

# Response to Reviewer 1

We thank the reviewer for the careful and constructive review of our manuscript. We appreciate the time and effort devoted to providing detailed feedback. The comments helped improve the clarity, methodological transparency, and overall structure of the manuscript.

## Summary of revisions

In response to the reviewer's comments, the manuscript has been revised to clarify the methodological framework and improve the interpretation of the results. A key improvement in the revised version is the replacement of the earlier Google Earth Engine (GEE)-based methane dataset with the WFMD TROPOMI methane product, which provides a more consistent scientific retrieval framework. The methodological description and filtering strategy have been updated accordingly.

### The main revisions include:

- improved description of the TROPOMI/WFMD retrieval methodology and the filtering strategy used to reduce potential surface reflectance artefacts;
- clarification of the conceptual scope of the study, emphasizing that the analysis focuses on spatial clustering in observed XCH<sub>4</sub> column concentrations rather than emission quantification;
- strengthened explanation of satellite data coverage and the number of valid observations underlying the spatial averaging grid;
- application of a log transformation to EDGAR emission data prior to spatial autocorrelation analysis in order to account for the multi-order-of-magnitude variability in emission values;
- revision of terminology throughout the manuscript to consistently distinguish between methane concentrations (TROPOMI XCH<sub>4</sub>) and methane emissions (EDGAR inventory data).

Detailed point-by-point responses to the reviewer's comments are provided below:

## Comment 1:

### Precautions when using satellite total column data – surface reflectance artefacts

As for other spaceborne GHG satellite instruments, TROPOMI methane total column data is obtained through a Full-Physics retrieval process, aiming to estimate the state of the atmosphere and of other interfering variables (e.g. surface reflectance) by matching the infrared spectrum observed by the satellite with a simulated spectrum (e.g. Hasekamp et al., 2025). Through this process, surface-related retrieval artefacts may occur, yielding patterns of XCH<sub>4</sub> that are spatially correlated with surface reflectance features. While these effects can be corrected to some extent (Lorente et al., 2023), some artefacts remain, for example along coastlines as the retrieval process is more difficult for pixels containing a mix of bright land and dark water (Balasus et al., 2023). Consequently, using data such as TROPOMI XCH<sub>4</sub> calls for a lot of precautions to carefully filter out surface-related artefacts, even more if the focus of the study is on spatial patterns.

## Response 1:

We thank the reviewer for highlighting the importance of accounting for surface reflectance artefacts when analysing satellite-derived methane columns. In the revised manuscript, we shortened and clarified this part of the discussion, because these effects are handled primarily through the use of the WFMD scientific product together with the applied quality filtering. We now state this more explicitly and document the filtering criteria used to reduce reflectance-related artefacts, especially in coastal and mixed land–water pixels.

### **Changes in manuscript**

- Section 2.2.1 now more clearly describes the full-physics retrieval framework of the TROPOMI/WFMD methane product.
- The applied filtering criteria are now explicitly listed: XCH<sub>4</sub> quality flag = 0, cloud proxy < 1.2, solar zenith angle < 75°, apparent surface albedo 0.02–0.35, and land fraction ≥ 95%.
- A short clarification has been added noting that potential reflectance-related artefacts are particularly relevant in coastal and heterogeneous surface conditions.
- The discussion of surface reflectance artefacts has been shortened, emphasizing that the updated WFMD product and the applied filtering already mitigate most of these effects.

### **Comment 2:**

#### **On total columns, enhancements, and emissions**

The definition of a “column-averaged dry-air mole fraction” is also very important to take into account when using the data to discuss emissions. It is a (normalized) vertically-integrated quantity (from the surface to the top of the atmosphere), computed from the vertical concentration profile of the considered atmospheric specie. This vertical concentration profile is the sum of (1) a background profile due to global scale transport and mixing, etc. and (2) of possible local concentration enhancements due to nearby emissions. Consequently, it is appropriate to estimate the local background column and remove it from the data in order to only leave local enhancements due to local emissions. This background removal can be done in different manners (e.g. removing local mean: Buchwitz et al., 2017). In addition, because methane concentration is higher in the troposphere than in the stratosphere, total columns over high-altitude areas are lower than at sea-level, making it seem that valleys or coasts have enhanced methane compared to the neighboring mountains, and thus elevated emissions. This “natural” surface elevation effect on total columns must be accounted for before data is used to discuss emission patterns (e.g. Buchwitz et al., 2017).

