

***Interactive comment on* “Methane mapping, emission quantification and attribution in two European cities; Utrecht, NL and Hamburg, DE” by Hossein Maazallahi et al.**

Anonymous Referee #1

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This study presents an extensive set of vehicle-based measurements of methane and other compounds to investigate methane emissions in Utrecht, NL and Hamburg, DE. The authors used empirical equation developed von Fischer et al. (2017) and updated by Weller et al. (2019) to estimate the emissions from the natural gas distribution network using the methane enhancements they observed during their surveys. They also tested several approaches to determine the origin of these enhancements (biogenic, thermogenic, pyrogenic) such as the isotopic signature of methane, C2/C1 ratio or CH4/CO2 ratio. Finally they used a Gaussian dispersion model to estimate methane emissions from larger sources.

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Overall, despite the large ranges and uncertainties presented here, the study is a valuable contribution to the literature as the state of knowledge of urban methane is not very advanced compared to other pollutants. The measurement campaigns seems carefully done and well designed, I appreciated the authors described their interactions with the local distribution companies and showed how their work helped reducing the emissions. It was also nice to find a list of all the acronyms in the supporting information as there are so many of them in the text and I was a bit lost at first. I recommend publishing it after addressing few minor points (which are also mentioned in the detailed comments):

1) Structure: I would reorganize section 2 a bit and group all the source attribution approaches together in one subsection instead of having two subsections about isotopic analysis and information about the ratios scattered throughout the rest of the subsections. It would also make it easier for the reader if there was a table in this section summarizing these approaches and the limits used to attribute the emissions.

2) I would have liked more discussions about the uncertainty on the emissions estimated with the approach developed by Weller et al. (2019). I understand that this method is the reference for mobile surveys at the moment but the fit of the calibration curve presented in figure 4 of Weller et al. (2019) makes me wonder about the uncertainties associated with these estimates. Also, the authors used this empirical equation to estimate emissions from microbial and combustion sources whereas it was originally designed to estimate emissions from NGDN. While the classification into small, medium and large LIs depending on the maximum amplitude of the enhancement remains correct, I am not very comfortable using the empirical equation to estimate the emissions of these other sources. Biogenic emissions are very different from NGDN emissions, they are way more sensitive to atmospheric conditions (especially temperature) and are likely to vary in time unlike NGDN emissions which should be more constant. They could also potentially be located further away than the usual roadside emissions. Figure S16 examples illustrate this: microbial emissions from the

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water body are likely localized further away and the sewage system seem to emit at a higher level than the road level.

3) The presentation of the GPDM approach used to quantify the emissions from larger facilities should be reworked and expanded a bit. For examples, the authors should talk about the wind data they are using as it is a critical parameter in this approach. Did they adjust the wind direction so that the maxima of the observed plume is aligned with the maxima of the modeled plume, etc. . . The part about the selection of the sigma y and sigma z is also not very clear. The author should also specify here which observation they are fitting with the model (the measured concentrations? One plume at a time or all the plumes measured during all the surveys?)

Details: L18-19 and L30: Should be consistent with number notation (whether letters or numbers).

L22: This should be phrased differently, the largest emission rate in Utrecht is actually coming from the wastewater treatment plant.

L64: Typo? “high precision” is written twice in a row.

L76: Typo? A comma is missing after “(Giolo et al., 2012; Helfter et al.,2016)”.

L78-80: This sentence is too vague, most methods quantify emissions methane enhancements! The authors should specify which approach they used and which type of sources they used it for.

L83: Typo? Should it be “across the urban areas in these two cities” (rather than “across the urban areas is these two cities”)?

L93: Why specify the time needed to flush the cell for the G2301 but not for the G4302? Could you add a sentence about how the methane enhancements measured by the two instruments compare? This discussion is actually in the SI, the authors could add a sentence to refer it.

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L122: What do the level 2 and 3 roads correspond to?

L137: The authors should remove “at the following links: Utrecht and Hamburg”, the citation “(Maazahalli et al., 2020b)” is enough.

Section 2.4: I would merge sections 2.4 and 2.6.1 into a source attribution section that details the multiple approaches used in this study. I would incorporate in this section a table summarizing the different ratios/isotopic measurements and the ranges used to distinguish between fossil, combustion, microbial and unclassified sources.

L168: Did the authors only take samples for isotopic analysis in Hamburg? Why not in Utrecht?

L179: How far are the measurements tower from the studied sites? Wind parameterization are large sources of uncertainty in Gaussian plume dispersion model, especially since wind close to the surface can be very different from the wind measured at 10 meters at these towers.

L184: “It has been demonstrated that the algorithm adequately estimates the majority of emissions from a city (Weller et al., 2018).” The authors should specify that this method was specially developed to quantify methane emissions from the natural gas distribution network. In this sentence, the authors seem to imply that they could estimate the emissions from any type of sources from a city.

L 192-194: How did the authors know about the mole percent of CH₄ and C₂H₆ in the NGDN in Hamburg and Utrecht? Is it based on measurements or did the NG suppliers give them this information?

