

Response to Reviewer Comments

We thank the two reviewers for their thoughtful comments. Our responses are in blue. Line numbers are based on the revised manuscript.

Response to Reviewer #1

This study presents a multi-year top-down assessment of methane (CH₄) emissions over Europe using in-situ and satellite data. While there are several studies looking on European CH₄ emissions this study combines in-situ and satellite data and extends the studying period to 2023. The manuscript is well organised and overall scientific sound. However there are several key concerns that need to be revised/clarified. The manuscript lacks discussion in several statements the authors make. I first include some general comments, followed by comments about the parts of the manuscript that require the most attention/revision and later on specific lines, for minor adjustments/corrections/suggestions.

Some general comments:

Please use “Tg y⁻¹” as in your abstract and not “Tg a⁻¹”.

Also for the units in your map-figures use mg m⁻² d⁻¹ or mg m⁻² h⁻¹. That way its easier to compare your results with other studies.

Use the term “flux”/”fluxes” when you describe/show results with units “Mg Km⁻² a⁻¹”.

We thank the reviewer for the suggestions. Following the reviewer's recommendation, we now use Tg yr⁻¹ instead of Tg a⁻¹ throughout the manuscript and supplementary material. We also use "fluxes" when referring to spatially distributed quantities expressed in Mg km⁻² yr⁻¹.

Regarding the units used in the spatial maps, we retain Mg CH₄ km⁻² yr⁻¹. Our inversion optimizes annual methane emissions at the level of grid cell clusters (typical size 50-200 km). Using Mg CH₄ km⁻² yr⁻¹ helps avoid the potential misimpression that the inversion resolves finer spatial or temporal details, an implication that units such as mg m⁻² h⁻¹ might hint. Nonetheless, our unit can be readily converted to mg m⁻² h⁻¹ or mg m⁻² d⁻¹ if needed. This unit, or similar variants, has been adopted in previous global-scale studies (e.g., Maasakkers et al., 2019).

Maasakkers, et al.: Global distribution of methane emissions, emission trends, and OH concentrations and trends inferred from an inversion of GOSAT satellite data for 2010–2015, Atmos. Chem. Phys., 19, 7859–7881, <https://doi.org/10.5194/acp-19-7859-2019>, 2019.

Major comments

Data session

Section 2.1:

Could you elaborate more on which TROPOMI product you used (SRON?, BREMEN?) and how are you dealing with the over-passing orbits

We thank the reviewer for this comment. We have clarified the satellite products used in the revised manuscript. Specifically, we used the blended TROPOMI–GOSAT product developed by Balasus et al. (2023), which is based on the SRON TROPOMI product (Lorente et al., 2023) but apply a machine learning algorithm to correct known retrieval biases of the TROPOMI data, using CO₂ proxy GOSAT methane retrievals as a reference (Parker et al., 2020).

Regarding overlapping satellite orbits, all valid observations from different overpasses on the same day are retained, including overlapping pixels. For each individual satellite sounding, the model-equivalent XCH₄ is calculated by applying the corresponding averaging kernel and prior profile. The model-

equivalent XCH_4 values are then aggregated onto the model grid cells and averaged to produce daily gridded observations. Consequently, overlapping satellite footprints are naturally merged into a single grid-cell observation at the model resolution.

We now clarify in Section 2.1.

“We use column-averaged dry-air methane mole fractions (XCH_4) from the blended GOSAT+TROPOMI satellite product (referred to as TROPOMI observations hereafter) developed by Balasus et al. (2023), which employs CO_2 proxy GOSAT methane retrievals as a reference (University of Leicester GOSAT Proxy XCH_4 dataset v9.0) (Parker et al., 2020) to correct the retrieval biases of original TROPOMI data produced by Space Research Organisation Netherlands (Lorente et al., 2023).”

“We feed daily grid-cell averaged XCH_4 observations, along with corresponding model values that account for each retrieval's prior vertical profile and averaging kernels (see Section 2.4), into the inversion.”

Section 2.2

The fact that you compute daily daytime averages using data between 10 and 16 is technically correct. However this is valid only for the surface stations (see for example in Steiner et al. 2024, Ioannidis et al. 2026).

Figure 2 shows stations that are not surface stations, e.g. PUY, OXK. These are mountainous stations which mean only nighttime values should be assimilated because during night these stations are more representative of free-tropospheric conditions and are not influenced by pollution from daytime upslope winds which usually are not well represented in coarse resolution modelling.

If you want to assimilate daytime observations from mountainous sites then you need to make sure you are using the correct sampling height (e.g. in the middle between the model topography and the real altitude). If you did that already please mention it in the paper.

Otherwise, is there a scientific, other, reason you decided to keep daytime data from mountainous stations? If not, I recommend to either remove them completely from your assimilation or use nighttime observations from these sites.

We thank the reviewer for this valuable comment regarding the representativeness of mountainous sites. In the original inversion, we applied a surface-pressure threshold of 800 hPa and excluded observations with surface pressures below 800 hPa before calculating daytime averages. However, this pressure-based filtering may not be accurate for excluding mountain stations. Following the reviewer's suggestion, we therefore refine the observation selection by directly excluding seven mountain stations, including those highlighted by the reviewer. The 10:00–16:00 local-time averaging is now applied only to the remaining non-mountain stations. The inversion was rerun using this revised observation set, and all results and figures have been updated accordingly. Detailed information on the station selection is provided in Table S3, and Section 2.2 has been revised accordingly.

Section 2.1 and Section 2.2

You don't mention the model-data mismatch you used for the in-situ and tropomi data.

We thank the reviewer for this comment. We now report the model–data differences for both the ICOS surface observations and the blended TROPOMI observations in Section 2.6. We evaluate the mismatch using the root-mean-square error (RMSE) between the observations and the simulations. For ICOS observations, the RMSE decreases from 43.27 ppb for the prior simulation to 35.32 ppb for the posterior simulation. For blended TROPOMI observations, the RMSE decreases from 13.20 ppb to 12.36 ppb (Figure S6). These results show that the posterior simulation improves the agreement with both observational datasets.

We now also report the statistics for ICOS and TROPOMI observations to specify the observing system error covariance matrix (So) in Section 2.5, as well as Figure S3, S4 and Table S4. The error variance of individual observations is on average ~ 12 ppb for satellite and ~ 40 ppb for surface data. The spatial

correlation length is 500 km for satellite and temporal correlation length is 2 days for both satellite and surface.

Section 2.3

Are you using CH₄ freshwater a-priori emissions? Emissions from rivers/lakes but also coastal wetland are important. Are they included somewhere as an input? Discuss in the manuscript.

We thank the reviewer for this valuable comment. Freshwater methane emissions, including emissions from rivers, lakes, and coastal wetlands, are not explicitly included as separate source categories in our prior emission inventory. This is now clarified in the description of prior emissions (Section 2.3).