### **Response 2:**

We appreciate the reviewer’s comments regarding the interpretation of methane total column data in relation to emissions. In the revised manuscript, we clarified more explicitly the conceptual meaning of XCH<sub>4</sub> and the scope of our analysis. Specifically, we now state that the study analyses spatial clustering in the observed XCH<sub>4</sub> field over Iran and does not aim to quantify methane emissions or isolate local emission-driven enhancements. We also clarify that the WFMD full-physics retrieval already accounts for surface pressure and total air column mass, so the influence of surface elevation is considered within the retrieval framework. For this reason, and because the objective of the study is to analyse national-scale spatial patterns in observed methane columns rather than to attribute local enhancements to specific sources, no additional background subtraction was applied.

### **Changes in manuscript**

- The manuscript now clearly states that the study analyses spatial clustering in the observed XCH<sub>4</sub> field and does not attempt to quantify methane emission fluxes.
- Terminology has been revised throughout the manuscript so that methane concentrations are no longer described as emissions.
- Section 2.2.1 now clarifies that XCH<sub>4</sub> is a column-averaged dry-air mole fraction retrieved with a full-physics approach that accounts for surface pressure and total air column mass.
- The methodological explanation has been revised to clarify that, because the study focuses on national-scale spatial clustering in observed methane columns rather than source attribution or emission quantification, no additional background subtraction was applied.

- The manuscript now also states more clearly that surface elevation effects are addressed within the WFMD retrieval framework and are considered when interpreting spatial patterns across regions with different topography.

### **Comment 3:**

#### **On data coverage**

Finally, satellite data can have significant varying (e.g. cloudiness) or fixed (e.g. preprocessing excluding pixels with too much surface altitude change) coverage gaps. Because retrieved total columns have a random error (e.g. Hasekamp et al., 2025) and because locations with relatively little coverage are often challenging for the total column retrieval, it is important to monitor the number of satellite pixels underlying the average values reported in each element of temporal average grids. Grid cells relying on – relatively – too little data should be excluded from the analysis, or at least the results must be discussed against the data count.

The work by Vanselow et al. (2024) provides a comprehensive example for TROPOMI methane temporal averages of all the points of precaution made above. I strongly recommend that the authors read this article and some of its references.

I expect that including all these points of precaution in their analysis will help authors better discuss e.g. the significant hotspots that run along the Persian and Oman gulf coastline in Figure 3; or to assess e.g. to what extent grid pixels over mountainous areas should or should not be kept in the analysis.

### **Response 3:**

We thank the reviewer for emphasizing the importance of monitoring satellite data coverage and the number of observations underlying temporal averages. In the revised manuscript, we explicitly document how the number of valid observations per grid cell is evaluated and how grid cells with insufficient sampling are excluded from the spatial analysis to ensure reliable spatial statistics.

#### **Changes in manuscript**

- The manuscript now explicitly reports the minimum observation threshold required for grid cells to be included in the analysis.
- Grid cells are retained only if they contain at least 10 valid observation days during the averaging period, ensuring that spatial averages are not derived from insufficient data.
- The number of valid observations underlying the temporal averages is now explicitly considered when constructing the analysis grid, and cells not meeting the minimum data requirement are excluded from the spatial statistics.
- A clarification has been added noting that cloud-contaminated observations are removed during the initial filtering stage using the cloud proxy threshold applied to the TROPOMI retrievals.
- The manuscript now includes a short discussion on how varying satellite coverage (e.g., cloudiness or retrieval filtering) may influence the spatial representativeness of the averaged XCH<sub>4</sub> fields.

## Comment 4:

### Validity of the analysis comparing spatial autocorrelation hotspots between TROPOMI data and EDGAR v8

Assuming all points of precaution have been taken into account to use satellite XCH<sub>4</sub> data (see above), I still have some concerns on the analysis presented in this manuscript, especially on the comparison between TROPOMI and EDGAR autocorrelation analyses.

My first point relates to the difference in value distributions. TROPOMI XCH<sub>4</sub> data overall lies between 1850 and 1950 ppb, a rather narrow variability range compared to the baseline values. However, EDGAR v8 emission data spans at least 4 orders of magnitude across Iran (e.g. Figure 2 in Chen et al., 2023). As Moran-I statistics rely on the difference to the mean, I am concerned that the wide variability of EDGAR emission values across orders of magnitude somehow skews the mean and leads to misleading results. I would recommend that authors discuss the local Moran I cluster results for EDGAR, redoing it applying a log transformation on the emission data.

