

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 #2

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1 Overview

The authors present an extensive study of ground-based mobile measurements of methane and several related tracers (C_2H_6 , CH_4/CO_2 ratio, δ^2H-CH_4 and $\delta^{13}C-CO_2$ focused on quantifying and attributing methane emissions in two European cities, namely Utrecht and Hamburg, which both rely on subterranean pipelines as the delivery system for natural gas used in the households and otherwise. Such delivery systems are known to cause leaks that contribute to the anthropogenic global warming, and it has been demonstrated previously that fixing of these gas leaks can be a very cost-effective mean of climate change mitigation. Using a combination of in situ observations (with

C1

CRDS), discrete samples collected for identified leak sources as well as Gaussian plume modelling, the authors are able to identify approximately 100 leak sources in both cities over their study period, and thanks to the robust analysis of collected data, are able to differentiate them according to emission source (natural gas distribution system / microbial sources) and the respective source strength (with just several sources responsible for large parts of total emissions). A comparison of the results against previous studies conducted in US point to potential lower specific emissions for the studied cities. The authors also attempt to upscale the measurements performed over these limited campaigns in order to compare them to the publicly available aggregated data, albeit these results should be treated with care as the dataset is limited and much more robust studies are needed to achieve this goal (which the authors accurately point out).

The authors should be commended for the impressive amount of high-quality work that was put into design, execution and data analyses during those campaigns. This is no easy task, as the study encompassed simultaneously using many state-of-the-art techniques from very different scientific fields together with a very large amount of data (both measurement and supplementary) in order to achieved the stated goals. Simultaneous analysis of several tracers and isotopic composition is of particular interest, as it shows great promise in development the methods of precise small-scale emission estimations. I find this study to be a strong contribution to the discussion in the city-scale methane emissions, and the strategy developed here seems to be promising in developing both research-targeted and operational methods for leak detection and its strength estimation.

The article does suffer however, from this wealth of data and methods, and requires multiple improvements before final publication. For example, some sections of the text require more detailed information in order for the study to be considered reproducible. Also, the treatment of uncertainty in the source estimation and Gaussian plume modelling sections should be deepened. In the second case, the sensitivity of the method to the chosen meteorological parameters should be established. In methods section

C2

some restructuring is recommended, and the Discussion could benefit from introducing a clearly defined structure in order to appropriately focus attention.

I recommend publishing the article after addressing items listed below.

2 Major comments:

1. The Method description could do with an overhaul. In some places, more details need to be provided (see 'detailed comments' section below) for the experiments to be considered reproducible. In others, some information should be combined (2.6.4. and 2.4.).
2. Discussion of uncertainties in the urban emissions is very limited, with authors stating that 'We used a Bootstrap method (Nelson, 2008) to estimate 247 emission uncertainties similar to Weller et al. (2018) for the US city studies by resampling from all recorded LIs randomly 30,000 times.' No further comment is given, and in the discussion section the authors quickly skim over this and analyse the statistics of LI, without providing information on how precise those classifications might be. The method described by Wheller et al. (and earlier by Fisher et al.) relies heavily on assumptions regarding the distance to the source and (calibration using control releases) it stands to reason that this simplistic approach must produce very large uncertainties if not supported by multiple measurement repetition. This is critically important here as the data from limited detections is interpreted and up-scaled. As it is now, it is not possible to get a realistic impression about the numbers given, and puts the resulting data analysis in question. The discussion of uncertainty and potential biases should therefore be expanded.
3. The authors state that 'Emissions from facilities show significant contributions to the total emissions in both cities.' (supplement), but many details on the method

C3

used for estimating them are missing. This section needs to be expanded and more info should be given about the analysis as well as the uncertainty estimation and sensitivity of the method to the stated assumptions. For example, the use of measurement data from distant towers in order to drive the transport model raises an eyebrow, as these are critical for calculating the emission rate. What was the average distance between the tower and the measurement location and what was the elevation of that measurement? Are the wind speed and direction uncertainties reasonable? What about the elevation of the source, which is only very briefly discussed in the supplement? In the end, the reader should have a comprehensive view on whether the method is able to provide good emission estimation in a given setting, and at the moment the result with error bars (on the order of $\pm 50\%$ $1-\sigma$) suggest it is not. This should also be discussed in more detail.

