Responses to reviewer's comments. (Reviewer 1)

Ueyama, et al.

Evaluating urban methane emissions and their attributes in a megacity, Osaka, Japan, via mobile and eddy covariance measurements

Revisions are highlighted in red, and sentences that have been moved are highlighted in green.

We would like to express our sincere gratitude for the constructive and insightful comments to improve our manuscript. In response to the three reviewers' suggestions, we have thoroughly revised the manuscript by clarifying methodological assumptions and associated uncertainties, and by incorporating a wind sector analysis of the eddy covariance data. We consider that these revisions have substantially improved the manuscript and have enhanced its value for readers interested in urban CH_4 emissions. We have also reviewed the manuscript again and made minor revisions to address inconsistencies in terminology and other minor issues. Since the latest local inventory in 2020 was provided by Sakai City, the inventory-based CH_4 emissions for Sakai and the corresponding discussion were updated in the revised manuscript. We are confident that the revised version meets the standards of Atmospheric Chemistry and Physics. Once again, we sincerely thank you for your thoughtful and valuable feedback.

Accepted!

In my opinion, this is an excellent paper!

Temporal and spatial variability of greenhouse gas exchange between the atmosphere and various ecosystems is one of the most critical problems of global climatology/ecology/environmental science. Despite the growing number of measurement sites worldwide, it should be noted that (for various reasons) their number in cities is insufficient. And yet, cities are such intensive sources of GHGs to the atmosphere! In addition, the existing urban sites mainly focus on measuring carbon dioxide fluxes, and long-term measurements of methane fluxes are still few (the results of only a few long-term measurement campaigns in the UK, Poland, Japan, or Italy have been published). Thus, it should be emphasized that the research results presented in the manuscript are valuable to knowledge of urban methane emissions.

The article presents the results of a detailed planned research experiment during which measurements were made using the EC method and during mobile measurements with methane concentration sensors installed on a car and a bicycle. The study is written clearly, and the Authors describe the results in detail. Noteworthy is the detailed description of the methodology and the extensive discussion of the results' quality (and their comparison with inventories). Some discrepancies in the results obtained from mobile measurements by car and bicycle are also discussed in detail by the Authors. The Authors also devoted much attention to estimating CH4 flux components, which I consider the most critical achievement in the presented study.

Thank you for your kind encouragement and for recognizing the importance of this manuscript.

I have only two comments:

P4, figure 4 should be more detailed (larger?). In the current figure, the differences in development, and especially the EC footprint, are poorly visible!

To address the comment, we have added an enlarged view of the area surrounding the tower site and the flux footprint, now presented as Fig. 1b.

P6L168 – should be (Vickers and Mahrt, 1997)

We have corrected the name of the authors.

Responses to reviewer's comments. (Reviewer 2)

Ueyama, et al.

Evaluating urban methane emissions and their attributes in a megacity, Osaka, Japan, via mobile and eddy covariance measurements

Revisions are highlighted in red, and sentences that have been moved are highlighted in green.

We would like to express our sincere gratitude for the constructive and insightful comments to improve our manuscript. In response to the three reviewers' suggestions, we have thoroughly revised the manuscript by clarifying methodological assumptions and associated uncertainties, and by incorporating a wind sector analysis of the eddy covariance data. We consider that these revisions have substantially improved the manuscript and have enhanced its value for readers interested in urban CH_4 emissions. We have also reviewed the manuscript again and made minor revisions to address inconsistencies in terminology and other minor issues. Since the latest local inventory in 2020 was provided by Sakai City, the inventory-based CH_4 emissions for Sakai and the corresponding discussion were updated in the revised manuscript. We are confident that the revised version meets the standards of Atmospheric Chemistry and Physics. Once again, we sincerely thank you for your thoughtful and valuable feedback.

General comments

This paper reports on a study of CH4 emissions in two Japanese cities using both eddy-covariance (EC) and mobile measurements of CH4 and C2H6 at street level. The mobile measurement campaign covered a lot of ground using two modes of transport (car and bicycle). The aim of the study was to identify different sources of CH4, quantify their emissions, upscale them to city level and compare them to local emissions inventories. This is an interesting subject because there is a mounting body of evidence demonstrating that urban emissions inventories tend to underestimate CH4 emissions; identifying the potential missing urban sources of CH4 and/or using in situ measurements to correct emission factors is crucial for improving current inventories and planning mitigation strategies. However, in my opinion, the experimental datasets generated in this study were not used to their full potential. Great effort was put into upscaling localised emissions derived from mobile measurements to the city scale; this required 'calibrating' the mobile fluxes using EC values obtained for very different spatial and temporal scales. This must introduce substantial uncertainties, which seem almost impossible to fully quantify.

> Thank you for the insightful evaluation about significance of this study. As pointed out by the reviewer, when comparing the methods used in previous studies for mobile measurements with those of the eddy covariance (EC) method, we recognize that CH_4 emissions derived from mobile measurements have several uncertainties. On the other hand, we acknowledge that EC measurements represent only a portion of the urban area. Therefore, to estimate the city-scale CH_4 emissions, we propose that scaling based on a combination of EC measurements and mobile observations is the measurement-based approach (given the available data), albeit with inherent uncertainties. To our knowledge, there has been no publication that addresses these differences and uncertainties in detail for the benefit of the field and readers. Therefore, we have revised the manuscript to more thoroughly describe the assumptions, uncertainties, and limitations of the research methodology. We explain the individual details in the following responses.

