Satellite-based global monitoring of urban-scale methane emissions

Reviewer #2

This manuscript presents an improved source-pixel method to estimate urban methane emissions from single-pass TROPOMI observations over London, Los Angeles, and New York during 2021–2023. The use of PBL-weighted winds and an upwind background concentration is a useful development, and the manuscript is well organized and easy to follow. The manuscript would be strengthened by further clarification and justification of key method assumptions, particularly the wind fields and the calculation of background concentrations. I recommend publication after the authors address the following comments.

1. The methane enhancement observed by TROPOMI can reflect emissions accumulated and transported over several hours before the overpass. It would be helpful if the authors could discuss and test the sensitivity to a 3-hour pre-overpass mean wind compared with the 1-hour overpass-time wind.

Response: Thank you for this constructive comment and idea. We tested the sensitivity of orbit 17480, which passed over London on 26 February, and found that the emission estimate of 3-hour pre-overpass mean wind was 24.79 ± 14.37 th^{-1}, which is not significantly different from that derived using the 1-hour overpass-time wind 25.74 ± 14.13 th^{-1}, and well within respective uncertainty. In addition, in the present study, we have carefully considered the influence of dynamic changes in wind fields before and after the local overpass time, which propagate into the uncertainties of urban-scale emission estimates. The uncertainty in wind speed was estimated from the standard deviation of wind speed over the source region in a period of four consecutive hours around the TROPOMI local overpass time. These uncertainties were further propagated into the final methane emission estimates through Equation (5). This wind uncertainty analysis in generally analogous to the sensitivity test suggested by the reviewer and is therefore reflected in our uncertainty budget.

2. As the authors noted, single-pass TROPOMI observations can be sparse and uncertain over urban areas, while temporally averaged fields are often used for emission estimates. Could the proposed approach be applied to time-averaged methane enhancements (e.g., monthly means) to increase the number of usable observations?

Response: The proposed approach can be applied to time-averaged methane enhancements for further emission estimation in principle, but we wouldn't recommend it as boundary layer winds can vary significantly between orbits. The method is most valid when the wind speed and direction are known at the time of the overpass. In this study, we calculate methane enhancement at the orbit-level because daily observations can be tracked individually and can provide insight into day-to-day variability among different overpasses. Although integrating overpasses over a longer period can increase the number of valid observations, errors due to wind variability can compound. In addition, temporal variations in wind fields may be overlooked when time-averaged methane enhancements are used to estimate final emissions, because the wind field associated with each overpass is different. Therefore, we would recommend that flux is evaluated at the orbit level only, and any temporal averaging is applied thereafter (as we have done in this study for annual averages for example).

3. The approach implicitly assumes no significant methane sources in the background region, so that the methane enhancement can be attributed to the source region. However, Figure A1 suggests substantial emissions around some source regions, which could bias the background concentration and inferred emissions. It would be useful to discuss the sensitivity of the results to the size or location of the background region, or to screen the background regions using available inventories.

Response: This is a very insightful observation and something we laboured over significantly when applying and testing the new method. The calculation of methane enhancement in source pixel methods which do not use a directly upwind background assume that the background region doesn't contain significant emission sources and neglect possible enhancements in the background region caused by outflow from the source region. In those methods, this could bias the methane enhancements and emissions if substantial emissions occur around the source region. In our variation of the source pixel method, emissions in the background region are implicitly accounted for and subtracted such that the new method does not necessitate the background region to be completely free of methane emission sources. This is because we take an average concentration in an area that is directly upwind of the target source area, which other methods have not previously. This is an advantage compared with other source pixel methods. The background region in this study is defined as an equal area region located upwind of the source region. It is oriented such that two sides of the box are perpendicular to the pressure-weighted PBL wind direction, and the full width of the emitted urban plume is observable over the city. We have added an analysis of the variability in calculated methane enhancements for overpasses associated with different upwind-oriented background locations over the yearly period, as shown in a new additional Figure, A3. To better describe this, we have added new text. A clearer description can be found in the revised manuscript (Line 214-218, 225-231).