My second point relates to the overall meaning of comparing spatial autocorrelation clusters between TROPOMI XCH<sub>4</sub> data and EDGAR emission data. The clusters narrow down concentration and emission data into binary categories HH, LL, etc., dropping behind the full extent of information contained into the actual concentration/emission values that these data carry. The comparison of significant clusters in either or both TROPOMI and EDGAR data thus only allows for very qualitative conclusions at best. It also implicitly neglects the role of atmospheric transport that provides the link between concentrations (TROPOMI) and emissions (EDGAR). Inverse atmospheric modelling (e.g. Chen et al., 2023) can typically provide this link. Consequently, what is the added value of this spatial autocorrelation clusters comparison approach compared to inverse atmospheric modelling?

Finally, possibly because they treat atmospheric transport only implicitly in their analysis, I notice that the authors sometimes confuse concentrations for emissions, and vice versa, in the manuscript. For example: line 191: “The preliminary analysis of satellite-derived methane data across Iran revealed consistently high emission levels.” (concentration mistaken for emissions, what does high emissions mean, compared to what?).

line 314: “This high-resolution gridding enables consistent, grid-to-grid spatial analysis and direct comparison of methane concentrations between the two datasets.” (one dataset is concentration- TROPOMI - and the other is emissions - EDGAR).

## Response 4:

We thank the reviewer for the helpful comments regarding the comparison between TROPOMI methane column data and EDGAR emission inventories. In response, we revised the analysis to account for the wide distribution of EDGAR emission values by applying a log transformation prior to computing spatial autocorrelation statistics. We also clarified the conceptual purpose of the comparison, emphasizing that it serves as a spatial diagnostic comparison between observed methane column patterns and bottom-up emission distributions rather than a direct emission–concentration attribution.

### Changes in manuscript

- EDGAR v8.1 emission values are now log-transformed ( $\log_{10}$ ) prior to computing Local Moran's I in order to account for the multi-order-of-magnitude variability in emission values.
- The transformed EDGAR values are standardized before spatial autocorrelation analysis, and the resulting cluster patterns are interpreted accordingly.
- The manuscript now clarifies that the comparison between TROPOMI and EDGAR clusters is intended as a spatial diagnostic comparison between observed atmospheric methane column

patterns and bottom-up emission distributions, rather than a replacement for inverse atmospheric modelling approaches.

- A short clarification has been added noting that atmospheric transport is not explicitly modelled in this study, and that the comparison aims to highlight spatial agreements or discrepancies that may warrant further investigation using inversion methods.
- All instances where methane concentrations were mistakenly referred to as emissions have been corrected (e.g., “high emission levels” replaced with “elevated methane column concentrations”).
- The manuscript now consistently distinguishes between methane concentrations (TROPOMI XCH<sub>4</sub>) and methane emissions (EDGAR) throughout the text.

## Comment 5:

### Writing of the manuscript

I could not help but notice that, while the manuscript provides a smooth read, the discussion remains somewhat high-level, akin to the vague textbook prose that Large Language Models (LLMs) can produce. To verify this feeling, I had ChatGPT examine the whole manuscript with a prompt asking to identify if LLMs were used to write the manuscript, and if so in which sections. I am cutting out its extensive reply and only including here its final overview:

Abstract: high probability

Introduction: moderate to high probability

Materials and methods: low probability

Results: moderate probability

Discussion EDGAR vs TROPOMI: high probability

Conclusion: very high probability

To further confirm this result, I used GPTZero (<https://app.gptzero.me/>) to examine the EDGAR vs TROPOMI discussion text, and the conclusion:

lines 358-382 “Several studies [...] relevant mitigation measures”: GPTZero 4.1b model provides a 100% “high confident” assessment that this text was generated using LLMs.

lines 384-407, Conclusion: GPTZero 4.1b model provides a 90% “high confident” assessment that this text was generated using LLMs.

These tools cannot be fully trusted to perfectly assess the use of AI/LLMs in a given text, so I will not claim that I know for sure that LLMs were used to write at least parts of this manuscript. However, I have a strong feeling that it may very well be the case.

While the use of LLMs is not forbidden by Copernicus Biogeosciences’ guidelines, its use must be acknowledged somewhere in the manuscript (<https://www.biogeosciences.net/submission.html>) and it is not currently the case. Given my strong impression reading this manuscript, I urge the authors to include a statement on the

use or not use of LLMs within this manuscript (providing at least a section breakdown), should it be resubmitted after significant and critical revisions.