More specifically, the design of the study could have been improved. A second mobile measurement *campaign in a contrasting season would have been interesting since the EC time series demonstrates that there is a strong seasonality in emissions. The authors acknowledge that the chosen routes for the bicycle and car measurements might have biased the fluxes because of plausible differences in sources. This could have been tested by conducting a sampling experiment along the same routes with both modes of transport. Overall, there are many caveats, unknowns and sources of uncertainties associated with the mobile measurements, whilst the EC approach seem to have produced a robust time series exhibiting interesting temporal patterns. More could have been done with the EC data; e.g. the spatial distribution of EC fluxes was not even mentioned.*

We appreciate the reviewer's criticism. We accept that the value that further study would provide and make recommendations for others that build in supplementary measurements that would close the circle with regards to further making the conclusions more robust. This study presents our first phase extensive mobile campaign with a main focus on identifying CH_4 source sectors within the EC footprint and neighboring areas. We agree with the reviewer that it would be beneficial to conduct additional measurement in different seasons, and we acknowledge the limitation of analysis using the currently available data. However, we are sure the reviewer will understand that the project budget was not endless. Better quantification of CH_4 emission magnitude will be examined should additional data collection become available. To answer the reviewer's concern, we have made following revisions.

To investigate the spatial variability of CH_4 fluxes obtained from the EC method, we have added a radar chart in Fig. 9, illustrating the CH_4 fluxes during daytime in both summer and winter. To explain the new results, we have included the following sentences in Section 3.5, Lines 464-471: " Although nighttime CH_4 fluxes were consistent across seasons with an average of 26 nmol m^{-2} s^{-1}, daytime fluxes were lower in spring compared to other seasons (Fig. 8). Wind sector analysis for daytime fluxes revealed higher CH_4 emissions in the WSW, N, and NNW sectors during summer (Fig. 9a), as found by a previous study conducted in 2019 (Takano and Ueyama, 2021). These elevated emissions may be attributed to anthropogenic sources, such as high industrial-commercial development and extensive major road networks (Ueyama and Ando, 2016; Okamura et al., 2024), and presumably to sewage treatment plants in these wind sectors (Takano and Ueyama, 2021). A similar pattern of elevated daytime CH_4 fluxes in the same three wind sectors was observed in winter, although the magnitudes were lower than those in summer (Fig. 9b). "

We have also added a discussion of the wind sector analysis in Lines 642-650: "Daytime CH_4 fluxes measured by the eddy covariance method varied with wind direction (Fig. 9). Elevated fluxes from the NNW and WSW directions showed a pattern consistent with observations in 2019 (Takano and Ueyama, 2021), suggesting a persistent trend at this site. Westerly winds, which are prevalent during the day due to sea breeze, were coincident with elevated CH_4 fluxes. To the west of the tower site lie commercial and industrial zones with major roads, which have previously been linked to increased CO_2 fluxes (Ueyama and Ando, 2016) and NO_2 fluxes (Okamura et al., 2024) under similar wind conditions. This pattern suggests that CH_4 emissions may originate from such urban landscape. The wastewater treatment facilities are located near the outer edge of the source areas contributing 80% of the turbulent fluxes. Therefore, they may also contribute to the observed CH_4 emissions, and fluxes from the northwest and southwest may be influenced by plumes emitted from these facilities (Takano and Ueyama, 2021)."

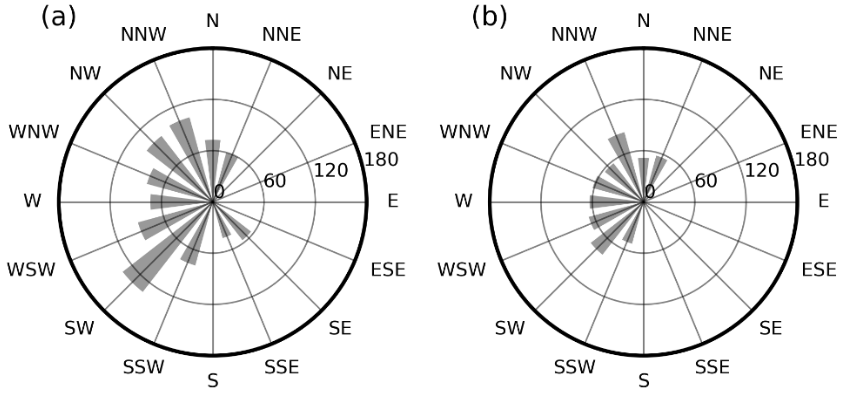
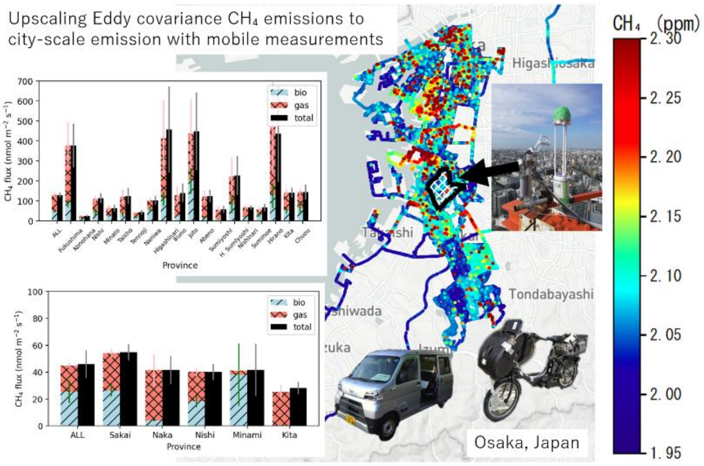


Fig. 9. Stacked charts of half-hourly CH_4 fluxes (nmol m^{-2} s^{-1}) during the daytime (09:00–17:00) for the 22.5° wind sector in summer (a; June–September) and winter (b; December–March) of 2023.