In our method, it is important that the length scale of the upwind edge (perpendicular to mean PBL wind) is the same for the source region and the background region, otherwise key assumptions in the method (such as accounting for sources upwind) become invalid. Therefore, changing the size of the background region would not be a valid approach here, and expanding it further in the upwind direction would not add new information, and could potentially add unwanted information that could bias the background. What is important is that the area upwind is similar to the scale of the source region, which validates the idea that any emissions advected from the background area to the source area are implicitly accounted for. We hope this makes more sense. It is a very difficult concept to convey, but it is clear that the reviewer understands these issues well - the challenge here is in optimizing the validity of the assumptions implicit to emissions mass balancing, which is what we believe we have achieved by using a background that is directly upwind, and of the same size as the source region under evaluation.

4. Negative ΔXCH_4 values and negative emission estimates, especially for London, are not physically meaningful. A more careful treatment of these cases is needed, for example by testing sensitivity to alternative background regions or excluding them from averaged emission estimates.

Response: Thank you for pointing out the potential concern regarding the treatment of negative methane enhancement and emission estimates. We agree it is not physically meaningful, but there is an important nuance to how to treat such retrievals to avoid bias. Negative values in methane enhancement and emissions are relatively common when emission estimates are derived from TROPOMI retrievals. Previous studies, such as Maasakkers et al. (2021) and Vanselow et al. (2024), which used different methods over different regions, also reported negative flux values during the flux quantification process and didn't exclude them when averaging emissions, for entirely valid reasons. Selecting only positive fluxes would bias results because a negative flux can sometimes result in response to random TROPOMI retrieval error. As random errors can be expected to average out over a significant number of independent measurements, it is important to include negative results when averaging over annual timescales. Excluding negative values could introduce a positive bias into the final results, even though we applied a set of stringent filtering criteria to preserve statistical reliability and avoid systematic overestimation or underestimation of emissions. We therefore follow, and agree with, previous approaches and retained negative values in the final emission estimates. What is more important is that the emissions uncertainty reflects this source of potential error, which is accounted for by the method and shown in the error bars in each case. In addition, a boxplot of methane enhancement for each overpass over the source region, calculated using the upwind background locations according to the PBL pressure-weighted wind direction, has been added to the revised manuscript as Figure A3.

Maasakkers J D, Omara M, Gautam R, et al. Reconstructing and quantifying methane emissions from the full duration of a 38-day natural gas well blowout using space-based observations. Remote Sensing of Environment, 2022, 270: 112755.

Vanselow S, Schneising O, Buchwitz M, et al. Automated detection of regions with persistently enhanced methane concentrations using Sentinel-5 Precursor satellite data. Atmospheric Chemistry and Physics, 2024, 24(18): 10441-10473.

5. The methane enhancement ΔXCH_4 appears small in some cases, for example below 2 ppb in London. Do such weak enhancements meet the detection/signal-to-noise criteria discussed in Jacob et al. (2016), and how reliable are the resulting single-pass emission estimates?

Response: Thank you for your comment. We carefully examined this issue during our data analysis. Jacob et al. (2016) reported that with a detection limit of 4 t h^{-1}, TROPOMI can detect methane emissions from a single overpass (1 day). For the valid overpasses over London during 2021 - 2023, only three orbits did not meet this reported detection threshold when considering the absolute emission values, indicating that the detectable influence is negligible. Fluxes from individual orbits with relatively small methane enhancements and correspondingly small emission estimates are averaged during the temporal aggregation. Emissions from any city can be expected to be highly variable - for similar reasons to including negative emissions when averaging over annual timescales, it is equally important to include small emissions that meet the data selection criteria for consistency. To directly answer the final question - an emission estimate from a single overpass is far from reliable, but an average over a large number of overpasses converges for greater accuracy. What is perhaps more important is that a systematic/methodological error/uncertainty is derived for fluxes from each overpass, which we calculate explicitly.

