

We are grateful to the referee for thorough evaluation of our manuscript. Our responses to the comments are detailed below. Please note that *the referee's comment* and our responses are in different styles. The page and line numbers shown below correspond to those in the revised manuscript.

We note the following corrections: (1) Figure 4 (b) has been modified with color shades added according to another referee's suggestion. (2) The analyzer's name "MIRA ULTRA" at every place has been changed to "MIRA Ultra" to be consistent with the producer. The labels of Figure 3 have been corrected accordingly.

This study provides valuable insights into urban methane emissions in Tokyo, employing mobile vehicle-based measurements of CH₄ and C₂H₆. The work is timely and relevant for improving city-level emission inventories, especially given discrepancies between bottom-up (Tokyo Metropolitan Government, EDGAR) and observation-based estimates. The integration of a controlled release experiment is a strength, although it also highlights significant uncertainties in emission quantification.

The manuscript makes a novel and policy-relevant contribution to understanding urban methane emissions. I recommend minor revisions to address the issues of emission quantification uncertainty, spatial representativeness, and inventory implications. With these improvements, the study will provide a strong addition to the literature on methane emissions in megacities.

Overall, I find this paper suitable for publication after addressing several concerns outlined below.

We again thank the referee for constructive evaluation of our manuscript. Please see our responses below.

Minor Comments

L20-24: The abstract's conclusion is vague. It mentions an inconsistency with local government reports for residential areas but doesn't specify how. To enhance the impact, clearly state whether the measured emissions were higher or lower than the reported values. This highlights the need for improved accounting in specific sectors and underscores the value of direct-measurement research.

The abstract effectively introduces the scope and importance of the study but lacks quantified uncertainty. Including error ranges (e.g., $\pm$ values for emission estimates) or highlighting explicit policy implications would improve clarity for broader audiences.

We appreciate this comment which pinpoints importance of this study. It is unfortunately difficult for the moment to clearly state possible under/overestimation of local government reporting due to the large uncertainties in our emission estimates. Our current best arguments from the preliminary emission calculations are (1) the estimates are well correlated with the reporting for the areas with waste-sector facilities (Chiyoda, Minato and Koto Wards), showing actual key roles of emissions from the facilities, and (2) fossil fuel emissions in the residential areas (Shibuya and Toshima Wards) are comparable in magnitude to those in the areas with the waste facilities, but they are not accounted in the reporting. We have revised the last sentence of the abstract as follows:

P1 L23: "In the areas with biogenic facilities (landfill and wastewater treatment plants), our emission estimates are well correlated with local government reporting, indicating actual key contributions of the waste-sector facilities. On the other hand, in the residential areas, CH₄ emissions were predominantly of fossil-fuel origin, with a magnitude comparable to the area with waste facilities. However, such fossil-fuel emissions are not accounted for in local

government reporting. This result highlights the need for improved accounting of urban fossil fuel-related emissions.”

Line 14-16 The phrase “conversion is not straightforward” is vague. Suggest clarifying what aspects are most uncertain (e.g., dispersion variability, background definition, or calibration transferability).

We have corrected the sentence and added a new sentence as follows:

P1 L14: “The empirical equation derived from the experiment was significantly different from those reported by previous studies, suggesting the limitation of such enhancement-to-emission rate conversion, which is a source of large uncertainty in estimating urban CH₄ emissions based on street-level measurements. The uncertainty stems from different experiment settings and underlying assumptions (e.g., source distance and height) which do not always represent actual urban measurement environments.”

L64-70: I agree with that “Discrepancy between the local government reporting and the global data commonly used in the atmospheric science community”. The discussion of the discrepancy between local government reporting and global datasets is important and well-motivated. While the year-to-year differences in reported values may not be dramatic, the comparison currently refers to different years (2021 vs. 2023). I recommend explicitly noting this difference in the manuscript, as it will help readers interpret the magnitude of discrepancies more clearly.

We appreciate the referee for the important comment. Since Bureau of Environment of Tokyo Metropolitan Government updated the greenhouse gas emission reporting in the meantime, we have shown the emission for 2023, by which comparison to EDGAR is now made for the same year. The update is small as in general CH₄ emission in the Tokyo reporting gradually decreases. In addition, we have modified the relevant sentence as follows:

P3 L73: Given the general small decreasing trend of CH₄ emissions of Tokyo (e.g., 0.5 % yr⁻¹ in the Tokyo Metropolitan Government reporting), comparison of these datasets indicates discrepancy in magnitude and attribution of CH₄ sources in Tokyo between the local government reporting and the global data commonly used in the atmospheric science community, highlighting the importance to improve activity-based CH₄ emission datasets for Tokyo.

L93-95: The study covers ~2,000 km of road network (~10% of Tokyo), with higher coverage in selected wards, particularly near waste facilities. This uneven sampling may affect representativeness and source attribution. A brief note on this limitation and its implications for generalizing results would improve clarity.

We acknowledge the possible sampling bias, which is discussed in Section 5.2. For conciseness, we do not add explanation here.

L180-220: The Control Release experiment is a valuable addition. However, since it was conducted under specific wind conditions, its generalizability is somewhat limited. It would strengthen the manuscript if the authors briefly acknowledge this limitation and note that further experiments under varied meteorological conditions could be beneficial.

To mimic actual on-street measurements, the control release experiment was conducted downwind of the point source with wind speed of <3 m s⁻¹, as described in the manuscript. The meteorological condition agrees to predominant summertime weather in the area, and more importantly, the wind speed was well within the variability in the Tokyo area (Japan Meteorological Agency, 2025). Therefore, we consider that the result is well representative and useful for the present study, although we acknowledge that differences from actual

measurements were made under to some degree various meteorological conditions. We have added the following sentence in the revised manuscript:

P8 L201: “These weather conditions agree to those typically observed at the site and in Tokyo (Japan Meteorological Agency, 2025).”

Japan Meteorological Agency, Tables of Monthly Climate Statistics:

<https://www.data.jma.go.jp/stats/data/en/smp>, last access: 29 October 2025.

L231-233: The choice of a 0.1 ppm threshold for defining LPs is central but appears somewhat arbitrary. While Tokyo’s relatively low CH₄ enhancements justify a lower threshold, a statistical validation (e.g., false positive/negative analysis) would provide stronger support.

We have added the following sentences in Section 4.1:

P10 L249: “For instance, a higher threshold of 0.2 ppm would miss ~60% of LPs with smaller enhancements. In contrast, a lower threshold of 0.05 ppm would more than double the LP counts, but, due to comparable magnitudes of the observed baseline CH₄ variability, the LP detection would entail larger uncertainty.”

L264-272 and overall results: The use of C2/C1 thresholds (<0.005 biogenic, 0.005–0.1 fossil fuel, >0.1 combustion) is appropriate, but the combustion category is underrepresented (only ~5% of LPs, ~30 samples). Given the small sample size, confidence in combustion attribution is limited.

In dense residential areas with potential mixed sources, interpretation could be more complex. It would be useful to briefly acknowledge this limitation and, if appropriate, mention complementary methods (e.g., isotopes, co-tracers)

We agree to the referee. The following sentence has been added. For conciseness of this manuscript, we avoid additional discussion about possible complementary tracers.

P12 L288: “In this study, due to the very limited number of the combustion LPs, our analysis below mainly addresses biogenic and fossil fuel sources.”