It is no surprise that EC and street level mobile fluxes cannot be reconciled since the spatial scales and micrometeorological conditions are vastly different for the 2 approaches. Fundamentally, trying for/forcing a reconciliation feels like shoehorning. Why not acknowledge that the 2 approaches yield very different estimates (for obvious reasons), shorten the discussion on this and emphasise instead the unique information that mobile measurements can provide and that EC cannot? For example, the findings about increased emissions near restaurants is interesting as is the hypothesis that these might be caused by the type of stoves used.

We acknowledge that EC and mobile measurement methods differ in spatial representativeness, and thus differences between the two were expected. As noted by the reviewer, EC provided more robust estimates within its flux footprint, but, as shown in the new Figure 1b, it represented only a portion of the city area in Sakai or Osaka Metropolitan area. To address this limitation, we combined mobile measurements as a proxy to spatially scale EC-derived fluxes to areas not directly covered by EC. In other words, we used the mobile detections as a proxy to scale the total and sector emissions across the parts of the cities not covered by EC measurements. This intent was not clearly conveyed in the initial manuscript.

To clarify our approach, we have revised the manuscript structure: the section on the EC measurements (now Section 3.4) precede the city-scale emission estimates from mobile data (now Section 3.5). Additionally, the section on point source origins ("Restaurants and sewage treatment plants") is moved earlier to Section 3.3. These changes are intended to better highlight our key methodological approach, and we believe they have improved the clarity of the manuscript. We are grateful to the reviewer for pointing out this important issue. To clarify the framework of the study, we have also added a graphical abstract.



I would like to see a rewrite where the use of the mobile flux data is mostly limited to partitioning the street-level aggregated fluxes into natural gas, biogenic etc… whilst the EC measurements are used to calculate robust seasonal and annual budgets. Combining the two outputs (fraction of fossil fuel to biogenic emissions from mobile measurements) and seasonal and annual budgets can give a more robust estimate of the magnitude of emissions segregated by source, and improve the comparison with values from emissions inventories.

As answered to the above comments, the EC method alone can only evaluate CH_4 emissions in a partial area within Osaka, specifically in Sakai City, Sakai Ward. While the EC method provides robust results for the target area, it cannot be directly applied to assessments of the entire city area, which may only be achieved via broad assumptions with associated uncertainties. One such assumption is to use the leak indications and source attribution from the mobile measurements as a proxy (rather than a direct measurement) for the total methane emissions from each ward. In this study, the first step was to assess the relationship between EC estimates and mobile measurements for the same areas as the EC footprint, and then use this relationship (by applying a scale factor) to extend the evaluation to Sakai City and Osaka City. Although this approach carries uncertainties, using mobile leak indications as a proxy for area CH_4 emissions is an observation-based approach in contrast to other possibilities relying on statistics such as population or number of households. Considering that such regional evaluations have not been examined in Japan to date, this approach serves as an important start point to advance research on urban methane emissions in Japan. As the reviewer pointed out, we agree that a more detailed discussion of the EC results is necessary, and we have added relevant discussion accordingly.

The new panel (b) in Fig. 1 now shows zoomed view including the limited area (relative to the total size of Sakai and Osaka cities) covered by the EC footprint, which highlights why an extrapolation approach is needed (in this case based on mobile measurements) to estimate whole-city emissions.

Specific comments

Equations 1-4: change the asterisk (*) to a multiplier sign (×) or omit altogether.

Based on the reviewer’s suggestion, we have revised the equations accordingly.

Lines 211-212: “The local background concentration was defined as the 5th percentile value during *a 5-minute moving window". How were these limits/criteria set?*

> We calculated the baseline concentration based on previous studies and our own examination of variety of criteria settings. We have added the following sentences in Lines 224-227: "Previous studies used median values from the length of the moving window, ±2.5 minutes (Maazallahi et al., 2020; Weller et al., 2019), but we have used the 5th percentile as a baseline, a compromise solution based on the methods described in Dowd et al., (2024) and Tettenborn et al., (2025). As the approximate baseline was 2.0 ppm CH_4 throughout the campaigns, the choice of baseline metric is not anticipated to have material impact on the overall results. "

Line 220: "spatial resolution of 12 s".

How was this aggregation threshold obtained?
Consider changing the sentence to "a spatial resolution equivalent to 12 seconds of travel time (approximately 37 m)."
I don't understand the leak detection procedure and the data aggregation. I read the first sentence as meaning that the maximum enhancement within the 12 s block is used, but the next sentence states that multiple data points are available within each 12 s time chunk.

> We apologize for the error in this sentence, which may have caused confusion. The information about the unit in meters was incorrect, so we have revised the value and provided an explanation for why this value was used. We have added a sentence to clarify this in Lines 234–238, explaining the spatial resolution of the aggregated emission sources: "The identified CH_4 enhancements were aggregated at a spatial resolution of 12 seconds in both latitude and longitude (approximately 471 m in longitude and 372 m in latitude) by obtaining the maximum CH_4 enhancement (hereafter referred to as a leak indication, LI) to avoid double counting of single LI. This spatial resolution was determined based on measurements showing that detected plumes sometimes extended over more than 200 m, and that the majority of distances between neighbouring LIs exceeded this value.".

Lines 229 – 231: "We found that it underestimated the regional CH4 fluxes in the empirical model, as noted below. Consequently, the use of units in the emission rate might be misleading; therefore, we used the unit of CH4 enhancement (i.e., ppm) but a definition consistent with that used in previous studies."

What underestimates the regional CH4? Weller's empirical model? How was the regional flux calculated?
Why is the use of units misleading?
What definition of enhancement was used? What studies is it consistent with?
Additionally, these comments read like results and should therefore not be in the methods section.