4. The discussion section would benefit from introducing subsections to provide focus for specific items under discussion.
5. I find the overall quality of language very high, yet there are multiple minor deficiencies that still need to be addressed. Below I have listed some of them. I believe that this is mostly due to heavy editing during manuscript preparation, and I ask the authors to take special care of that issue before resubmitting.
6. The supplement is large and – I'm sorry to say that – poorly edited (tables are too large - the font can safely be made much smaller; order of figures and sections does not correspond to the manuscript reference order). In some cases, it is a source of important information that is also in some places missing from the main text (already mentioned section 2.4. and corresponding S.2.4, figure S16). If the authors want to keep some technical details apart from the main text (understandable with that much material), then I would ask to consider putting the more important parts in the Annex, in order to a) maintain the high editing stan-

C4

ard and thus make reading easier, b) keep the important information together with the text. At the very minimum the editing of the supplement needs to be improved.

3 Detailed comments:

Line numbers are given for identification. Comments for figures are given at the end of the list.

L25, also later in the text: ACP requires exponential notation of units, consult the 'manuscript preparation' on the ACP website for details (www.atmospheric-chemistry-and-physics.net/for_authors/manuscript_preparation.html)

L35: I'd suggest putting ppm outside of parentheses and mole fraction inside, as the ppm/ppb notation is the dominant one in the manuscript.

L46-49: Sentence needs rephrasing

L60-62: This paragraph does not fit well here, would be better if info given as part of previous or next.

L64: 'high precision' used twice

L75-77: What were the main findings from these studies? Specifically, it would be good to comment on whether these methods can be useful for up-scaling.

L78: 'We quantified emissions in this study using measured CH₄ enhancements above background, which were detected' - This needs revision; also, it feels like Weller et al. 2019 should already be quoted here, perhaps something like: 'In this study, we have quantified the CH₄ emissions using the method described by Weller et al., who demonstrated ...'

C5

L91: Was the reproducibility tested by the authors? Picarro currently gives 0.5 ppb for 5 s raw data. If the reproducibility was tested by the authors, please provide some details on the testing (either reference, or brief description of the experiment). Was the water correction modified or the factory settings were used? Please state that explicitly and also provide information if necessary.

L99: Discard 'about' or the approximation sign

L100-101: Similar to comment for L91, please provide more info.

L104: Info on how the delay was calculated should be given here, but can be found later in L202. Please combine both (see major comment no. 1)

L111-112: Please spell out the main findings of the discussed comparison. Also, the reference to annex section number (S.2.) where it is discussed should be present (next to table S3 ref.). In general, sections of supplement should be referenced and not only tables or figures from it.

L129: When reading the sentence for the first time I have understood that the gas pipeline network corresponds to the street network 1:1. Is that correct, or the general coverage of municipal areas is meant? Please clarify.

L137: discard 'at the following links: Utrecht and Hamburg'

L147-148: Please briefly explain how the vehicles can be methane sources (with reference for subsequent discussion further in the text).

L148: Please state clearly how many revisits were usually made.

L159-161: Have any cases where new leaks have occurred in-between surveys been observed?

L166-167: Parentheses missing? 3 L bag for a price of 2 L bag is too good to be true.

L168-169: More details on sampling are needed. Was the data collection stationary

C6

or also mobile? How was the plume / non-plume location determined? What was the flushing time? Was the sample dried? How?

L173: See major comment no. 3.

L191-192: Case shown in S5 is special and I strongly recommend to remove it from here and discuss later. As it is now, the text does not explain it, and thus may imply that all the cars are potential sources of CH₄, which is certainly not the case.

L194: Consider providing these standards in the parentheses or the supplement section S.7.

L196: How many such cases were observed? Could they be important for the overall budget?

L198: Please state the reasons for this exclusion, briefly.

L199-200: Just a small comment, no action needed: I don't see the benefit of this artificial increase of the data frequency. This brings no new information at the cost of tripling of the data that needs to be processed.

L200-201: Wording. If the time was just converted to UTC, then calling it 'a correction' is not warranted. Consider changing to: 'Following the interpolation step, the data was converted to UTC, and subsequently corrected for ...'

L202: About the delay time estimation: 5–30 seconds is a very broad range. Were the ranges so variable for both instruments, or was it 5 for one and 30 for the other? Also, how was the pulse generated? Can you estimate precision of that delay estimation (even grossly)?

L204: Reference order needs correction. Previous reference supplement figure was S5 (L190).

L207: In CO₂ signal (Fig. S8), it can be clearly seen that the background line is sometimes higher than the observed signal. Since this plot is about the background, it

C7

would be good to change the limits of y axis to make the calculated background visible clearer, especially for methane. Please give some comment about the possible negative enhancements after subtracting such background (can it affect the estimation of emissions?).

L217: Please add 'peak' after enhancement, to make it clear that it's not about the release height.

