We appreciate the feedback from the reviewer. The comments from the reviewer are displayed in blue text, along with our responses, which are displayed in black text.

Response to RC1:

This paper uses an improved flux divergence algorithm to estimate sources of CH4 in the middle east. The results are compared with different emission inventories, and discrepancies are noted with both over and under estimations of emissions. Overall, the paper is well written and informative, the method seems sound and the results are interesting, timely and relevant. I am happy to recommend publication.

General comments

One weakness was some missing citations of recent work that is relevant. You cite Beirle et al., 2019, but should really also consider Beirle et al., 2022 which contains version 2 of the flux divergence method. Sun, 2022, provide further developments to the method. Although both of these are for NO2, they are for the same sensor and there is sufficient overlap with CH4 that merits their mention. De Foy and Schauer, 2023 estimate CH4 emissions in urban areas, including Tehran. It would be interesting to see the difference in estimates. Finally, Roberts et al., 2023 specifically look into the impact of missing data on CH4 flux retrievals. There may be further papers – it would be good to compare this paper with the latest publications.

We thank the reviewer for providing us the relevant information. Beirle et al., (2023) and Sun et al., (2022) have been added to the revised manuscript at L68. We believe the conclusion about the impact of missing data on the divergence method in Roberts et al., (2023) is not very suitable to be cited in our study because it specifically examines the impact of missing data at a very high spatial resolution (the typical radius of an emission field is 200-400 meter).

The emission estimated by de Foy et al., (2023) for 2017-2021 is 953 kt/yr over Tehran based on Table C1, which is 8.3 times higher than EDGAR v6.0's estimates (114 kt/yr) used in that paper. Our estimate over Tehran is 220 kt/yr for 2018-2021, which is about 2 times higher than EDGAR v6.0 and much closer to the conclusion in that paper: "We show that methane emissions from urban areas may be underestimated by a factor of 3–4 in the EDGAR greenhouse gas emission inventory". The possible reasons could be different methods used to determine the background and calculate the total emission of the area. The Gaussian model in that paper treated the urban area as one large source and integrated the emissions along the "plume", whereas our total emission for a certain area is the sum of individual sources derived from the divergence. We added this comparison in our revised manuscript at L515:

"Our result (220 kt/yr for 2018-2021) is much lower than the emission estimated by de Foy et al., (2023) (953 kt/yr for 2017-2021) over Tehran, which is 8.3 times higher than

EDGAR v6.0's estimates (114 kt/yr) used in that paper. The possible reasons could be different assumptions of the regional background and the methods to calculate the emission of the area. The Gaussian model used by de Foy et al., (2023) treated an urban area as one large source and integrated the emissions along the "plume", whereas our total emission for a certain area is the sum of individual sources that are derived from the divergence method."

Line 167 to 187: I was a bit skeptical of the boundary layer treatment: fixing PBLH at 500m seems rather crude. However, I notice that you published this already in your previous paper. I do wonder what would happen if you used actual PBLH from ERA5,

Liu et al., (2021) found that enhancements of XCH_4 due to the transport in the upper atmosphere should be removed before calculating the divergence, as they are irrelevant to the ground-level sources. Therefore, identifying the height of PBL is not very important, but estimating the XCH_4 in lower atmosphere is more relevant to the ground emissions. We used surface pressure, XCH_4 and total dry air density from TROPOMI observation, as well as the XCH_4, temperature and relative humidity profile from EAC4 (60 layers for the whole atmosphere, whereas TROPOMI uses 12-layer profiles for retrieval, which is too coarse to resolve meteorological dynamics), to estimate XCH_4 below 500 meters (referred as "PBLH" in our paper). The favorable height is suggested to be 500-700 meters above the ground considering the systematic difference between EAC4 dataset and TROPOMI observations (Liu et al., 2021). Using either a too shallow or too thick layer as "PBLH" can magnify the bias. To reduce the uncertainties caused by singular values in model simulation, we fixed the height at 500 meters. The way to describe the "500 meters" as "PBLH" in our original manuscript is indeed a little confusing. We addressed this in the revised paper at L191 and L201:

"Estimating the XCH_4 in lower atmosphere is quite important since the enhancement due to the transport in the upper atmosphere is irrelevant to the ground emissions."