> Previous studies classified the magnitude of LIs based on CH_4 emissions estimated using the Weller empirical equation, rather than simply reporting increased CH_4 concentrations. However, we found that CH_4 emissions estimated by the Weller model involve substantial uncertainties; therefore, we chose to report LIs in units of ppm. Based on the reviewer's suggestion, we have added the emission rate (unit in L min^{-1}) in addition to CH_4 enhancements (unit in ppm) in the revised manuscript. We have revised the sentences to address the reviewer's comment in Lines 246–250: "CH_4 emission estimates derived from LIs using the empirical model by Weller et al. (2019) showed considerable uncertainty in Osaka, as discussed below. Because the emission rates calculated for individual LIs may carry large uncertainties, expressing these values in standard emission units could be misleading. Therefore, we presented CH_4 enhancement relative to the baseline concentration (in ppm) for each emission category in addition to the emission (in L min^{-1}), ensuring consistency with the definitions used in previous studies.".

The CH_4 enhancement was defined in Lines 221-222: “the CH_4 concentration was elevated by more than 0.1 ppm with respect to the local baseline concentration (hereafter referred to as CH_4 enhancement; ΔCH_4)”.

Based on the reviewer’s comments, the determination of the *A* value using the eddy covariance method, which was previously described in the Methods section, has been moved to the Results paragraph in Lines 491-504 with slight modifications.

Lines 233 – 235: so you forced the estimated CH4 fluxes obtained from the mobile measurements to match the EC values by applying a correction factor. This seems problematic because of the difference in spatial scales, source distribution, intensity etc… Essentially, the assumption is that street-level spot measurements must be identical to EC values from a larger, spatially-integrated flux footprint. This is a dubious assumption and I doubt that it is defensible.

We agree with the reviewer’s concern, and this is a key point that we emphasized in this study. Applying certain calibrations to empirical models (e.g., Weller et al., 2019) appears to be the most reasonable approach given the current limitations. We have carefully addressed the assumptions and associated uncertainties in the Methods, Discussion, and Conclusion sections. This additional information may help readers understand that current estimates based on empirical models—including both this study and previous works—contain substantial uncertainties.

In the initial manuscript, we described the use of EC results to “calibrate” the mobile measurement data. However, we acknowledge that this term may not be appropriate in this context, and have therefore revised the wording to “scale” instead. This change was made to clarify that the mobile measurements were used to extend the EC-based estimates to a broader area. Additionally, as mentioned in our response above, we have restructured the relevant section in the manuscript to improve clarity on this point. We have added further explanation regarding the assumptions and uncertainties associated with this scaling approach in the following sections.

In the method section, we have added the sentences in Lines 262-270: “To estimate regional CH_4 emissions on the basis of mobile measurements, we applied scaling EC-derived daytime CH_4 fluxes to the city-scale fluxes using the mobile measurements and an empirical equation (Weller et al., 2019), extending the fluxes beyond the EC footprint to estimate CH_4 emissions at the city scale. This scaling method clarified the relationship between the results from empirical models and the regional CH_4 fluxes observed using the EC method, with the goal of spatial extrapolation. Since CH_4 fluxes measured by the EC method reflect both street-level emissions and sources located on building rooftops, walls, and other vertical surfaces, the scale factor may capture the relationship between street-level emissions and vertically integrated emissions at the city scale. This scaling method also quantified the differences between mobile and EC measurements, determining the extent to which ground-based mobile measurements deviate from the spatially representative EC results.”, and Lines 290-298: “This approach to estimating regional CH_4 emissions is widely adopted (Vogel et al., 2024; Weller et al., 2019) with the assumption that the spatial distribution of CH_4 emissions obtained from mobile measurements is representative of the emission patterns across the entire study area. This study evaluated the assumption by comparing upscaled CH_4 emissions derived from bicycle-based measurements (complete spatial coverage) with vehicle-based measurements (coverage only a portion of the area but are assumed to reflect the broader spatial characteristics). The total flux calculation was also performed for all LIs (not categorized). CH_4 fluxes upscaled by the two estimates (with and without source attributions) were compared to understand the uncertainties associated with the procedure. Note that the correction factor, *A*, close to 1 indicates low uncertainty, whereas a deviation from 1 suggests a larger correction with greater uncertainty in the estimates derived from mobile measurements.".

In the discussion section, we have added the sentences in Line 732-739: "The regional flux estimates based on unscaled empirical models were considerably underestimated compared to those derived from models where the flux totals are scaled to match eddy covariance fluxes, highlighting the substantial impact of scale factor adjustment on the results. While the scale factor adjustment provides the best available estimate at present, its validity cannot be fully verified due to the lack of independent evaluation data. Smaller estimates derived from unscaled empirical models may suggest the significance of CH_4 emissions originating from rooftop sources, such as exhaust vents, smokestacks, and gas-powered air conditioners located on building rooftops (Stichaner et al., 2024). Another potential explanation could be the cumulative effect of numerous small sources that went undetected in the street-level measurements.".

In the conclusion, we have added the sentences in Lines 779-781: "This highlights that mobile measurements at near ground level may be considerably underestimating city-scale CH_4 emissions, highlighting that a large underestimation may be common to other city surveys.", and Lines 785-793: "Simultaneous EC and mobile measurements could enable the quantification of uncertainties in CH_4 emissions derived from mobile surveys, facilitate the development of improved scaling techniques beyond those used in this study, and support the identification of emission hotspots and source attributions. Simultaneous measurements are also useful for evaluating CH_4 emissions outside the EC footprint, as they allow for comparison with the emission inventory provided by the local government. Future studies should revisit improvements in empirical estimation methods based on other top-down methods, such as atmospheric or EC measurements. Further understanding the reasons for the discrepancy between the top-down EC measurements and mobile measurements is key to understanding the mitigation potential for urban methane in Japan. Continuous measurements of turbulent fluxes and atmospheric concentrations of CH_4 and C_2H_6 could enhance our understanding of urban CH_4 emissions, their attributions, and their temporal dynamics.".

