully disentangled in one map. A second set of maps (Figure S1b) has been added, showing the number of days with at least one observation per model pixel. The combination of both figures gives a broader understanding of the observation count distribution.

However, the content of the map is not what is provided as an input to the model: the model takes into account each observation individually, with the associated date and location, thus the associated topography, wind, etc. This figure is not related to the model uncertainty.

5. I noticed that you do not use any of the surface observation constrained emissions calculated from coal mines in Poland (Tu et al., 2022). How do your computed emissions compare with these results? Since they are independent from your inversion, such a comparison would provide a truly independent comparison.

Comparing the emissions from our inversions to the ones from coal mines in Poland would provide a relevant comparison (not independent though, as TROPOMI XCH4 is used to constrain the emissions in Tu et al., 2022). We propose the two following comparisons at local scale, in Poland and Romania. Please note that the coal mining (Poland) and oil and gas production (Romania) emissions can vary from one year to another, making it hard to compare different years.

Poland: in the Upper Silesian Coal Basin in Poland (49.3-50.8°N, 18-20°E), emissions retrieved from TROPOMI XCH4 (SRON product) and TROPOMI + IASI TXCH4 are respectively about 496 kt/yr and 437 kt/yr, and they are compared to the coal mining emissions from the E-PRTR inventory (448 kt/yr) and the CoMet inventory (555 kt/yr), for the year 2021.
For the same region (see figure below), total methane missions in 2019 are 722 kt/yr for the prior (coal mining emissions are from EDGAR v8), 762 kt/yr for the posterior of SRON, 670 kt/yr for BLENDED, 289 kt/yr for WFMD and 638 kt/yr for the surface-based inversion.
Except for WFMD, total emissions are higher than those from Tu et al. (2022), because the prior of EDGAR has higher emissions. While SRON leads to increased posterior emissions, the Surface and BLENDED inversions are consistent and lead to total emissions closer to the results of Tu et al. (2022). WFMD leads to very low emissions in comparison to the others, as the specific pixels of this region have very strong negative increments. These locally high gradients may be unrealistic. This is also the case for the next case in Romania (see the increments on Figure 10a).

