Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
https://doi.org/10.5194/acp-2021-606-RC1, 2021

Comment on acp-2021-606

Anonymous Referee #2

Referee comment on "Continuous CH_4 and $\delta^{13}CH_4$ measurements in London demonstrate under-reported natural gas leakage" by Eric Saboya et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-606-RC1, 2021

In the manuscript entitled "Continuous CH4 and δ13CH4 measurements in London demonstrate under-reported natural gas leakage," the authors utilize tower measurements of methane to evaluate the inventory representation of urban methane in London, in both the global EDGAR inventory as well as the national NAEI. The study investigates urban methane emissions which remain, despite the proximity to a large portion of the global population, a poorly characterized part of the methane budget. The finding that methane emissions associated with the natural gas infrastructure are undercounted in inventories is consistent with other studies of urban centers around the global, and points to an area where potential mitigation efforts are tangible, impactful, and requiring of further study.

The methodologies presented are consistent with those established in the literature previously, namely the use of an established atmospheric dispersion model used in conjunction with a gridded emission inventories to generate simulated signals for comparison with observations and isotopic source analysis with Keeling plots. The manuscript is organized in a logical manner and the writing is concise and mostly clear. At times, however, it reads more like a report than a research article. On a several occasions (detailed below in the 'specific comments'), important details or context are missing from the text. With the inclusion of these additional details and discussion, I believe the manuscript meets the threshold for publication and would be of interest to the readers of ACP.

Specific Comments

- Introduction: The authors motivate their work by highlighting previous works investigating urban CH4 emissions, both in London and around the world, as well as other recent studies based on δ13CH4 measurements. There is, however, no statement in the introduction justifying why further measurements are needed. Is it that previous studies have suggested that urban methane is higher than inventoried, but the cause of the discrepancy is not yet know (i.e. need for attribution using isotopic measurements)? A stronger statement of why the presented work is important would aid the reader in understanding how this work adds value to the existing body of work.
- Lines 208-210: "To compare the simulated excess CH4 mole fractions to the measurements at ICL, we subtract daily background CH4 mole fractions from the Mace Head Observatory (Arnold et al., 2018; Manning et al., 2011) from the 20-minute

averaged measurements at ICL." Is this background methodology consistent with other works? Is that why those references are included? If this is consistent with previous studies, it makes sense to explicitly state that. Is the location of Mace Head Observatory representative as a typical upwind location for the domain? Are there time periods where the CH4 signal at Mace Head Observatory is not representative of the background for the urban domain?

- Section 2.4.2: Why use EDGAR v4.3.2 when newer versions are available? Was this the newest version when the work began? If so, would expect anything to change if the newer versions (v5.0, v6) was used instead? This especially relevant because a work is cited (Klausner et al. 2020) that compares their flux measurements to EDGAR v5.0.
- Lines 272-274: "Subtracting the 25 km NAEI emissions from the 25 km EDGAR emissions (Fig. 3e-f) indicates the largest differences between inventories were in cities; London, Birmingham and the Leeds-Sheffield area, which have higher emissions in the EDGAR inventory." What is the takeaway from this statement? That the largest discrepancies in inventory representation of ch4 appear in cities, suggesting that inventory don't capture these emissions well? As written it is not really clear.
- Figure 8: I find this presentation of this data as a time-series difficult to interpret. If the goal is look at the relationship with wind direction and δs, a correlation plot (e.g. wind direction vs. δs) or a polar wind chart would show this more directly.
- Section 3.2.1: I believe the inclusion of nighttime tower observations in this section requires more discussion. As the authors state, the model transport error is smaller in the afternoon. Accordingly, it is not clear from the manuscript as written if the non-afternoon measurements add anything to the findings. Additionally, including nighttime observations is a deviation from several previous tower-based urban studies (including Mckain et al. cited in the introduction), and thus requires more discussion to support the interpretation of this data. I understand that the nighttime observations are used in 12-hour Keeling plot analysis, however, without further information it unclear if in the simulated methane for 'all hours' we are just seeing the influence of higher transport error.
- Section 3.2.1: Similar to the previous comment, what does the role of higher transport uncertainty in the non-afternoon hours play in the interpretation of the model-observation mismatch of δ13CH4? In Figure 13 only afternoon hours are shown in the natural gas scaling test. Does the focus on afternoon hours indicate lower confidence in the nighttime simulations?
- Lines 516-521: This paragraph provides some references to other works examining London as points of comparison, but no discussion is included as to why the presented results may or may not differ from these previous works. Without this information it is unclear how the findings presented here fit into the existing body of knowledge for urban methane in London.

Technical Corrections:

- Line 184 – The reference to the supplementary material should be to specific section to aid the reader.
- Figure 2 caption: The version number of EDGAR should be included, especially since newer versions are now available.
- Lines 327-328: "We focus on δ13CH4 measurements from May 2019 onwards in our analysis as the associated measurement uncertainty is smaller (Sect. 2.2.3)." I believe it would really aid the ready to briefly recap why the uncertainty is lower for May 2019 and onward, even just briefly. It is likely the reader will not recall this detail from earlier in the paper.
- Lines 413-415: "Background mole fractions exert a significant leverage on the values of β. We account for this by randomly varying the background mole fractions based on

their standard deviations and calculating the β values 150 times.” It is unclear which standard deviations are being used here. Further clarification is needed.

- Line 453: leaks, not leak