## Response 5:

We appreciate the reviewer’s comments regarding the writing style and the possible use of AI-assisted tools. We confirm that the scientific content of the manuscript—including the study design, data processing, statistical analysis, and interpretation—was developed entirely by the authors. In accordance with Copernicus Biogeosciences guidelines, we have now added a transparency statement acknowledging the

limited use of AI-assisted language editing tools for grammar refinement and readability improvements. In addition, the discussion and conclusion sections have been revised to strengthen the scientific interpretation and reduce overly general language.

### **Changes in manuscript**

- A transparency statement regarding the limited use of AI-assisted language editing tools has been added in the manuscript in accordance with Copernicus Biogeosciences guidelines.
- The EDGAR vs TROPOMI discussion section has been revised accordingly, with generic wording reduced and additional region-specific interpretation included.
- The conclusion section has been rewritten to improve clarity and focus on the key scientific findings of the study.
- A general revision of the manuscript language has been carried out to improve precision and reduce overly general or textbook-style phrasing.

## **Comment 6:**

### **Other miscellaneous points**

Given the critical nature of the three main points that I raised, I cannot currently recommend the publication of this manuscript. Besides these points, I did note other points of concerns/questions that I list here without much elaboration:

Use of “secondary references”

Some basic facts are referenced using citations of what could be called “secondary sources”, that cite the main reference, rather than being referenced with the citation of the main reference directly.

For example, in the introduction about methane 80 GWP:

Zhang et al., 2022: The Spatial and Temporal Distribution Patterns of XCH<sub>4</sub> in China: New Observations from TROPOMI

Jackson et al., 2024: Human activities now fuel two-thirds of global methane emissions

South et al., 2024: Methane Emissions from Oil and Natural Gas Operations—30 Percent Reduction by 2030 Possible if Domestic and International Actions “Stay the Course”

Subraveti and Anantharaman, 2025: Methane enrichment from dilute sources: Performance limits and implications for methane removal and abatement

Lerner., 2025: How the USA can feasibly cut methane emissions 30% by 2030: anaerobic digestion of organic waste and various measures in oil and gas production

Why cite those instead of the IPCC report?

The same goes for TROPOMI presentation in Section 2.2.1. For example, the mission presentation article is not cited: Veeffkind et al., 2012.

Figure 1b

Why are central bars missing in the histograms? What does “Mode” mean? Why is “Mode” significantly lower in 2023 and 2024?

Figure 2

I cannot see any LH and HL, can you provide illustration and explanations on such cases?

Section 3.2

Line 306: “Overall, methane levels in Iran show a pronounced upward trend”. We know that methane concentration is increasing globally. How is the trend in Iran different compared to the global one? Likewise for the seasonal variability: how is the Iranian seasonal variability in Methane concentration different that the one expected in the northern hemisphere?

## **Response 6:**

We thank the reviewer for these additional comments and suggestions. In the revised manuscript, we addressed the points raised regarding citation practices, figure clarity, and the interpretation of methane trends. Secondary references have been replaced with primary sources where appropriate, figure explanations have been clarified, and the discussion of methane trends and seasonal variability has been strengthened by placing the Iranian observations in the context of global methane behaviour.

### **Changes in manuscript**

- Secondary references: Where appropriate, secondary citations have been replaced with primary sources. For example, the methane GWP value is now referenced directly to the IPCC report rather than to secondary literature.
- TROPOMI description: The TROPOMI mission reference (Veefkind et al., 2012) has been added to Section 2.2.1 when describing the Sentinel-5P instrument.
- Figure 1b: The figure has been corrected following a layout issue identified in the original panel. The histogram construction and the meaning of the mode statistic are now clarified in the revised figure caption.
- Figure 2 (HL and LH clusters): Additional clarification has been added describing the interpretation of HL and LH spatial outliers, and illustrative examples are now provided in the manuscript and supplementary material.
- Section 3.2 – Methane trend: The discussion of the methane trend over Iran has been revised to place the observed increase in the context of the global methane growth rate, with comparison to NOAA global methane trends.
- Seasonal variability: The manuscript now compares the seasonal cycle of methane over Iran with the typical Northern Hemisphere seasonal variability, providing additional context for interpreting the observed patterns.

Sincerely,  
The Authors