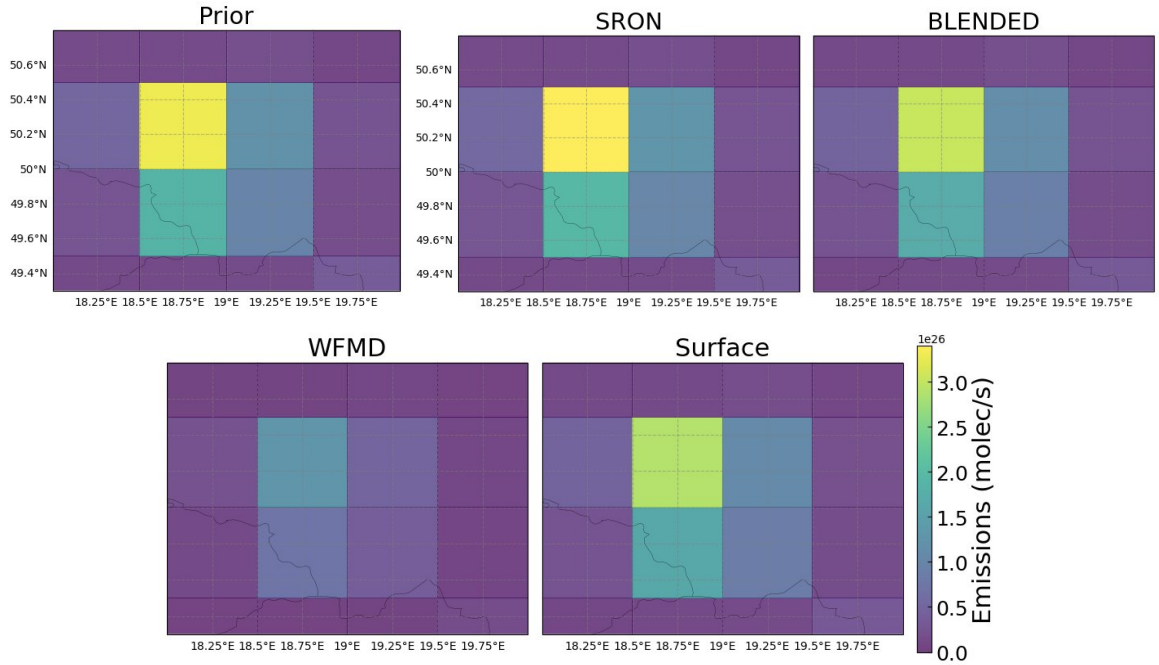

Figure – Maps of prior and posterior CH4 emissions for the 4 inversions performed in this study, over the USCB region studied in Tu et al. (2022), in 2019. Emissions are in molec/s.

Romania: in the study area of the ROMEO campaign in Romania, emissions retrieved from the airborne measurements are respectively about 227 kt/yr in 2019 (Maazallahi et al., 2025) and from 76 to 175 kt/yr in 2021 (four scenarios in Kuhlmann et al., 2025), to compare to 71kt/yr in 2019 and 66 kt/yr (national inventory report for UNFCCC), and 80 kt/yr for IEA.
For a similar region (44-45.5°N, 23-27°E), total methane missions in 2019 are 292 kt/yr for the prior (mainly from EDGAR v8), 256 kt/yr for the posterior of SRON, 240 kt/yr for BLENDED, 56 kt/yr for WFMD and 235 kt/yr for the surface-based inversion.
Except for WFMD, every inversions make emissions closer to the values of Kuhlmann et al. and Maazallahi et al., and once again BLENDED and Surface inversions are consistent in this area.

These comparisons are limited by the following arguments:

- Extrapolating the comparison to different years may introduce errors.

- The area is not clearly defined for Romania, and the studied areas contain only a few 0.5°x0.5° pixel, making local estimations very sensitive to high pixel increments. This is why we suggest to focus on comparisons at the country scale, rather than the local scale.
- Our CH4 emissions contain all the anthropogenic and natural emissions. Even if the emissions are dominated by coal mining in Poland, and oil/gas production in Romania, comparing totals is theoretically not correct.

For these reasons, and also as we already have included comparison with surface measurements, we have chosen not to add these comparisons in the manuscript. The manuscript is already long, and we want to keep the focus on the relative differences between emissions from the different TROPOMI products, to avoid making the paper unclear. We mention the following references for future work: a deeper and dedicated analysis is required to make a proper and relevant comparison and to validate the emission outputs of satellite-based inversions against surface measurements.

References :

Kuhlmann, G., Stavropoulou, F., Schwietzke, S., Zavala-Araiza, D., Thorpe, A., Hueni, A., Emmenegger, L., Calcan, A., Röckmann, T., and Brunner, D.: Evidence of successful methane mitigation in one of Europe's most important oil production region, Atmos. Chem. Phys., 25, 5371–5385, https://doi.org/10.5194/acp-25-5371-2025, 2025.

Maazallahi, H., Stavropoulou, F., Sutanto, S. J., Steiner, M., Brunner, D., Mertens, M., Jöckel, P., Visschedijk, A., Denier van der Gon, H., Dellaert, S., Velandia Salinas, N., Schwietzke, S., Zavala-Araiza, D., Ghemulet, S., Pana, A., Ardelean, M., Corbu, M., Calcan, A., Conley, S. A., Smith, M. L., and Röckmann, T.: Airborne in situ quantification of methane emissions from oil and gas production in Romania, Atmos. Chem. Phys., 25, 1497–1511, https://doi.org/10.5194/acp-25-1497-2025, 2025.

Tu, Q., Schneider, M., Hase, F., Khosrawi, F., Ertl, B., Necki, J., Dubravica, D., Diekmann, C. J., Blumenstock, T., and Fang, D.: Quantifying CH4 emissions in hard coal mines from TROPOMI and IASI observations using the wind-assigned anomaly method, Atmos. Chem. Phys., 22, 9747–9765, https://doi.org/10.5194/acp-22-9747-2022, 2022.

6. My largest worry/concern is that the inverse optimized emissions compare worse against surface observations than the non-optimized emissions. This point needs to be explored more deeply.Lines 608-611 and associated tables/figures/etc.