To have a first-order assessment of their potential influence, we examined published estimates for these sources. Available studies suggest river emissions of approximately 0.8 Tg CH₄ yr⁻¹ over Europe (Rocher-Ros et al., 2023; <https://www.nature.com/articles/s41586-023-06344-6>), lake emissions of approximately 0.26 Tg CH₄ yr⁻¹ (http://geoschemdata.wustl.edu/ExtData/HEMCO/CH4/v2022-11/Lakes/Maasakkers_Lakes_2015.01x01.nc), and coastal wetland emissions of approximately 0.39 Tg CH₄ yr⁻¹ estimated from Weber et al., (2019) (<https://www.nature.com/articles/s41467-019-12541-7>). Collectively, these sources are relatively small compared with total European methane emissions and are therefore expected to have a limited influence on the national-scale emission estimates. Nevertheless, uncertainties in freshwater methane emissions may contribute to regional biases and are discussed as a limitation of this study. We have added a discussion of this limitation in the revised manuscript (lines 337-344, Section 5.1).

Table S1: There is no way readers would go through your excel table and check the numbers. Add a nice barplot in the supplementary material showing prior/posterior emissions per year and per country in your domain.

We thank the reviewer for this suggestion. We agree that graphical summaries are useful for highlighting the main results. In this study, however, the main countries and sectoral emission changes are already presented and discussed in the main text (Figures 6–10). We prefer to retain Table S1 in the Supplementary Material because it provides the complete record of the inversion results, which may be useful for some readers.

Section 5

Please rephrase the title. ‘Correction’ is misleading. This term indicates that the a priori emissions you used are wrong, while both bottom-up and top-down emissions have uncertainties.

We have revised the title accordingly to avoid the confusion. The title for Section 5 now reads “2019–2023 mean methane emissions”.

This session also lacks a bit of discussion about figure 8. Discuss about the spatial distribution, patterns in the a-priori, the a-posteriori. It is also important to know whether posterior emissions show similar spatial patterns as the prior. Papers usually also show a map of the a-priori emissions.

Following the reviewer's suggestion, we have added a description and discussion of Figure 8 in Section 5. This new text highlights the broad agreement in spatial distribution between the prior and posterior emissions, as well as the regions where the inversion introduces notable adjustments. These findings are then examined in greater detail in Subsections 5.1 and 5.2. We now add additional references to Figure 8 when it is discussed in the two subsections. In addition, we have updated Figure 3 to include the spatial distribution of total prior emissions alongside the sector-specific prior emissions already shown.

The new text reads “Overall, the posterior estimate yields a spatial distribution of methane emissions across Europe that is broadly consistent with prior knowledge (Figure 8). For instance, the prior and

posterior show that methane emissions concentrate in central and western Europe, reflecting intensive agriculture, waste treatment, and fossil fuel production (Figure 3). Meanwhile, the inversion reveals substantial regional adjustments to the prior estimates. Notable upward revisions occur in regions with intensive agriculture and waste sources, such as the Netherlands, northwestern Germany, and northern Italy, as well as in urban centers across France, Spain, and the southern United Kingdom. In contrast, downward adjustments are found in offshore oil production regions in the North Sea and in boreal wetland areas (Figure 8). In the following sections, we discuss these inversion-informed revisions to the 2019–2023 average methane emissions for each of these regions”.

I also miss some discussion on geological and wetlands CH_4 emissions since there are important over Europe and their prior (e.g. for geological) have large uncertainties.

We thank the reviewer for this insightful suggestion. Indeed, these important sources, and their impact on the estimation of anthropogenic sources, are barely discussed in the previous version.

We now add a paragraph in Section 5.1 (lines 337–344) to discuss the limitation of our method due to uncertainties in wetlands and geological emissions: “Despite the above evidence, our sectoral attribution is inherently limited in regions with spatial overlap among major sectors or poor prior representation of key sources. Coastal wetland and freshwater emissions, absent from our prior, may partly explain the inferred increase over the Netherlands (Buzacott et al., 2024). Geological emissions introduce major uncertainty in Italy. Etiope et al. (2019) estimated $\sim 2 \text{ Tg yr}^{-1}$ from geological seeps, compared to negligible amounts in our prior, mirroring large discrepancy in global geological emissions using different approaches (Hmiel et al., 2020; Etiope et al., 2019). This leads to considerable uncertainties in both total emission estimation and sectoral attribution, particularly where agricultural and geological sources are spatially closely located like northern Italy. Given their important implications for distinguishing natural from anthropogenic emissions, these limitations warrant further targeted investigation.”

We also briefly discuss the results regarding wetland emissions from northern Europe in Section 5.2 (lines 373–378). Wetland emissions are major emission sources in Northern Europe. The inversion indicates a general downward adjustment of wetland emissions in this region (Figure S11). However, observational constraints are overall weak because of sparse observation coverage of both satellite and surface observations, limiting our ability to robustly assess wetland emissions in this region.

Section 5.1

You talk about agriculture and waste emissions and then in line 265 you say something about urban emissions which first does not fit with the sentence before and also contradict what you say later on about urban areas in lines 279 to 284 (specifically the lines 282 to 284). This part confused me a lot; the way to wrote it and what your take away message want to be. Please revise carefully the part about urban emissions.

We thank the reviewer for pointing out the unclear language here. To avoid confusion, we have restructured this part of the discussion into two separate subsections. The analysis for France, Spain, and the United Kingdom now appears in a new subsection, Section 5.3, titled "Urban Hotspots." And the title of Section 5.1 is now revised to “Agriculture dominated regions”.

I miss a discussion and plots on the seasonal cycle of CH_4 emissions over Europe (as a whole or for different European countries or for specific sub-regions etc). Previous papers, like Bergamaschi et al. 2018 (<https://doi.org/10.5194/acp-18-901-2018>), East et al. 2024 (<https://doi.org/10.1029/2024GL108494>), Ioannidis et al. 2026, discuss the seasonal cycle of CH_4 emissions. It would be really interesting to compare the seasonal cycle between the a-priori and the top-down emissions from the different inversion simulations, e.g. how do in-situ only or satellite only

assimilation drive the seasonal cycle compared to in-situ + satellite assimilation. Your paper would be more complete if you add a session on seasonal cycle and discuss your results also with respect to previous studies.

We agree that the seasonal cycle can provide useful information on methane emissions. However, our inversion optimizes annual emissions and is therefore not designed to provide observation-informed results for seasonal emissions (see Section 2.5). We now have clarified this limitation in the Methods. We also perform a brief evaluation of seasonal biases in simulated methane concentrations (newly added Figure S5; Section 2.6), which shows that both the prior and posterior simulations reproduce the main observed seasonal variability in XCH₄. Furthermore, we suggest that our future study should make use of the information embedded in seasonality (Section 5.1).