Line 244: why not use data from the LI-7810 measured at 1.85 above street level?

We have added a sentence explaining why data from the Mira Ultra were used instead of the LI-78100 in Lines 258-260: "the tendency that CH_4 enhancements measured by Mira Ultra (0.5-m height) are larger than those by LI-7810 (1.85-m height) (see section 3.1). Hereafter, the CH_4 measurements refer to those by Mira Ultra.".

Equation 6:

A: correction factor. Why not use the EC fluxes directly since you're forcing the spot measurements to match the EC values? This correction factor bothers me because the assumption seems to be that most emissions of CH4 are from roads (whether it be from leaks in the gas distribution network or else), but what about buildings etc…? If I'm barking up the wrong tree, please correct me and rewrite the relevant part of the methods section to make the approach more explicit.
Emmean: what area was Em averaged over?
Multiplication by total road length within a ward: the underlying assumption is that the hotspots density and intensity is uniformly distributed along all roads. Is this a reasonable assumption?

Since the EC measurements represent only a limited area within Sakai City (specifically Sakai Ward), they cannot be used to evaluate CH_4 emissions for the entirety of Sakai City or Osaka City. Therefore, in this study, we scaled the EC-based flux to the entire city using the mobile measurements as a proxy, ensuring full spatial coverage of the streets within the EC footprint. The upscaled mobile measurements were then used to evaluate CH_4 emissions outside the EC footprint. As the emission inventory provided by the local government is only available at the city level, comparisons with the inventory can only be made at the city-wide scale. Em was calculated for each ward or city, and an explanation has been added in Line 282: 'Em_{mean} represents the areal mean of *Em* within each ward or city.'. To investigate the spatial variability of CH_4 fluxes obtained from the EC method, we have added a radar chart to Figure 9, illustrating the CH_4 fluxes during daytime in both summer and winter. The upscaling assumes that emission characteristics are spatially similar across the city, an assumption commonly made in studies utilizing mobile measurements (e.g., Vogel et al., 2024).

Lines 282 - 283: "We estimated the upscaled CH4 fluxes via Eqs. 5, 7, 8, and 9 and considered the range of upscaled fluxes as an uncertainty." Does this mean that estimates using Eq.6 were not used after all? If that is the case, there is no point in discussing Eq.6 at all.

Eqs. 5, 7, 8, and 9 were used to estimate Em, which was then used to calculate regional fluxes in Eq. 6. We have revised the sentence in Lines 503–504 accordingly: "We estimated the upscaled CH_4 fluxes using Eq. 6 with different values of *Em* derived from Eqs. 5, 7, 8, and 9".

Line 295: another correction factor (0.64). I assume that this is adjust the mobile flux to the average diel cycle of the EC measurement, but it isn't clear from the text how it was obtained. What time period was covered? Was the correction applied to the upscaled fluxes at ward or city level? I am concerned about the mounting number of simplifying assumptions, correction factors, etc… and the usefulness of the end estimates given that uncertainties are probably extremely large and difficult to quantify.

The assumptions and uncertainties associated with the parameter for diurnal variations have been added in Lines 321–326: "To consider the diurnal variability, we multiplied the upscaled fluxes by 0.64, which was the ratio of the daily mean to the daytime mean flux (9:00–17:00) in 2023, when calculating the daily fluxes. Based on the locations of emission point sources identified by mobile measurements (discussed below), CH_4 emissions across the study area were likely to exhibit diurnal variation. Although extrapolating the diurnal correction factor (0.64) introduces some uncertainty in regional CH_4 emissions, we applied the correction to avoid overestimations due to the absence of nighttime measurements.".

Lines 297 -299: "The annual emissions were only calculated for the vehicle measurements because the bicycle measurements were only conducted for the city center and residential areas and might underrepresent the rural areas." This sentence seems to contradict the statement at line 262 "To determine the A factor, we used bicycle measurements […]". The A correction factor forces fluxes from street-level spot measurements to match EC values, which arise from all sources of CH4 within the EC flux footprint, road and non-road. In this context, it would seem more logical to derive the A correction factor from the car measurements since a more varied landscape was sampled.

As mentioned in the above reply, this study used bicycle-based measurements—which fully cover all streets within the EC flux footprint—to scale mobile CH_4 measurements with the EC data. Subsequently, vehicle-based measurements were used to evaluate CH_4 emissions outside the EC footprint, specifically for the entire Sakai City and Osaka City. At present, the total CH_4 emissions estimated through upscaling from bicycle-based and vehicle-based measurements are generally consistent (Fig. 10). However, since the bicycle measurements cover all roads within the EC footprint, unlike the vehicle measurements which only cover a subset, we believe that using the bicycle data to determine the correction factor is more logically justified.

Lines 322 -329: This section is out of place. It would make sense to move it to the end of the sentence at line 242 since the justification of why the 0.5-m height measurements was favoured.

According to the reviewer's suggestion, the paragraph has been moved earlier in the section (Lines 252–260).

Fig.3: The red dots are the data for which the correlation coefficient > 0.7, the blue dots <= 0.7; so what are the grey dots?

We have added the following sentence to the caption (Lines 378–379): "Grey dots represent data that were not classified as leak indications because of the CH_4 enhancements smaller than the criteria (0.1 ppm).".

Fig.6-8: consider a different colour scheme because red, blue and green are difficult to distinguish from one another for some people.