As presented in Table 6 and Figure 13, the concentrations simulated with the posterior emissions are not clearly improved by the inversion: they are slightly improved for BLENDED (lower bias and RMSE, fit between observed and simulated concentrations improved for 53% of the stations), but degraded for WFMD and SRON. We have pinpointed the gap between the satellite and surface “points of view”: optimizing emissions with respect to satellite observations does not imply a better fit for surface concentrations. This is an important issue for the comparability of satellite-based and surface-based studies. Comments have been added in the text and in the conclusion. A deeper analysis is indeed required to improve the consistency of these categories of inversions, but we feel it goes beyond the scope of our work, which focuses on satellite XCH4 datasets.

7. Figures 11a and 10c also point out issues which occur when analyzing emissions at higher temporal frequency. I would like to see how these vary at weekly or day-to-day comparison.

The two figures below are similar to 10c and 11a, but at a weekly frequency. The control vector is optimized at a weekly resolution (in both the inversions and OSSEs) and a priori emissions have a monthly resolution, thus the day-to-day variations do not provide additional information.

Fig 10c: the weekly and monthly curves have similar shapes, with more noise especially for the *WFMD* and *Surface* inversions. The discussion of the comparison of temporal variations between the four inversions remains similar.

Fig 11a: the weekly variations show a lot more noise than the monthly ones. We are not so interested in the absolute variations, but rather in the relative differences with one scenario or another. From this point of view, the relative « positions » of the curves are overall similar and lead to the same analyses as presented in the manuscript. A sentence has been added in Section 3.4.1 to mention the fact that monthly variations smooth higher frequency variations, but relative differences are overall conserved. For the sake of readability, we prefer to keep the monthly variations in the manuscript.

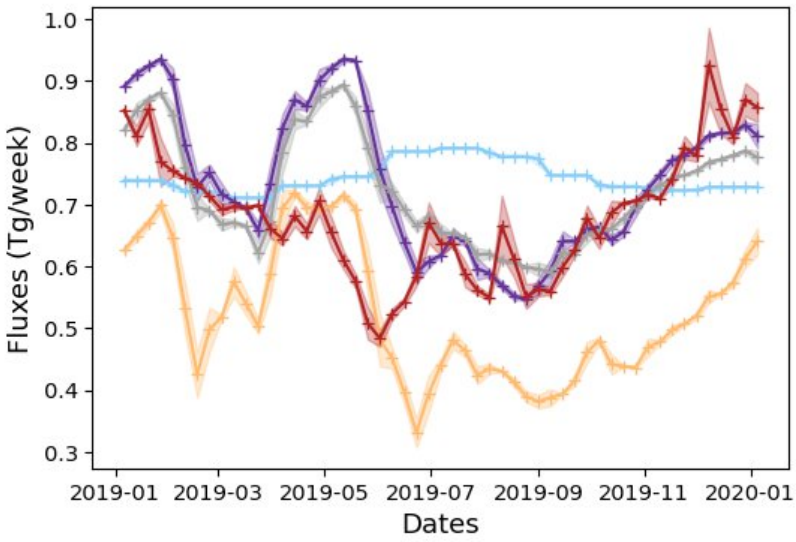

Figure – Similar to Figure 10 (c), but at a weekly temporal resolution.