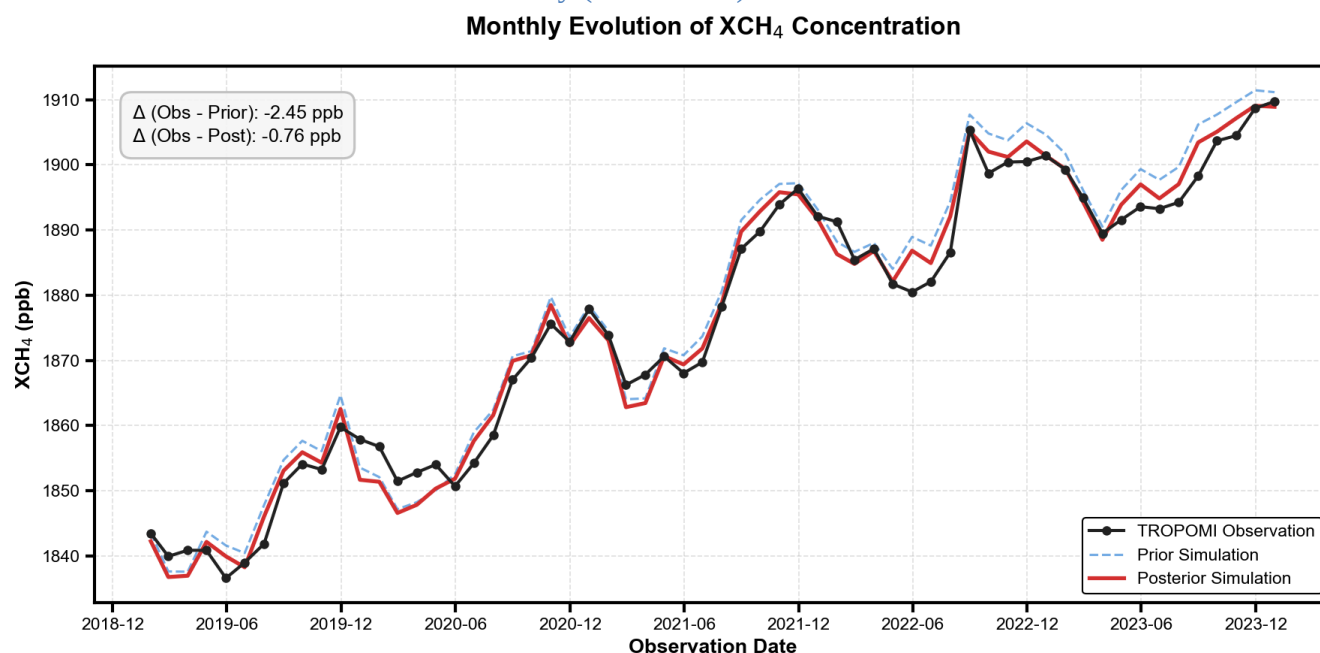


Figure 1. Monthly evolution of XCH₄.

Minor/More specific comments:

Line 65: add “and whole Europe “ after “ in the Benelux countries” and also Ioannidis et al. 2026 in the references there after Steiner et al. 2024

Accepted.

Line 70: say which tool (transport model, inversion technique) you are using for the inversion analysis

Accepted.

Lines 86-90: It would be useful to know the number of observations across the study domain for all years, not only 2019. You can easily add for example a barplot in the supplement material showing the number of observations per month and per year.

Accepted, see in Figure S1.

Line 97: delete flagged as – its written twice

Accepted.

Session 2.2: It would be useful to add a table in the supplement material showing if the station is a mountain site, the elevation, inlet height etc similarly to Steiner et al 2024 or Ioannidis et al. 2026.

Accepted, see in Table S3.

Line 101: You say in the text that Fig.2 shows the average CH₄ concentrations for 2023. The caption in Fig.2 says 2019-2023 average. Which one is the truth? In any case show 2019 similarly to TROPOMI figure.

We thank the reviewer for identifying this inconsistency. The text and figure have been revised to consistently show the 2019–2023 average methane concentrations.

Line 106: How can you really make that statement when you plot different years?

Following the reviewer's suggestion, we have revised the figures and text to consistently use the 2019–2023 average observations, ensuring that the comparison between the satellite and surface datasets is based on the same time period.

Line 115: Provide a table that summarises all the input a-priori emissions, their original spatial resolution and their temporal resolution.

Accepted, see in Table S2.

Line 115: How do the geological emissions from Maasakkers et al. 2019 compare to Etiope et al. 2019 especially over Italy?

We thank the reviewer for this comment. We have added a discussion comparing the geological seepage inventory of Maasakkers et al. (2019), which is used as the prior in this study, with the estimates of Etiope et al. (2019). In particular, we discuss the substantial uncertainty in geological methane emissions over Italy, where published estimates differ by nearly two orders of magnitude (lines 337–344).

Line 131: Interesting that GEOS-chem includes stratospheric loss fields. Are there tools provided by the GEOS-chem community or output parameters from your model that you could use to discuss the importance (or not) of those loss fields on CH₄ emissions in Europe? Especially for OH?

We thank the reviewer for this suggestion. In this study, GEOS-Chem uses archived monthly OH, Cl, and stratospheric loss fields, and these chemical loss fields are not optimized in the inversion. Because methane has a relatively long atmospheric lifetime, regional methane concentrations over Europe are influenced primarily by surface emissions and lateral boundary conditions rather than by regional variations in OH. Furthermore, our boundary conditions are constrained using atmospheric observations, which partly accounts for the large-scale influence of OH on background methane concentrations. We therefore do not expect uncertainties in the prescribed OH fields to substantially affect the inferred European methane emissions.

Line 159: “with a 50% relative uncertainty assigned to each emission flux”. Each input flux? Or are you grouping the input fluxes into categories (e.g. anthropogenic, natural) and you optimise (assign uncertainties) to each category? Are your anthropogenic emissions only EDGAR emissions or there are the sum with biomass burning? Your text is vague; clarify and please explain how and which input emission fluxes you are optimising and their uncertainty. Also move here from the supplement material the text about the spatial and temporal correlation scale. Also it’s missing information on the window length you are using.

We thank the reviewer for pointing out that the original description was unclear. We have revised the Methods section to clarify the definition of the optimized state vector and its prior uncertainty. The prior emissions consist of anthropogenic emissions from EDGAR together with natural emissions from wetlands, biomass burning, termites, and geological seepage. The inversion optimizes the total methane emissions of each spatial cluster rather than individual source sectors. A relative uncertainty of 50% is assigned to each optimized state vector element (i.e., each emission cluster), rather than to individual source categories. We have also expanded the description in the main text and added Table S4 to summarize the relevant parameters, while retaining the detailed implementation in the Supplementary Material. The inversion is performed separately for each calendar year using a one-year inversion window.

Line 197: What about TCCON data? There are observations in a few sites in your domain for your simulation years. Is there a reason you did not include them in your validation?

We thank the reviewer for this valuable suggestion. Following the reviewer's recommendation, we evaluated the inversion results using independent TCCON observations. The comparison is presented in the new Figure S9, with the TCCON site information summarized in Table S3. Among the seven

TCCON sites within the study domain, the posterior simulation reduces the root-mean-square error (RMSE) at five sites relative to the prior simulation. We have added this evaluation and discussion to the revised manuscript (lines 233-244).

