

This manuscript by Rahimi et al. leverages observations by the spaceborne instrument TROPOMI to study spatial and temporal distribution patterns of methane total column (XCH<sub>4</sub>) over Iran. Global and local spatial autocorrelation analyses of TROPOMI XCH<sub>4</sub> data are performed, and the results are compared to a spatial autocorrelation analysis of inventory emission data from EDGAR v8. Temporal variability of XCH<sub>4</sub> over Iran from 2019 to 2024 is also described.

The manuscript reads well and its topic could – loosely – fit within the scope of Copernicus' Biogeosciences journal. However, it currently fails to meet minimal state-of-the-art precautions in using satellite total column data. Besides, I also have strong concerns on the validity of the analysis and on the writing of the manuscript. In the light of all these elements, in this current state, I cannot recommend the publication of this manuscript. The following paragraphs elaborate on these main points and provide a pathway towards significant revisions, should the authors aim to resubmit this work.

## **1 – Precautions using satellite total column data**

### ***On 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.

### ***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).

### ***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.

## **2 – 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)

### 3 – 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.

#### **4 – 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?

## References

Hasekamp et al., Algorithm Theoretical Baseline Document for Sentinel-5 Precursor Methane Retrieval v2.9.0, 2025, [https://sentiwiki.copernicus.eu/\\_attachments/1673595/SRON-S5P-LEV2-RP-001%20-%20Sentinel-5P%20TROPOMI%20ATBD%20Methane%20retrieval%202025-2.9.0.pdf?inst-v=484bd6a0-b045-46f7-904c-77cf35a903b1](https://sentiwiki.copernicus.eu/_attachments/1673595/SRON-S5P-LEV2-RP-001%20-%20Sentinel-5P%20TROPOMI%20ATBD%20Methane%20retrieval%202025-2.9.0.pdf?inst-v=484bd6a0-b045-46f7-904c-77cf35a903b1)

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