6. For coastal cities such as Los Angeles and New York, the upwind background region may fall over the ocean, where TROPOMI retrievals are filtered out under Sect. 2.1.1. Does this occur, and if so, how are background concentrations calculated?

Response: Thank you for pointing this out. Yes, this can indeed occur and we looked in detail at the impact of this. For some overpasses, the upwind direction may originate from the ocean, resulting in insufficient valid observations within the upwind background region for further background concentrations calculation. These overpasses are therefore excluded from the aggregated monthly or yearly emission estimates, resulting in a large amount of lost overpasses for coastal cities. For example, in Los Angeles 2021, there were 129 overpasses over the source region, but only 32 of them were used in the final emission estimation because of the stringent filtering criteria applied in this study.

7. The apparent increase in Los Angeles emissions from 26 t h-1 in 2021 to 62 t h-1 in 2023 appears inconsistent with previous reported decreasing trends in this region (https://doi.org/10.1021/acs.est.5c09817, https://doi.org/10.1088/1748-9326/acb6a9, https://doi.org/10.1038/s41467-023-40964-w), and may reflect sampling or methodological biases. Similarly, the large error bars and limited data coverage limit confidence in the inferred seasonal cycles. Further discussion of the uncertainties in these temporal variations would be helpful.

Response: We appreciate the reviewer's insightful suggestion and we agree that is not possible to confirm a trend (within error) as the uncertainties overlap in our results between years. It's even possible (within error bounds) that methane emissions over Los Angeles exhibit a decreasing trend similar to that reported in previous studies, given the large error bars in the final emission estimates and the substantial overlap among them. In addition, the influence of urban domain definition on urban-scale emission estimates should also be considered. For example, the three studies that reported the decreasing methane emissions focused on the Los Angeles Basin, whereas Whiting et al. (2026) reported a different increasing trend, similar to our study, using a different square urban domain definition. We have discussed this discrepancy in the revised manuscript (Line 351-353).

Regarding the influence of large error bars and limited data coverage on the inferred seasonal cycles, we have highlighted this limitation and the associated uncertainties in these temporal variations in the revised manuscript (Line 260-268, 479-484 in the revised manuscript) and removed any affirmative suggestion that there is a definitive trend in emissions between years.

8. The comparison with previous top-down studies in Sect. 3.4 needs more clarification. The statement that this study finds "generally lower, yet overall consistent" emission estimates could be misleading, because the urban domains used here are much smaller than those in previous studies. Although the authors rescale previous estimates using emission intensity, this assumes spatially uniform emissions, which not hold for heterogeneous urban sources such as landfills and gas infrastructure. A more direct, apple-to-apple comparison would be preferable; otherwise, the uncertainty introduced by the rescaling method should be acknowledged.

Response: Thank you for this comment. On reflection, We agree this was somewhat of an oxymoron. We agree that further clarification regarding the comparison of emission estimates among different studies is needed in Section 3.4. A direct apple-to-apple comparison is difficult because gridded emissions were not provided in most previous studies. We acknowledge that differences in the definitions of urban domains and background concentrations make direct comparisons complicated. And we have carefully considered the rescaling method used in the present study and revised the manuscript accordingly (Line 415-423, 439-445).

Minor Comments:

1. In the Abstract, "combat climate changing emissions" would be better phrased as "mitigate climate change" or "reduce climate-forcing emissions."

Response: Thanks for this comment, we have revised the manuscript accordingly (Line 2).

1. Some statements in the Abstract and Introduction overstate the limitations of previous studies and imply that this study provides a more accurate emission estimation method (lines 4-5, lines 60-62). I suggest softening these claims and focusing the motivation more on the simplicity, efficiency, and urban-scale applicability of the source-pixel approach.