Following the reviewer's suggestion, I have changed the color to a lighter shade and applied different hatch patterns to it. Thank you for pointing out the important details.

Line 505: The agreement between EC and upscaled mobile measurements is perhaps a little misleading and circular because the upscaling was forced to match the EC data. I think that the authors are trying to highlight that mean annual EC values are close to the upscaled mobile data obtained for 2 months of the year only.

We agree with the reviewer's comment. We acknowledge that the term "agreement" may have been misleading, and therefore we have removed the corresponding sentence from the manuscript. As explained above, we have also restructured the manuscript so that the results from the EC measurements have been presented prior to the discussion on upscaling.

Line 585: In my opinion, this conclusion is somewhat overreaching because despite the agreement between car- and bicycle-borne measurements, there is a demonstrated discrepancy between the street-level fluxes and the EC fluxes. So, in terms of estimating urban emissions the mobile measurements are limited by the fact that they require calibration using EC measurements, and that they only capture temporal snapshots. In fact, calculating the emission budgets from the EC measurements comes with fewer uncertainties than using mobile measurements. However, the strength of the mobile measurements lies in the ability to identify hotspots and in source attribution. The authors should tone done the claims of usefulness in terms of estimating emissions at higher temporal and spatial scales.

We agree to the reviewer's concern. We have added the following sentence in the conclusion to highlight the limitation in Lines 785-789: "Simultaneous EC and mobile measurements could enable the quantification of uncertainties in CH_4 emissions derived from mobile surveys, facilitate the development of improved scaling techniques beyond those used in this study, and support the identification of emission hotspots and source attributions. Simultaneous measurements are also useful for evaluating CH_4 emissions outside the EC footprint, as they allow for comparison with the emission inventory provided by the local government.".

Line 647: what "21 uncertainties"? Do you mean sources of uncertainties? If so, name a few.

The unclear sentence has been revised in Lines 779-781: "This highlights that mobile measurements at near ground level may be considerably underestimating city-scale CH_4 emissions, highlighting that a large underestimation may be common to other city surveys.". The value "21" was originally included to indicate the *A* factor, referring to the discrepancy between the results from the EC measurements and the mobile measurements. However, we consider that presenting this as an uncertainty may be misleading. Therefore, we have revised the manuscript to remove the specific value and instead describe the discrepancy qualitatively.

Line 688: refrain from using a non-standard acronym (LI, in this instance) in the conclusion as some readers might skim the paper and only read the abstract and the conclusion initially.

According to the reviewer’s suggestion, we have added 'leak indications (LIs)' in Line 782.

References

Vogel, F., Ars, S., Wunch, D., Lavoie, J., Gillespie, L., Maazallahi, Röckmann, T., Nęcki, J., Bartyzel, J., Jagoda, P., Lowry, D., France, J., Fernandez, J., Bakkaloglu, S., Fisher, R., Lanoiselle, M., Chen, H., Oudshoorn, M., Yver-Kwok, C., Defratyka, S., Morgui, J. A., Estruch, C., Curcoll, R., Grossi, C., Chen, J., Dietrich, F., Forstmaier, A., Denier van der Gon, H. A. C., Dellaert, S. N. C., Salo, J., Corbu, M., Iancu, S. S., Tudor, A. S., Scarlat, A. I., and Calcan, A.: Ground-Based Mobile Measurements to Track Urban Methane Emissions from Natural Gas in 12 Cities across Eight Countries. Environmental Science & Technology, 58, 2271-2281, https://pubs.acs.org/doi/10.1021/acs.est.3c03160, 2024.

Responses to reviewer's comments. (Reviewer 3)

Ueyama, et al.

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The paper by Ueyama et al. describes eddy covariance and mobile observations of methane and ethane in the Osaka metropolitan area in Japan. It covers an important and relevant research topic. The dataset is quite detailed and unique in that the authors try to combine different datasets from eddy covariance to mobile measurements. Regarding the interpretation of their results, I have a couple comments and questions. I recommend publication after consideration of my comments as outlined below.

Thank you for your kind encouragement and evaluation of the importance of this study.

Major comments:

Section 2.5: the proposed method of Weller to infer fluxes from road-side observations is fundamentally challenging and uncertain, because it assumes that methane is emitted at street level (e.g. leaks below ground), for which the method has been designed. However there is growing evidence (see also Stichaner et al., 2024, doi: 10.1016/j.atmosenv.2024.120743), that methane emissions are largely emitted via standard operation procedures (e.g. pre-flush or partially burned natural gas) and then emitted at roof-top level through smoke stacks. Methane sources are therefore not solely distributed horizontally, but also exhibit a strong vertical gradient within the urban roughness layer. This could easily bias the approach of Weller by a factor 2-3 and therefore the approach can be qualitative at best. One would not expect to see a good correspondence with EC observations conducted above the urban roughness layer. The EC observations in this study also provide indirect evidence that methane is linked to natural gas consumption (e.g. diurnal cycle) rather than leaks, as other cited studies have shown. (e.g. Pawlaw and Fortuniak, 2016, Helfter et al. 2016)

We acknowledge the uncertainties associated with our methodology and have accordingly clarified the assumptions and limitations in the revised manuscript. Since EC measurements can only represent a limited portion of the city, mobile measurements remain one of the few viable approaches for estimating city-wide CH_4 emissions, despite the associated uncertainties. To address this limitation, we combined mobile measurements as a proxy to spatially scale EC-derived fluxes to areas not directly covered by EC. In other words, we used the mobile detections as a proxy to scale the total and sector emissions across the parts of the cities not covered by EC measurements. To acknowlege these concerns, we have added the following sentences to the Methods section (Lines 262–270): "To estimate regional CH_4 emissions on the basis of mobile measurements, we applied scaling EC-derived daytime CH_4 fluxes to the city-scale fluxes using the mobile measurements and an empirical equation (Weller et al., 2019), extending the fluxes beyond the EC footprint to estimate CH_4 emissions at the city scale. This scaling method clarified the relationship between the results from empirical models and the regional CH_4 fluxes observed using the EC method, with the goal of spatial extrapolation. Since CH_4 fluxes measured by the EC method reflect both street-level emissions and sources located on building rooftops, walls, and other vertical surfaces, the scale factor may capture the relationship between street-level emissions and vertically integrated emissions at the city scale. This scaling method also quantified the differences between mobile and EC measurements, determining the extent to which ground-based mobile measurements deviate from the spatially representative EC results."