L237: Wording. Why should results from different cities be comparable? The authors clearly mean that the analysis software used on a given dataset should be comparable. Please clarify. Actually, this whole paragraph can be limited to information that 'Our software was compared to analysis tools developed by CSU (von Fisher et al. 2017, Weller et al. 2019) and no significant differences were observed (see SI, section S.2.7)'.
L251: Erase 'areas'.

L252: Erase empty parentheses.

L253 and L384: 'drive-by'- I propose 'mobile'. I was surprised to find it used in Fisher et al. (albeit only once), as in U.S. this word is sometimes used to describe something much more nefarious than GHG observations.

L254: 'We report (. . .)' – Unclear what is meant in this sentence, please rephrase it.

L256-257: Erase 'both' and 'each day's'

L269-274: How was the release height determined? How is the uncertainty of this determination included in the uncertainty of emission?

L283: This section should be combined with 2.4.

L286: Info on the isotopic scales used in this study needs to be given.

L293-294: Please provide explanation on why these particular ranges were selected.

C8

For signatures, specifically also provide references supporting the choice of isotopic signatures. Please keep in mind that for fossil fuel signatures, figure 7 from Rockmann et al. 2016 doesn't give a full picture – see e.g. Sherwood et al. 2017 for a broad overview of isotopic signatures for fossil methane.

L314-315: 'were correspond' – corresponded

L315: What is the uncertainty here? This relates to major comment 2.

L378: Why uncertainty given only for wind direction?

L394: von Fisher – V should be capitalised at the start of the sentence (Von Fisher).

L404: 'About 50 %' - please give the specific number that was used in the calculation.

L420-422: These numbers are in fact very similar. The variability of $\delta^{13}\text{C}$ in the natural gas can be quite substantial. See e.g. Fig 4 in Sherwood et al, 2017.

L426: % used twice

L433: Reference to Figure 1?

L436-438: a) Fig S16 also points to the local sewage system as potentially important source, but this is not mentioned here. b) Please be more precise in the argument here - i.e. explain why measurements around the lake point to anaerobic methanogenesis specifically. Linking to a), please include info on the potential role of the sewage system if needed. Is it possible that the sewage is seeping into the lake?

L443: 'because there is no publicly available activity data for associated activity data' – please rephrase.

L452: Too many parentheses. I suggest '(...) 40 kg km⁻¹ yr⁻¹ (for other material, p < 200 mbar; see p. 130 in Peek et al., 2019, for details)

L473: 'credibility interval' – confidence interval

L491: '(...) factors can be gas pipeline age and material, sewer system.' Part of sen-
C9

tence missing? Please rephrase.

L531: 'were' – where

L533: 'as shown' used twice.

L542-544: The scheme from S18 cannot be treated as a 'protocol' without a proper description of the method. In reality, it describes the main components of the method applied in the study, so in fact the manuscript itself is more of such a protocol. As it stands now, consider either expanding the description in the supplement (so gas companies might actually use it as a protocol) or discard it altogether.

L561: 'corresponding to emissions of about 107 t CH₄ / yr' – exactly 110 t yr⁻¹ is given in L332.

L562: Please state the method, e.g. 'These estimates, based only on the studied area, were then up-scaled for the total municipal area, using the road network map as a proxy to (...)'

L567: 'were from' - I suggest 'originated from'

Figure 2: a) Please fix the x axis description - extra arrow unnecessary. b) the plot is cropped in the lower part, by several pixels. c) extra grid dashed lines (green) are unnecessary, make the labels difficult to read.

Figure 3: a) again, the arrow from total emissions to 'Road classes' seems unnecessary. b) please explain the arrows from the plot in the caption.

Figure 4: I recommend plotting all US cities in a single colour (grey?) and simply label the line as ('15 US Cities', Weller et al.) or similar. The colours are indistinguishable anyway.

Figure 5: Excellent plot! a) Please add comment about the uncertainty of $\delta^{13}\text{C}$ and $\delta^2\text{H}$ signatures in the caption (they were only plotted for C2/C1. I also softly suggest to label the plots with a-b-c-d and move the labels into figure caption. b) On previous plot

the units were placed in parentheses, consider keeping notation consistent.

Figure 6: a) Plumes of what? b) Please also provide information on whether the peaks are on the same scale; if yes, then what is the plotted range of mole fractions (if those are mole fractions)?

4 References

2017, Sherwood et al., Global Inventory of Gas Geochemistry Data from Fossil Fuel, Microbial and Burning Sources, version 2017, Earth Syst. Sci. Data, 9, 639–656, 2017 <https://doi.org/10.5194/essd-9-639-2017>

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-657>, 2020.