Lines 214 to 218: Why do satellite data offer little information? Limited coverage? Someone would expect that satellite data would better constrain the emissions than 31 points in a big domain. Elaborate a bit more in your text.

We thank the reviewer for this important question. Although satellite observations provide far more individual retrievals than the surface network, neighboring satellite pixels and adjacent orbital tracks are strongly correlated in space, reducing the amount of independent information available to constrain the inversion. In contrast, surface observations are continuously measured in time and are generally more sensitive to boundary layer methane concentrations that are directly influenced by local surface emissions. Consequently, the relatively dense ICOS network over central and western Europe provides stronger constraints on regional emissions than the satellite observations alone.

Line 225: For Europe here you mean your whole domain as shown in your figures?

Here, "Europe" refers to the European countries included in our analysis rather than the entire inversion domain shown in the figures. The inversion domain also includes surrounding regions (e.g., parts of Russia and neighboring areas) to properly represent atmospheric transport. We have clarified this definition in the revised manuscript.

Line 248: replace “ the inversion’s corrections relative to prior emissions” with “the increments, calculated as the difference between the posterior and the prior fluxes”.

We have revised the text accordingly and now describe the inversion results as the differences between the posterior and prior emissions, rather than as "corrections" to the prior emissions.

Line 248-249: you say top 10 methane emitting countries in Europe. Based on what? Elaborate or add a reference.

We have clarified that the countries are ranked according to their posterior anthropogenic methane emissions. The text has been revised accordingly.

Line 259: replace corrections with adjustments.

Accepted.

Line 261: “.. in these countries ..” which countries? As shown in Fig. 9? Be more specific.

We have revised the text to explicitly refer to the Netherlands, Germany, and Italy, avoiding the ambiguous phrase "these countries".

Line 269: “... this suggests that systematic biases may persist in national level livestock emission...” well no because your sentence before is about urban emissions measurements. Also the Netherlands is a really specific example regarding the livestock emissions. Add a more general discussion for the whole Europe and refer to previous studies.

We thank the reviewer for this important comment. We agree that the Dutch farm study alone cannot support a general conclusion for Europe and that the relationship between the urban studies and livestock emissions was not sufficiently clear. We have therefore reorganized the discussion in Section 5.1 (lines 325–335) to distinguish the evidence related to agricultural and urban emissions and to provide a broader basis for interpretation. We now first refer to a recent satellite inversion over western and central Europe that inferred higher livestock emissions relative to the prior inventory (Oak et al., 2026), while retaining the Dutch farm study as local-scale supporting evidence and explicitly noting its substantial uncertainty and limited representativeness at the national scale. The urban measurements are discussed separately and are used only to indicate that urban sources alone are unlikely to explain the

inferred increases in the Netherlands, Germany, and Italy. Based on this combined evidence, we now state that these results point to potential systematic biases in national-level livestock emission accounting. We further discuss the limitations of this sectoral attribution in the following paragraph.

Lines 271, 279, 361: replace corrections with adjustments
Accepted.

Line 272: interesting results for UK because of the long range flow influence in the UK and especially when using in-situ observations (which are only 2 in the UK as shown in your map). Do you get these results over this part of the domain because of the correction you applied in your nested domain? Can you add a bit more discussion about your findings and not just describe them?

We thank the reviewer for this insightful comment. We have expanded the discussion in the revised manuscript (lines 381-388). The satellite-only inversion does not show a comparable increase over southern United Kingdom, indicating that the inferred adjustment is primarily constrained by the local surface observations rather than by the satellite data. These surface observations are more sensitive to regional boundary-layer methane enhancements and therefore mainly reflect local emission signals. In contrast, offshore emissions around the United Kingdom are constrained only indirectly through atmospheric transport because of the lack of nearby observations, resulting in substantially larger uncertainty. We have clarified this limitation in the revised manuscript.

Line 311: Do you mean wetland emissions by the term natural?
We intended to refer specifically to wetland emissions and have revised the text accordingly.

Line 354: Add a reference at the end of this sentence "... and a recent surface-data inversion".
Accepted.

Line 370: Even when using only satellite data?

We thank the reviewer for this question. The satellite-only inversion produces somewhat larger posterior–prior differences over Romania and Ukraine than the surface-only inversion, indicating that satellite observations provide the dominant constraint in these regions. However, the overall changes remain modest, suggesting that the effective sensitivity of the available observations to national-scale emissions is still limited.

Lines 371 to 376: give some numbers for the trends
Accepted. We have added quantitative trend values for the main countries discussed in the text to complement the qualitative description of the spatial patterns.

Comments for the figures:

Figure 1: I do not understand what you mean when you say

"Observational data constraints for the inversion are shown below the black dashed line" I could not link the above sentence with anything on your main text; please elaborate.

We thank the reviewer for pointing this out. The black dashed line indicates the northern limit (55°N) of the satellite observations used in the inversion rather than the extent of all observational constraints. Satellite observations poleward of 55°N are excluded because of snow cover, unfavorable illumination, and increased model uncertainty associated with stratospheric methane. We have revised the figure caption to clarify this point.

Figure 2: Correct caption or be consistent with what is written in the main text. See comment earlier.
Accepted.

Figure 3. OK but move it to the supplement material. Also what is OtherAnth? First time I see this term in

this plot, not defined in the text.

We thank the reviewer for this suggestion. We prefer to retain Figure 3 in the main text because it provides the spatial distribution of the prior sectoral emissions, which serves as essential context for interpreting the posterior emission differences and the subsequent discussion of sectoral attribution. To improve clarity, we have revised the figure caption and defined "OtherAnth" as the remaining anthropogenic EDGAR sectors, including energy, industry, transport, residential combustion, chemical production, and iron and steel production.

Figure 5: caption – this is the average of the 5 years right? You say annual inversion in the caption. We thank the reviewer for pointing this out. Figure 5 shows the five-year mean averaging-kernel sensitivities derived from the annual inversions for 2019–2023, rather than the result from a single annual inversion. We have revised the caption to clarify this.

Figure 7: Your caption does not agree with the title of Fig. 7a. Revise.
Accepted.

Figure 8: Title Fig.8a also TROPOMI + ICOS? Add to be consistent with Fig.8b
Accepted.

For Fig.8b: Can you change the colour of the colourbar and use the gradients of one colour for the negative values (usually blue) and the gradients of another colour for the positive values (usually red), with 0 being white? Otherwise its difficult to see the differences in your map.

Accepted. We have revised Figure 8b using a diverging blue–white–red color scale centered at zero. The same color scheme has also been applied to other figures displaying positive and negative differences for consistency.

Figure 9: Are the green diamonds and purple circles data re-gridded to your domain? Because these studies are in a different resolution than yours so their country-totals would be different than yours. We thank the reviewer for this important comment. No regridding was performed because the published studies do not provide gridded emission fields. Instead, Figure 9 compares the country-total emissions reported in each study.