Furthermore, we have addressed the vertical distribution of emissions and the associated uncertainties in the Discussion section (Lines 732–739): "The regional flux estimates based on unscaled empirical models were considerably underestimated compared to those derived from models where the flux totals are scaled to match eddy covariance fluxes, highlighting the substantial impact of scale factor adjustment on the results. While the scale factor adjustment provides the best available estimate at present, its validity cannot be fully verified due to the lack of independent evaluation data. Smaller estimates derived from unscaled empirical models may suggest the significance of CH_4 emissions originating from rooftop sources, such as exhaust vents, smokestacks, and gas-powered air conditioners located on building rooftops (Stichaner et al., 2024). Another potential explanation could be the cumulative effect of numerous small sources that went undetected in the street-level measurements."

We also emphasize this point in the Conclusion section (Lines 779–781): "This highlights that mobile measurements at near ground level may be considerably underestimating city-scale CH_4 emissions, highlighting that a large underestimation may be common to other city surveys."

Additionally, we highlight the importance of conducting simultaneous EC and mobile measurements (Lines 785–789): "Simultaneous EC and mobile measurements could enable the quantification of uncertainties in CH_4 emissions derived from mobile surveys, facilitate the development of improved scaling techniques beyond those used in this study, and support the identification of emission hotspots and source attributions. Simultaneous measurements are also useful for evaluating CH_4 emissions outside the EC footprint, as they allow for comparison with the emission inventory provided by the local government."

We hope these revisions help readers better understand the estimated CH_4 emissions along with the methodological uncertainties involved.

Section 3.4: why would sewage treatment plants be a big methane source? They are typically run aerobically, and methane emissions should be very low for well managed plants.

Sewage treatment plants in the study area use the activated sludge process for wastewater treatment. However, as part of the treatment process, many facilities also employ anaerobic digestion, particularly for sludge treatment, which is known to emit substantial amounts of CH_4 in Japan and other countries. We have added the following sentences to clarify this point in Lines 679–683: "Sewage treatment plants in Japanese megacities—including all plants examined in this study—primarily employ the activated sludge process for wastewater treatment. In addition, these plants typically use anaerobic digestion for sludge treatment, a process that is known to emit more CH_4 than systems without anaerobic digestion (Song et al., 2023). Experiments conducted at sewage treatment plants have shown that significant amounts of CH_4 are emitted during the sludge dewatering process and from storage tanks containing digested sludge (Oshita et al., 2014)."

Section 3.5: Diurnal variations and weekend to weekday variations suggest sources other than leaks, which would be independent of activity (ie. gas usage) as gas distribution networks are typically maintaining a constant pressure and leak related emissions therefore are largely independent from temperature and activity changes.

We agree with this suggestion. As discussed in Lines 623-624, we note that "A small nighttime CH_4 flux could suggest that steady gas leaks are minimal in Sakai, as CH_4 emissions would be high at night if steady gas leaks were more significant.". We have slightly modified this sentence for the clarifications.

Fig. 10: The authors only state that there is a seasonal cycle with two peaks, which is quite unusual (all other sites with multi-year direct EC CH4 observations show a clear seasonal cycle with only one peak). What is the cause of this – can the authors provide a reasoning? The only explanation I can think of is drastic seasonal changes in the flux footprints, which the authors have not analysed to the extent necessary to understand diurnal and seasonal variations of EC fluxes.

Thank you for raising this important point. As reported in our previous studies (Ueyama and Ando, 2016; Ueyama and Takano, 2022), CO_2 fluxes also exhibited double peaks in summer and winter, which were linked to increased gas demand for air conditioning. We have added a paragraph to explain the potential reasons for the seasonal variations in CH_4 flux in Lines 629-640: "The two seasonal peaks observed in summer and winter (Fig. 8) can be attributed to increased gas demand associated with high and low temperatures, respectively. Within the study area, gas demand rose in summer when daily air temperatures exceeded 20°C, and similarly, in winter, demand increased as daily temperatures dropped. Natural gas consumption increased during both summer and winter, with summer usage being higher than winter usage, as observed at both the study site (Fig. S9 in Ueyama and Takano, 2022) and at university buildings in Sakai (Fig. A3 in Ueyama and Ando, 2016). Consequently, the CO_2 flux measured at this site exhibited a similar seasonal pattern, characterized by two distinct peaks (Ueyama and Ando, 2016; Ueyama and Takano, 2022). This gas demand could contribute to the seasonal variation in CH_4 flux. Other possible CH_4 sources in the summer include biogenic sources, such as sewage manholes and ditches surrounding ancient tombs (Takano and Ueyama, 2021). Detailed flux footprint analysis suggested that daytime CH_4 fluxes were influenced by a sewage treatment plant located in the bay area (Takano and Ueyama, 2021). Since the diurnal cycle of the dominant wind direction was controlled by land–sea breezes throughout the year (Fig. A1 in Ueyama and Ando, 2016), seasonal differences in the flux footprint likely had a limited influence on the seasonal variations in CH_4 flux."