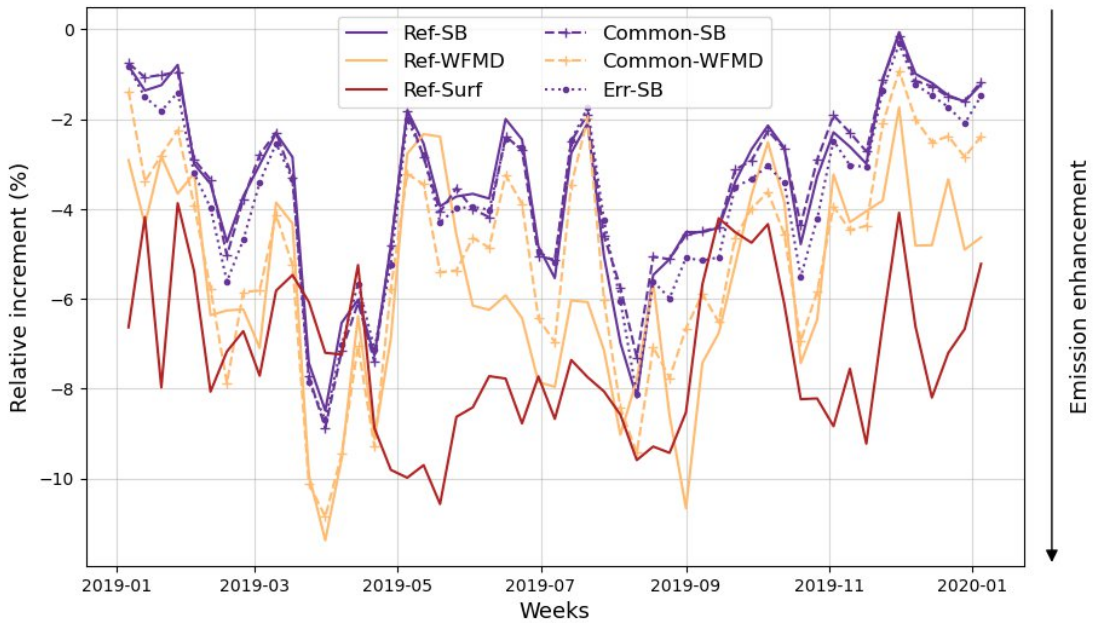

Figure – Similar to Figure 11 (a), but at a weekly temporal resolution.

8. I am concerned that your CH4 retrieval uncertainty is too low, based on your own statement on lines 143-146 and on lines 199-200. There have been multiple recent papers pointing out that there is a substantial impact of aerosols on the TOA and upward radiances in this region based on studies using AERONET (Tiwari et al., 2023), MISR (Liu et al., 2024), and TROPOMI and AEROENT (Tiwari et al., 2025). Furthermore, there are studies which also demonstrate that there are substantial impacts of aerosols on the ATM and hence absorption radiances in this region based on using sunphotometer and aerosol microphysical observations in tandem (Guan et al., 2025; Yang et al., 2025). A larger uncertainty on the column retrieval will lead to a change in the emissions by changing the spatial and temporal variation of emissions (Lu et al., 2025), as well as call into question both very high and very low emissions values (Zheng et al., 2025).

The purpose of our study is to compare emissions from inversions assimilating TROPOMI datasets as close as « a data user would do ». Therefore, we do not try to improve the retrieval uncertainty: they are only modified in the OSSE « SB-Err », to evaluate the impact of the inconsistencies between errors of SRON/BLENDED and WFMD.
Also, increasing the uncertainty would only make the posterior closer to the prior. It would not address the biases of the observation datasets and their impact on the inversion results, which we explore in this study. For these reasons, we do not modify the XCH4 uncertainties.

However, the papers you mention are very important in terms of the sources of differences between products and the analysis of optimized posterior emissions. The substantial impact of aerosols is consistent with the analysis of Section 3.1. A discussion on the retrieval uncertainties and the inferred emissions in Section 2.1.3.

9. Your simple comment about the change in the OH dominating the changes in the CH4 on lines 181-182 requires a deeper set of explanations. New work demonstrates that increases in coal mining (and hence CH4 emissions) are paired with increases in CO and BC emissions (from steel and iron production, as well as biomass burning)(Frausto-Vicencio et al., 2023; Mallet et al., 2024), and hence these other co-emitted species will also impact the lifetime of CH4 (by competing with OH radicals) during this period of time (Hu et al., 2024; Li et al., 2025; Wang et al., 2025; Deng et al., 2024; Pitt et al., 2019).

This sentence had been written to explain the XCH4 time variations of Fig. 2a, which are related to the hemispheric seasonal cycle of CH4 in the atmosphere. We agree that the work you mention shows that the competition between chemical species is more complex than presented here.

Integrating a discussion would go out of scope of the study, because chemistry is neglected in the transport model due to the small ventilation time scale of the domain (~10 days). Therefore, we remove this oversimplified comment from the text.