L196: If I understand correctly, this whole part is used to explain how you differentiate car exhaust signals from NG signals. This is not really clear, the authors should introduce it up front to help the reader follow the organization. This could probably also be moved to the source attribution section.

L204-207: I don't understand why do the authors use different approaches to estimate

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the CH₄ and CO₂ backgrounds? This should be explain.

L229: Did the authors really need to convert decimal degrees to Cartesian coordinates in order to cluster enhancements? Doesn't it introduce additional uncertainties than directly estimating the distance between enhancements using decimal degrees?

L233: Why did the authors assigned the maximum observed enhancement to the cluster rather than a weighted average just like for the location? Wouldn't that artificially increase the emissions?

L240: The "visited at least twice" criterion in von Fischer et al. (2017) and Weller et al. (2019) was implemented to identify enhancements from the natural gas distribution network that are considered to emit continuously. I would mention that you are using another source attribution method instead.

Section 2.6.3: I was surprised that the authors did not talk about wind measurements in this section given that this is one of the biggest source of uncertainty of this technique. Maybe they should move part of section 2.5 here.

L252: What do the authors mean by "These data were evaluated using a simple point source GPDM"? What are the authors evaluating?

L252: Typo? "(" should be removed.

L265-266: The authors should be consistent with the notation: zsource (which is equal to 0 in the text) and h are to the same thing.

L276-279: This part is not very clear. Do you select sigma y and sigma z separately? Could you end up with a sigma y of a given Pasquill-Gifford stability class and combine it with a sigma z from another stability class?

L288: It would be appropriate to at least in a sentence or two explain the isotopic analysis so the reader doesn't need to go back and read these papers (which analyzer, how long were the samples measured. . .).

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L314: Typo? “Utrecht and Hamburg correspond to” rather than “Utrecht and Hamburg were correspond to”

L321: Typo? “Figure 2” looks weird.

L332: You showed previously that different types of road had very different LI rates per km depending on cities, why didn't the authors use these road-specific emission factors to upscale their emissions?

Figure 5: Typo? “of collected air samples” instead of “of air samples collected”. The authors should also show the microbial and pyrogenic clusters on these figures (L342).

L352: Typo? “combustion-related” instead of “combustion, related”.

L360: Not clear which criteria for CH₄/CO₂ ratios the authors used to classify LIs as combustion-related in the end. CH₄/CO₂ > 0.2 ppb/ppm?

Figure 6: This figure is relatively difficult to interpret, it is difficult to visualize the shape of the observed plumes when they superimposed like this. It would have been interesting to see how and where you triangulated the location of the source for this site. How many sources did you find for this site? In wastewater treatment plant, the main methane source usually correspond to the sludge treatment areas that can be spotted with Google Earth.

L375-377: The definition of the error estimate is very confusing, what are the 5 sets of measurements if there were only 3 days of measurements at the wastewater treatment plant?

L395: Typo? Extra space before “74%”.

L426: Typo? One of the “%” should be removed.

L413-432: The author should expand the discussion about the different source attribution approaches, is it necessary to use all of them? Which approach would the authors recommend to use in the future?

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L479: Shouldn't it be the "annual natural gas leakage rate per capita" rather than the "annual natural gas consumptions per capita"?

L480: Typo? "per km of pipeline" rather than "per km pipeline"?

L491: The authors already explained several times that natural gas emissions depends on the age of the pipelines and the type of material used for these pipelines. I am not sure it is useful to repeat it here, especially since it will be discussed again later (L514).

L545-549: The authors should choose one unit for the emissions and use it for all the sources, it would make easier for the reader to compare these emissions (wastewater treatment plant in t/yr, wells in kg/h. . .).

L557: Typo? "For emissions from the NGDN, the urban. . ." rather than "For emissions from the NGDN the urban. . .".

L545-557: Did the authors also looked at the ratios of these larger facilities? It could be also be an interesting information.

Supplementary information: Section 1: "Figure S2a and Figure S2b show total length. . ." rather than "In Figure S2a and Figure S2b total length. . . are shown". Same for "In Table S1 and Table S2".

Section 2.1: Typo? Should it be "CH₄-only mode, which show" (rather than "CH₄-only mode. which show"). It is indeed very strange that the higher inlet measures higher methane enhancements than the bumper inlet. Would it possible that this source was located above the ground ("chimney" emissions or like the sewer pictures showed below)?

Section 2.2: What does "the ratio of the sum of CH₄ enhancements (in ppb) to the sum of CO₂ enhancements (in ppm)" mean? Does it correspond to the area under the plume? There is no mention of Figure S7 in the text.

Section 2.4: "Errors in wind speed are estimated to be $\pm 10\%$ and for wind direction \pm

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5°” this seems low to me considering that the wind was not measured on site but at a tower located away from the site. Table 5 caption should be better isolated from Table 4, this is a bit confusing at the moment.

Section 2.6: In Figure S10a, shouldn't the authors constraint delta13C, deltaD, C2H6 and CO2 before clustering? It would avoid clustering enhancements from different types of sources. Figure S11: caption not very precise.

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