Figure 9 and 10: Are you showing the sum or the average of CH₄ emissions for 2019 to 2023? Clarify in the caption.

We thank the reviewer for pointing this out. Figures 9 and 10 show the methane emissions for 2023 rather than the mean over 2019–2023. Although the panel titles indicate the year, we agree that this was not sufficiently clear from the figure captions. We have revised the captions accordingly.

Figure S5: Why don't you compare your results against TCCON observations?
Accepted.

Figure S6: When you say independent ICOS observations you mean the 31 stations you used in the ICOS-only and ICOS-TROPOMI inversion right? Remove independent its confusing. We thank the reviewer for pointing this out. In the revised Supplementary Material, this cross-validation analysis is now shown in Figure S7. In this case, the ICOS observations are used only to evaluate the TROPOMI-only inversion and are therefore independent of that particular inversion. We have revised the caption to make this distinction clear.

Figure S8: Both TROPOMI-only and ICOS-only inversion agree on the upward adjustments over Italy, while they disagree on the sign over the Netherlands. Can you elaborate on the text why? This is an interesting finding. Also both agree on the downward adjustment over Moscow where Figure 5a shows

values above 0.6 so there is quite good constrain. However it's still puzzling to me how the constrain is so high over Russia when using only ICOS data while there are not in-situ data over Russia. Elaborate on the text.

We thank the reviewer for highlighting these interesting differences. We have expanded the discussion in the revised manuscript (lines 319-321). The agreement between the TROPOMI-only and ICOS-only inversions over Italy suggests that the inferred increase is robust to the type of observation assimilated. Over the Netherlands, however, the two inversions differ in sign. We interpret this difference as reflecting their distinct sensitivities: surface observations are more sensitive to regional boundary-layer enhancements and short-range transport, whereas TROPOMI constrains column methane over a broader spatial scale. Because the Netherlands is small and located close to major emission regions in Germany and Belgium, cross-border transport and spatial attribution may contribute to the different responses of the two inversions.

For Moscow, the relatively high averaging-kernel sensitivity in the ICOS-only inversion does not imply direct local observational coverage. The Russian state-vector elements cover relatively large spatial clusters, which can be constrained indirectly by ICOS observations elsewhere through modeled atmospheric transport. The mapped sensitivity therefore partly represents information on the larger cluster rather than a purely local constraint over Moscow. We have clarified this interpretation and noted that the spatial attribution within such large clusters remains uncertain.

Response to Reviewer Comments

Our responses are in blue. Line numbers are based on the revised manuscript.

Response to Reviewer #2

General

The authors estimate European methane emissions based on surface (ICOS) and satellite (TROPOMI) observations. They obtain interesting new results that fit well in the scope of ACP and should to be published. The paper is well structured and presented. However, a few major issues should be addressed.

- A. The description of the method is very similar to an earlier publication. The authors should make more transparent what is new in their work and what has been published before (see comment 1 below).

We thank the reviewer for this comment. We now add an opening paragraph in Section 2, in which we clarify that our methodology builds on our previous work, and highlight that the incorporation of existing satellite and surface observations is new in assessing European methane emissions. Additionally, we have revised the text in subsections to highlight detailed differences from the previous work.

- B. The description of the method lacks some details that are important for the interpretation of the results (see comments 3–11 below).

We have revised the Methods section to provide the additional methodological details following the suggestion. The specific changes are addressed in the responses to the individual comments below.

- C. The discussion should be extended and address possible weaknesses of the method. Ideally, I would expect the discussion to answer the following questions:

- What are the most relevant approximations made in the method?
- What are other possible weaknesses and sources of uncertainties (if any)?
- How can these approximations and weaknesses affect the results?
- Is the expected impact (of approximations and weaknesses) on the results larger or smaller than the posterior uncertainties?

The latter two questions can be very difficult to answer and should be considered optional. But the authors should at least identify and describe the most crucial approximations made in their work.

These questions can already be addressed in the description of the method (Section 2). But if the authors identify any possible biases or approximations that may affect the results beyond the posterior uncertainties, this should also be mentioned in the discussion of the results. This discussion could then be included in Section 5 (see comment 15 below).

We agree that a more comprehensive discussion of the assumptions and limitations strengthens the manuscript. Accordingly, we have expanded the discussion throughout the revised manuscript, including the uncertainty in wetland and geological emissions, the interpretation of sectoral attribution, and observational constraints. The detailed revisions are described in the responses to the relevant.

Specific comments

Section 2

1. The whole method seems very similar to Zhang et al. (2022) (citations as in the paper). The authors should make transparent what is new in their work and where they closely follow earlier publications. If the same method has been published before for different observation data and a different target region, this must be clearly stated at the beginning of the method section.

We thank the reviewer for this important comment. We now add an opening paragraph in Section 2, in which we clarify that our methodology builds on our previous work, and highlight that the

incorporation of existing satellite and surface observations is new in assessing European methane emissions. This paragraph reads,

“We assess the distribution, trends, and observational constraints of European methane emissions using an analytical Bayesian inversion framework adapted from earlier work (e.g., Maasakkers et al., 2019; Zhang et al., 2022; Chen et al., 2023; Liang et al., 2023; Lu et al., 2023) to the European domain. Satellite and surface observations are jointly assimilated, enabling a comparative assessment of the information content provided by each observation type. The following sections detail the observation datasets, prior emissions, forward model, inversion procedure, and evaluations against independent measurement.”

Section 2.2

2. The authors apparently exclude mountain sites from the observation dataset (JFJ, ZSF, PRS, and CMN). This should be mentioned in the text and I recommend to briefly comment why this selection was made.

We thank the reviewer for this comment. In the original inversion, we applied a surface-pressure threshold of 800 hPa and excluded observations with surface pressures below 800 hPa before calculating daytime averages. However, we agree that this pressure-based filtering alone may not fully address the representativeness of daytime observations at mountain stations. Following another reviewer’s suggestion, we therefore revised the observation selection by directly excluding seven mountain stations. The inversion was rerun using this revised observation set, and the posterior emission estimates and related figures and tables have been updated accordingly. The exclusion of mountain sites is explicitly described in Section 2.2, *“High-altitude mountain stations are excluded from the inversion due to representativeness issues associated with daytime observations at these sites (Ioannidis et al., 2026; Steiner et al., 2024) (Table S3)”*. Table S3 has also been revised accordingly to provide detailed information on the station selection.