Response: The advancement of our method lies in the incorporation of hourly resolved PBL-pressure weighted wind fields and an explicitly defined upwind background concentration. These refinements enhance the conceptual accuracy of the approach relative to the conventional source pixel method, which does not explicitly account for upwind background direction or the vertical structure of wind within the PBL. The relevant statements have been explicitly clarified in the revised manuscript(Line 3-6, 64-67).

2. Abstract and line 375: Factors of 0.1 and 0.3 indicate emissions lower than the inventory estimates, not higher. The two reported ranges are inconsistent and need clarification.

Response: The value has been corrected accordingly in the revised manuscript (Line 17, 400).

3. Line 22: Revise to "...the second most significant anthropogenic greenhouse gas..."

Response: This has been clarified in the revised manuscript accordingly (Line 25).

4. Line 30: Grammar issue in "natural and processes".

Response: This grammar issue has been corrected in the revised manuscript (Line 33).

5.Line 33: Tibrewal et al. (2024) refers to fossil fuel sectors rather than all sectors mentioned in the preceding sentence.

Response: Thank you for bringing this to our attention. We have now cited the appropriate references in the revised manuscript (Line 36-37).

6. Line 53: "actual emissions" is too strong and could be removed.

Response: The manuscript has been revised accordingly (Line 56).

7. Figure 1, line 156, and elsewhere: Please correct the resolution format, such as "0.25° × 0.25°", "1 km × 1 km", or "1 × 1 km2".

Response: We have corrected and standardized the resolution format throughout the revised manuscript, including Figure 1 and line 161 (Line 161).

8. Section 2.2: From my understanding, the authors mainly remap individual TROPOMI pixels onto a 0.1°× 0.1° grid, which differs from oversampling that usually averages multiple overpasses

to obtain a higher effective spatial resolution. I suggest using “regridding” or “remapping” instead of “oversampling”.

Response: We appreciate the reviewer’s concern regarding the use of the term “oversampling.” We agree that, in this study, we do not oversample the irregular original TROPOMI pixels into grid cells finer than the pixel size through temporal averaging. Instead, we apply an area- and uncertainty-weighted remapping method to map valid pixels from each individual overpass onto a regular $0.1^\circ \times 0.1^\circ$ grid. This enables orbit-level methane emission estimates to be derived consistently. We have therefore revised the terminology and clarified the difference between the method used in this study and traditional oversampling methods in the revised manuscript (Section 2.2).

9. Lines 316 and 369: “Landfills and waste” is inaccurate, as landfills are part of the waste sector.

Response: We have corrected this statement in the revised manuscript (Line 337, 394).

10. Line 377-379: Please clarify the reference used for the reported percent differences.

Response: These percentage differences were derived from comparisons between emission estimates reported in our study and publicly available inventories, no other previous researches were referred to in this comparison. This has been clarified in the revised manuscript (Line 401-404).

11. Figure 7: Please clarify why the adjusted Los Angeles estimate is larger than the original Nesser et al. (2024) estimate, given the smaller domain used here.

Response: The urban domain for Los Angeles used by Nesser et al. (2024) is based on the U.S. Census Bureau’s Topologically Integrated Geographic Encoding and Referencing system (TIGER)/Line urban area definition, corresponding to an area of approximately 4288 km^2. By contrast, although the administrative urban boundary used in our study is smaller, our emission estimates are calculated based on the square source box defined by the urban boundary, which covers an area of approximately 4948 km^2. This difference in the source region area explains why the adjusted Los Angeles estimate is larger than the original result reported in that previous study.

12. Previous intercomparisons suggest that de Foy et al. (2023) lies the upper bound of top-down urban methane estimates (https://www.science.org/doi/full/10.1126/sciadv.adz9308). This would

be useful to acknowledge when discussing consistency with previous studies.

Response: Thank you for this valuable comment. This has been clarified in the revised manuscript (Line 415-423, 442-445).