"We fixed the PBLH at 500 meters above the ground considering the PBLH from the reanalysis dataset has large uncertainties and is occasionally too shallow (Guo et al., 2021). The favorable height is suggested to be 500-700 meters above the ground considering the systematic difference between EAC4 dataset and TROPOMI observations (Liu et al., 2021)."

Coastal regions present a particular challenge in terms of data filtering. Some of your figures do seem to suggest that there are anomalous retrievals near coastlines. It might be that more careful filtering of boundary retrievals is necessary compared with the default land/water mask used. This could be discussed for future reference.

Actually, we applied a new mask to identify water, land and the boundary, but that was not mentioned in the main text. Here we added the content in our revised manuscript at L116:

"Another aspect that is addressed is the distinction between land and water bodies, especially over the coastlines. TROPOMI use different retrieval strategies for data over land and ocean. The retrievals over ocean are only available in sun glint mode. We find the data over ocean can be quite noisy. Furthermore, the data continuous from land to ocean are checked. We selected pixels locating at several 1° ×1° areas covering half land and half ocean at the coastlines of Oman, Yemen and along the Red Sea. We found there are not many differences between pixels over land and ocean (see Figure S1 in SI). Therefore, we built a water-land mask at the same spatial resolution as our emission data (0.2° ×0.2°) based on Global Land Cover Characterization (GLCC) of the United States Geological Survey (USGS) (United States Geological Survey, 2018a, b) to distinguish water, land and the coast (transition grids from land to water). Only grid cells that are marked as land and coast are used to build the regional background and are used to calculate the daily divergence."

Line 325: Is it really feasible that a farm in the desert would produce a detectable amount of CH_4? I think that this should be backed up with a bit more information if it is to stay here – information about the agricultural emissions, comparisons with farms with known CH_4 emissions, threshold values for TROPOMI.

Actually, there is not only one farm in the grid cell. At least, there are about 135.000 cattle in six big farms in this area through Google Earth Image (see the figure below), one of them is the AlMarai farm which is one of the biggest dairy farms in the world (Shadbolt et al., 2013). But indeed, we do not have observation or reference to directly compare with.

Reference:
Shadbolt, P. : Milking the desert: How mega-dairies thrive in Saudi sands, CNN news, available at: https://www.cnn.com/2013/12/18/world/meast/milking-the-desert-saudi-dairy-farms (Last access: 16 June, 2024), December 18, 2013

Minor points:

Fig. 5g&h: "Mehttane"

Changed.

Line 137: despite *the fact* that the three

Changed

Line 449: 3 kg/m2 (remove extra /)

Changed to "3kg/km^2/h"

Suggested References:

Beirle, S., Borger, C., Jost, A. and Wagner, T., 2023. Improved catalog of NOx point source emissions (version 2). *Earth System Science Data Discussions*, *2023*, pp.1-37.

de Foy, B., Schauer, J.J., Lorente, A. and Borsdorff, T., 2023. Investigating high methane emissions from urban areas detected by TROPOMI and their association with untreated wastewater. *Environmental Research Letters*, *18*(4), p.044004.

Sun, K., 2022. Derivation of Emissions From Satellite-Observed Column Amounts and Its Application to TROPOMI NO2 and CO Observations. *Geophysical Research Letters*, *49*(23), p.e2022GL101102.

Roberts, C., IJzermans, R., Randell, D., Jones, M., Jonathan, P., Mandel, K., Hirst, B. and Shorttle, O., 2023. Avoiding methane emission rate underestimates when using the divergence method. *Environmental Research Letters*, *18*(11), p.114033.