Section 2.3

To me, the description of the prior left a few questions open that should be answered in the paper:

3. To my knowledge, EDGAR v8 does not cover the year 2023. Which dataset was used instead for 2023?
We thank the reviewer for pointing this out. EDGAR v8 does not provide emissions for 2023. We therefore used the 2022 EDGAR emissions as the anthropogenic prior for the 2023 inversion, assuming that the spatial distribution and magnitude of anthropogenic emissions remained unchanged between the two years. This information has now been explicitly stated in Section 2.3 and summarized in Table S2.
4. What is the temporal resolution of the prior emissions? If the authors used yearly averaged values for emissions with strong time dependence (such as wetlands or biomass burning), then this should be mentioned as a possible limitation of the method. If they use monthly or higher resolved values, they should mention the seasonal cycle.
We have added Table S2 to summarize the source, reference year, and temporal resolution of each prior emission component. Wetland emissions were prescribed at a monthly temporal resolution, whereas biomass-burning emissions were prescribed at a daily resolution. We have clarified this treatment in Section 2.3.
5. The authors refer to Maasakkers et al. (2019) for geological emissions. Does this mean that prior geological emissions were constructed using the same procedure as described by Maasakkers et al.?

In this case the authors might consider citing also the original data sources. Or is the posterior from Maasakkers

et al. used? In any case, the formulation should be adjusted for clarity.

We thank the reviewer for pointing this out. The geological methane emissions were compiled by Maasakkers et al. (2019) as the prior inventory of their inversion. We now have revised the manuscript to cite original sources.

6. Figure 3 shows the prior emissions, but it seems like geological and biomass burning emissions are missing. Please make sure that all prior emissions are provided in Figure 3 (and in Table S1, see below).

The original version of Figure 3 displayed only the major prior emission categories and did not separately show geological and biomass-burning emissions. We have revised Figure 3 to include additional panels for these two source categories. Table S1 has also been updated.

Section 2.4

7. The procedure for obtaining initial and boundary conditions remains unclear to me:

- 7.1. What is the setup for the global simulation? Are the global and regional simulation online coupled or are these separate simulations? What are the initial conditions and the spin-up time of the global simulation?

We thank the reviewer for this comment. The global and regional simulations were conducted separately and were not online coupled. For the present study, we performed a separate global methane simulation to provide the initial and lateral boundary conditions for the regional simulations. We have made clarifications and cite the paper reporting the results for the global simulation in Section 2.4.

- 7.2. The bias correction (mentioned in line 135) remains unclear to me. Which observations were selected? How was the bias correction computed? At which scales (temporal, horizontal and vertical) does the bias correction vary?

We thank the reviewer for noting that the description of the boundary-condition bias correction was insufficient. The correction was computed separately to the western and northern boundaries, by comparing simulations with satellite or surface observation in ocean or background grid cells near the boundaries. Satellite observations over ocean are mainly used for the western boundary, while high-latitude surface stations located close to the northern edge of the domain are mainly for the northern boundary. We find that these biases do not show apparent long-term trends and seasonal patterns, and therefore the correction was applied as a constant offset for each boundary that does not vary temporally. Also, the correction is applied to the entire tropospheric profile at the boundary.

We have now clarified the bias correction procedure in Section 2.4 of the Methods section: “We then quantify and correct residual systematic regional biases in the boundary condition, using satellite and surface observations from background grid cells near the western and northern boundaries, which are the primary inflow pathways into the European domain (Figure S2). The derived correction, 12 ppb for the western boundary and 30 ppb for the northern boundary (Figure S2), is applied uniformly to the entire tropospheric vertical profile at both boundaries.”

We further evaluated the resulting simulations against independent IAGOS-CARIBIC aircraft profiles, which show improved agreement between simulations and observations.

- 7.3. An explanation of the bias correction could make use of Figure S4. Does the bias correction also use satellite observations above the ocean as shown in Figure S4? This point should be clarified in the caption of Figure S4 or in the explanation of the bias correction.

Yes, the TROPOMI observations over the ocean shown in Figure S4 were used to derive the correction applied to the western boundary conditions. We have now clarified this point in the

caption of Figure S4 (now Figure S2).

8. I did not see a citation of the IAGOS-CARIBIC dataset. Citing one version of the dataset could help understanding and reproducing the results.

The IAGOS-CARIBIC data source was previously given only in the caption of Figure 4. We have now added the corresponding data access information to the main text where the dataset is first introduced (line 158).

Section 2.5

9. The definition of the spatial clusters (state space) is important for the a priori uncertainties and for the interpretation of Figure 5. I therefore suggest to include Figure S1 directly in the paper.

We thank the reviewer for this helpful suggestion. We have now incorporated the spatial cluster map into Figure 5 in the main text.

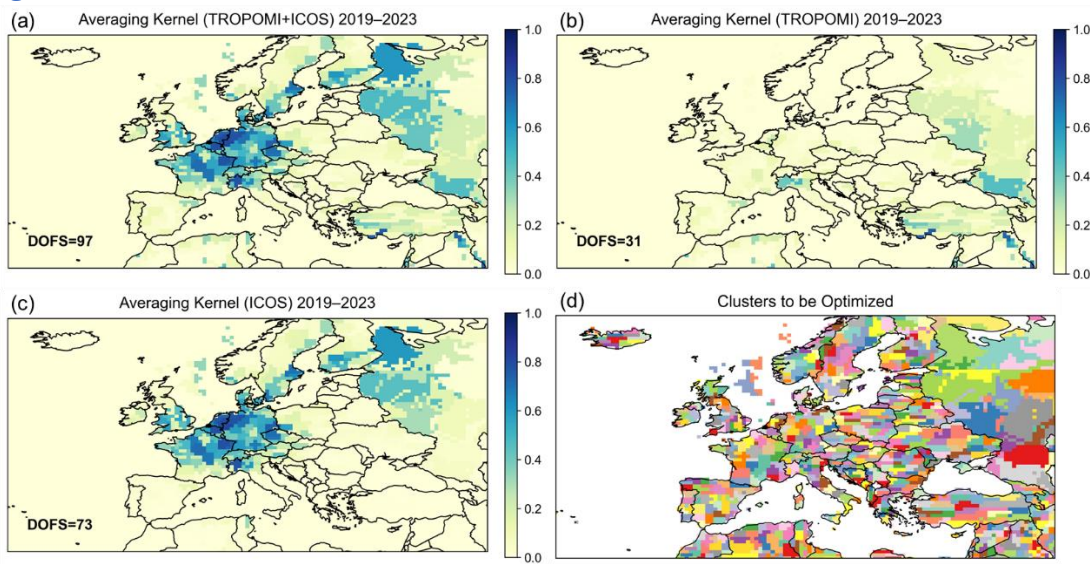


Figure 1. Spatial distribution of averaging kernel sensitivities and spatial clusters.

10. The description of constructing S_o in the supplement appears inconsistent with the main text.

Apparently, S_o in the supplement is equivalent to the full error covariance matrix S' in Zhang et al. (2022) and not the same as S_o in the main text. This should be clarified and the notation should be chosen such that the non-diagonal matrix $S' = \Sigma \mathcal{C} \Sigma$ and its diagonal approximation S_o are clearly distinguished.

We thank the reviewer for identifying the unclear and inconsistent notation. The original description in the main text was imprecise because it referred to S_o simply as a diagonal covariance matrix, without explaining that the observation-error variances were constructed using correlation-dependent weighting, as described in the Supplement. We have therefore revised the main text and now consistently denote the full error covariance matrix as S_o' and its diagonal approximation as S_o . We have also added text to describe the specification of error statistics for S_o' . The notation and description are now consistent throughout the manuscript.