As the dominant wind directions are fairly consistent at this site, with land–sea breezes being a typical wind pattern, we have added the following sentence in Lines 158-160; "The topography around the measurement area is flat, but the plain is surrounded by mountains on three sides (north, south, and east). Therefore, land–sea breezes prevail over the area, with westerly winds during the daytime and easterly winds at night throughout the year (Ueyama and Ando, 2016)."

Generally I miss a more thorough analysis of the flux footprints to understand the bimodal seasonal cycle that has not been observed at other sites. Is there a significant sea-breeze effect? What is the diurnal variation of the flux footprint? Could seasonal synoptic changes lead to the methane peak in summer? Are flux footprints changing seasonally? Antropogenic CH4 emissions would be expected to lead to a peak in winter (more heating demand), while biogenic emissions (e.g. wetlands) would show an opposite seasonal cycle. In this context it would be interesting to investigate whether CH4 fluxes *in Sakei City are temperature dependent, and whether there is a different temperature dependence in different seasons. Perhaps this could provide additional insights in their interpretation of EC flux data. Are there wetland emissions within the flux footprint or is there evidence of a superemitter (e.g. Stichaner et al., 2024) biasing the seasonal cycle?*

We conducted a detailed footprint analysis of CH_4 flux in our previous study and found that CH_4 fluxes were higher in certain wind sectors (Takano and Ueyama, 2021). Due to the tall tower measurements (112 m above ground), the representative area is very broad as shown in Figure 1, and as a result, we were unable to identify single large emission sources using footprint analysis, as discussed in Stichaner et al. (2024). Therefore, we have added an additional wind sector analysis, which is summarized in the new Figure 9. To explain the results, we have included the following sentences in Section 3.5, Lines 564-571: " Although nighttime CH_4 fluxes were consistent across seasons with an average of 26 nmol m^{-2} s^{-1}, daytime fluxes were lower in spring compared to other seasons (Fig. 8). Wind sector analysis for daytime fluxes revealed higher CH_4 emissions in the WSW, N, and NNW sectors during summer (Fig. 9a), as found by a previous study conducted in 2019 (Takano and Ueyama, 2021). These elevated emissions may be attributed to anthropogenic sources, such as high industrial-commercial development and extensive major road networks (Ueyama and Ando, 2016; Okamura et al., 2024), and presumably to sewage treatment plants in these wind sectors (Takano and Ueyama, 2021). A similar pattern of elevated daytime CH_4 fluxes in the same three wind sectors was observed in winter, although the magnitudes were lower than those in summer (Fig. 9b)."

We have also added a discussion of the wind sector analysis in Lines 642-650: " Daytime CH_4 fluxes measured by the eddy covariance method varied with wind direction (Fig. 9). Elevated fluxes from the NNW and WSW directions showed a pattern consistent with observations in 2019 (Takano and Ueyama, 2021), suggesting a persistent trend at this site. Westerly winds, which are prevalent during the day due to sea breeze, were coincident with elevated CH_4 fluxes. To the west of the tower site lie commercial and industrial zones with major roads, which have previously been linked to increased CO_2 fluxes (Ueyama and Ando, 2016) and NO_2 fluxes (Okamura et al., 2024) under similar wind conditions. This pattern suggests that CH_4 emissions may originate from such urban landscape. The wastewater treatment facilities are located near the outer edge of the source areas contributing 80% of the turbulent fluxes. Therefore, they may also contribute to the observed CH_4 emissions, and fluxes from the northwest and southwest may be influenced by plumes emitted from these facilities (Takano and Ueyama, 2021)."

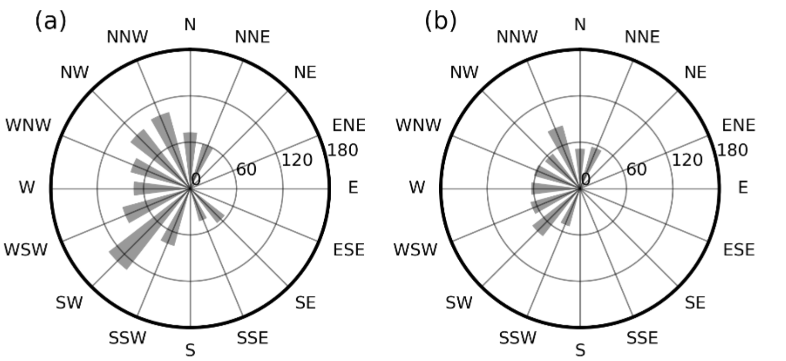

Fig. 8. Stacked charts of half-hourly CH_4 fluxes (nmol m^{-2} s^{-1}) during the daytime (09:00–17:00) for the 22.5° wind sector in summer (a; June–September) and winter (b; December–March) of 2023.

Line 593: Why would sewage treatment plants be such a big source of methane? They are typically *operated under aerobic conditions and methane emissions from such plants have shown to be comparatively small. This is actually in line with the street level observations suggesting that the sewage treatment plant is not a big methane emitter, which would be expected for a well operated facility.*

As mentioned in the previous comment, we have added a sentence explaining the reason in Lines 679–683.

Minor comments:

Line 58 **-** *565: it is Helfter and not Helfer – this typo needs to be corrected throughout the manuscript*

We have corrected all typographical errors throughout the manuscript.

Line 56, "The EC method has been used for measuring fluxes over terrestrial ecosystems (Baldocchi, 2014) but has been applied in urban areas to understand decade long greenhouse gas emissions. " change 'but' to 'and'

We have change to use "and" in Line 61.