11. The assumed uncertainties in S_o are a key parameter that should be provided and (briefly) discussed. Following the notation of Zhang et al. (2022), these are the standard deviations σ of individual observations (satellite and ground-based) and the regularization parameter γ . These parameters are crucial for understanding the results presented in Section 3. What assumptions were made on error correlation of satellite and ground-based observations? How were model uncertainties taken into account?

We thank the reviewer for highlighting the importance of the observing-system error parameters. We have revised text in Section 2.5 to clarify the specification of the full error covariance S_o' , from which

So is derived. We also add Table S4 to summarize the standard deviations and error correlations assigned to the satellite and surface observations. Specifically, the baseline error standard deviations were first estimated using the residual error method, yielding values of approximately 12 ppb for the blended TROPOMI observations and 40 ppb for the ICOS surface observations. We further estimated the temporal and spatial correlations for observations. The fitted correlation functions and correlation scales are now shown in Figures S3 and S4.

The revised text reads “*The observing system error covariance matrix, which accounts for random instrument and forward model errors, is represented by a diagonal approximation ($\mathbf{S}_0$) of the full matrix ($\mathbf{S}_0'$), which is populated with the error variance of individual observations (on average ~12 ppb for satellite and ~40 ppb for surface) along with spatial and temporal correlations between observations (spatial correlation length of 500 km for satellite and temporal correlation length of 2 days for both satellite and surface) derived from the error residual method (Figure S3, S4 and Table S4). The diagonal approximation ($\mathbf{S}_0$) of $\mathbf{S}_0'$ is solved following the method of Zhang et al. (2022) and Liang et al. (2023) (see details in Text S1).*”

12. Just a suggestion: Since Subsection 2.5 is quite long, the authors could consider splitting it at line 187 to have an own subsection (2.6) for the evaluation of the inversion.
We have revised accordingly.

13. lines 191–195: The authors argue that the sharp vertical concentration gradient observed in the IAGOS data in Germany indicates an upwards adjustment of the emissions. But it seems to me that the analysis shown in Figure S7 is prone to high uncertainties and model errors. Many IAGOS observations are likely very close to urban areas and cover various atmospheric conditions. Moreover, the inversion does not show an upwards correction in the area where most IAGOS airplanes start and land (Frankfurt airport). I therefore think that such an argument requires a more detailed discussion or a clear statement on its limitations.

We agree that the comparison with the IAGOS-CARIBIC profiles is limited by spatial coverage of the aircraft observations. We have revised the text to warn the readers of this limitation (lines 237–244, Section 2.6): “*In addition, we also evaluate the inversion against near-surface vertical gradients derived from independent IAGOS-CARIBIC aircraft observations (Figure S8). Since most vertical profile observations are only available over Germany, where IAGOS-CARIBIC aircraft take off or land, this evaluation only provides information at local to regional scales.*”

Section 3

This section is clear and well structured, but see comment 11 above.

Section 4

14. The construction of the prior uncertainties makes it difficult for the reader to estimate these uncertainties on a country level. The authors should therefore provide the prior uncertainties to help interpreting the results. This can be done by including prior uncertainties in Figures 6, 7a, 9, and 10.

We have added prior uncertainty ranges to Figures 6, 7a, 9, and 10.

15. When providing posterior estimates for sectors, it should be clearly stated how the posterior sector attribution is done. This is only indicated in line 262, but required (approximately) in line 227.

We have revised the text in Section 2.5 to describe how sector attribution is done: “*We derive*

sectoral methane emissions by partitioning posterior emissions in each grid cell according to prior sectoral fractions. This approach is more effective where major sectors are spatially segregated, but may misattribute emissions in regions with significant sectoral overlap (Lu et al., 2022). Although sectoral information is not directly constrained by methane observations, the sectoral estimates offer useful interpretive insights.”

Section 5

16. Section 5 contains a comparison of the results to other works. This discussion could be extended to mention possible limitations of the method. (See general remark C above)

We thank the reviewer for this helpful suggestion. We have expanded the discussion in Section 5 to better acknowledge the limitations of the inversion method. In addition, following the reviewer's recommendation, we also updated the comparison in Figure 9 using the more recent ITMS-D v2025 and ICOS/PARIS InTEM-NAME datasets.

17. Line 264–265: Chen et al. (2025) found on average 27% higher whole-farm emissions compared to the Dutch National Inventory, but this number has a very large uncertainty. The authors could mention this high uncertainty to avoid any wrong interpretation.

We have revised the text, which now reads “Our attribution to agricultural emissions also aligns with a recent local-scale survey of 51 Dutch dairy farms, which reported average whole-farm methane emissions 27% higher than Dutch National Inventory estimates, though those local measurements carried substantial uncertainties of their own and may not fully represent national conditions (Chen et al., 2025)”.

Figure 9

18. The comparison of Figure 9 and Table S1 indicates that Figure 9 shows only results for 2023. This should be corrected in the caption.

We have revised accordingly.

19. Since Figure 9 compares to results by Worden et al. and Bruch et al. for 2019 and 2021, why not show results for 2021 instead of 2023? This would be more consistent. When mixing results for different years in the same figure, this should be made clear in the figure caption.

(If the authors prefer to compare their results for 2023 to other works, they could have a look at <https://doi.org/10.18160/GR1Q-6SK4> and <https://doi.org/10.5281/zenodo.20429764>. UNFCCC reported emissions are now also available for 2023: <https://unfccc.int/ghg-inventories-annex-i-parties/2026>.)

We thank the reviewer for this helpful suggestion and for providing the additional datasets. The primary purpose of Figure 9 is to assess country-level methane emissions in the most recent year of our inversion, and we therefore retain the results for 2023. To improve temporal consistency, we have updated the UNFCCC values to emissions for 2023 and have also included the two additional datasets suggested by the reviewer. The updated comparison leads to conclusions similar to those in the original manuscript, with generally consistent emission magnitudes across most countries. However, because some external datasets report total methane fluxes rather than anthropogenic emissions alone, the comparison remains primarily a qualitative reference for the magnitude of national emissions. We have also noted that estimates with large reported uncertainties should be interpreted cautiously. The new results are shown in the following figure.

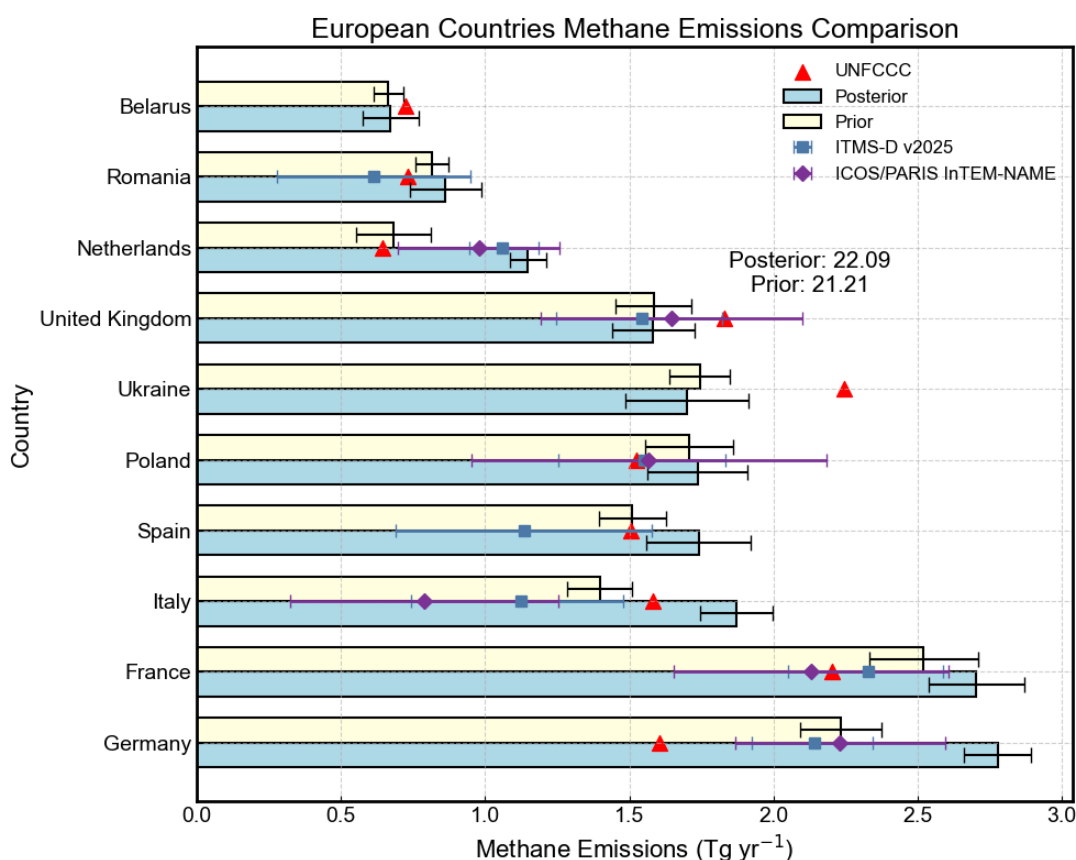


Figure 2. Comparison of Countries' result.

20. The data from Bruch et al. are total fluxes (anthropogenic plus natural) for 2021 with error bars for the 95% confidence interval (two standard deviations). This is not correctly described by the caption.

The original Bruch et al. (2025) comparison has been replaced by the 2023 ITMS-D v2025 and InTEM-NAME products. The revised caption now specifies the flux definitions and uncertainty ranges of both products.

21. For a precise reference to the UNFCCC data used in Figure 9, the authors should state the submission year of the reporting to the UNFCCC. (I assume that "UNFCCC 2021" means data reported for the year 2021 and not reported in the year 2021.)

We agree and have updated the UNFCCC values in Figure 9 from inventory year 2021 to inventory year 2023. The figure caption has also been revised to clearly indicate the inventory year of the reported emissions.

Figure 10

22. It should be mentioned explicitly in the figure caption that these results are for the year 2023.

Accepted.

Section 6

23. lines 323–324: The authors speculate that agriculture emissions in parts of western and central Europe might have a positive trend. It surprises me that they cite Bruch et al. (2025) and Mengistu et al. (2026) in this context. While both studies do see elevated agriculture emissions compared to the national inventories, they do not show or discuss such positive trends. (Mengistu et al. see a stagnation of agriculture emissions and Bruch et al. only show a single year.)

We thank the reviewer for pointing this out. We have removed Bruch et al. (2025) and Mengistu et

al. (2026) from the discussion of temporal trends, as these studies do not provide evidence for positive agricultural emission trends. Bruch et al. (2025) is retained only in Section 5.1 as contextual evidence regarding livestock emission accounting, while Mengistu et al. (2026) is no longer cited in the revised manuscript.

Supplement

24. The supplement seems unstructured. I recommend to add sections in the supplement that follow the structure of the paper:

1. Bias correction with Figure S4: As mentioned in comment 7.2 above, I would like to see an explanation of the bias correction. This could be part of the supplement or of the main text.
2. State space with Figure S1: This could be included in the main text, see comment 9 above.
3. Observation error covariance matrix $\mathbf{S}_o$ with Figures S2 and S3
4. Cross-validation exercise with Figures S5, S6, S8
5. Vertical gradient comparison to IAGOS-CARIBIC (Figure S7)

We thank the reviewer for this helpful suggestion. We have reorganized the Supplementary Material to provide a clearer progression from the inversion setup and error treatment to the evaluation and additional analyses. The final order differs slightly from the suggested sequence to accommodate the newly added figures and to preserve a coherent progression of the supplementary analyses.

Table S1

25. The geological emissions are missing in the table.

Accepted.

26. For many countries, the total emissions are lower than the sum of the sector emissions without geological emissions. Please explain this.

We thank the reviewer for pointing this out. We found that the original total emissions inadvertently included the soil methane uptake term. This has now been corrected. The corresponding values have been updated accordingly.

The following comments are just suggestions for improvement:

27. For convenience and to avoid misunderstandings, I would always provide some metadata (e.g. units) in such a table.

Accepted.

28. When providing such a table of the results, it would be helpful to include also prior and posterior uncertainties (at least for the total emissions).

Accepted.

Minor and technical remarks

29. There is a small inconsistency in the spelling: “western Europe” or “Western Europe”

30. line 47: Please check if there is a comma missing.

31. line 91: “in” doubled

32. line 97: “flagged as” doubled

33. line 120: possibly missing word: “... coal production **is** in ...”

34. line 121: “..., ~~are~~ **is** typically associated ...”

35. line 185 should be $\mathbf{S}_{\text{country}} = \mathbf{W} \mathbf{S} \mathbf{W}^T$ (transpose is missing)

36. line 186 should be $\mathbf{A}_{\text{country}} = \mathbf{I} - \mathbf{W} \mathbf{S} \mathbf{W}^T (\mathbf{W} \mathbf{S}_a \mathbf{W}^T)$ (transpose is missing)

37. line 239: the notation “ $p = \dots$ ” should be explained.

38. end of line 240–241: “posterior:” should probably be put in the first parenthesis.

39. Figure 3: When using an unevenly spaced colormap, I would prefer to have all tick labels (numbers) given explicitly. There are gaps between 0.05 and 0.5; 2.0 and 5.0; ...
40. Supplement: My personal recommendation would be to provide a PDF document instead of docx, but I leave it for the authors to decide on the file format.

We thank the reviewer for carefully identifying these minor and technical issues. All points in Comments 29–39 have been corrected in the revised manuscript and figures. We agree that a PDF version would improve readability and will follow the journal's requirements regarding the final format of the Supplementary Material.