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30th June 2022

Dear Dr Chen,

This letter is to accompany the resubmission of our manuscript entitled "*A quantitative comparison of methods used to measure smaller methane emissions typically observed from superannuated oil and gas infrastructure*", which we would like you to consider for publication. Our paper describes controlled release experiments at the METC facility in Fort Collins, USA that investigate the accuracy and precision of several methods commonly used to measure methane emissions. The controlled releases were all below 200 g CH_4 h^{-1} and the methods include: static chambers, dynamic chambers, a Hi-Flow sampling system, a backward Lagrangian stochastic method and the Gaussian Plume method. To our knowledge this is the first time that methods for measuring methane emissions from point sources between 40 and 200 g CH_4 h^{-1} have been quantitively assessed against a known reference source and each other.

We appreciate your time in reviewing our manuscript and look forward to hearing from you.

Kind regards,

Stuart N. Riddick (corresponding author)

and co-authors: Riley Ancona, Clay Bell, Mercy Mbua, Aidan Duggan, Tim Vaughan, Kristine Bennett and Dan Zimmerle

Dear Dr Chen,

We thank reviewer 1 for their comments. As suggested, we have amended the manuscript to address the reviewers' comments and have indicated changes to the manuscript in red text.

Please find our detailed responses below.

Reviewer 1 General comment 1:

First, it's unclear what the authors mean by "small". The emission rates that the authors are looking at (40-200 g/hour methane) are orders of magnitude larger than soil emission rates (e.g., agricultural soils). I am pointing out the agricultural soils because the authors are attempting to replicate methods for agricultural soils, although it's unclear if they are because the method doesn't seem to be the same – in particular the use of the fan and the lack of pre-installation of collars. It seems to be that the methods presented here are really focused on oil and gas wells. I suggest that the authors be clear on the applicability of the results and revise the title accordingly.

Response to reviewer:

As suggested, we have changed the title and text to highlight this study is focused on emissions from oil and gas infrastructure.

Changes to the manuscript:

Title: A quantitative comparison of methods used to measure smaller methane emissions typically observed from superannuated oil and gas infrastructure

At L7: Recent interest measuring methane (CH_4) emissions from abandoned oil and gas infrastructure has resulted in several methods being continually used to quantify point source emissions less than 200g CH_4 $hour^{-1}$.

At L41: Several methods are being used to measure emissions from these smaller point sources (less than 200 g CH_4 $hour^{-1}$) from abandoned oil and gas infrastructure.

Reviewer 1 General comment 2:

The authors claim in a few places that the static chamber is the simplest. But based on what's presented, the HiFlow sampler is the simplest – especially in terms of use and post-measurement analysis.

Response to reviewer:

This is a subjective observation that we are happy to remove.

Reviewer 1 General comment 3:

It would be easier for the reader if the Conclusions section is separated into two sections: discussions and conclusions.

Response to reviewer:

Have edited the manuscript as suggested

LINE-BY-LINE COMMENTS

Reviewer 1 Line-by-line comment 1:

Title: What is meant by “small”? Are the authors referring to the physical size of the emission source or the emission rate? Also, specify that this study applies mainly to oil and gas sources.

Response to reviewer:

As above, have changed the title and some of the text

Changes to the manuscript:

Title: A quantitative comparison of methods used to measure smaller methane emissions typically observed from superannuated oil and gas infrastructure

At L7: Recent interest measuring methane (CH_4) emissions from abandoned oil and gas infrastructure has resulted in several methods being continually used to quantify point source emissions less than 200g CH_4 $hour^{-1}$.

At L41: Several methods are being used to measure emissions from these smaller point sources (less than 200 g CH_4 $hour^{-1}$) from abandoned oil and gas infrastructure.

Reviewer 1 Line-by-line comment 2:

line 36: There are missing references here: Townsend-Small et al, 2021, Saint-Vincent et al. 2020, and El Hachem and Kang, 2022.

Response to reviewer:

Have added references as suggested.

Reviewer 1 Line-by-line comment 3:

line 38: replace "form" with "inform". Here and elsewhere, the paper needs proofreading.

Response to reviewer:

We believe “form” is correct and have proofread the rest of the manuscript.

Reviewer 1 Line-by-line comment 4:

line 48: grammatical error.

Response to reviewer:

Have deleted “using”

Reviewer 1 Line-by-line comment 5:

line 51-52: Tracer release has been used to measure active and abandoned wells in Romania. See Delre et al. (2022) published in Elementa.

Response to reviewer:

Have added the citation.

Changes to the manuscript:

At L55: Tracer release is technically demanding, takes a long time to make a single measurement and requires road access for measurement, although it has been used to measure nonproducing wells in Hungary (Delre et al., 2022)

Reviewer 1 Line-by-line comment 6:

line 72: spell out “PPR”

Response to reviewer:

Have added.

Changes to the manuscript:

At L76 personal protective equipment (PPE)

Reviewer 1 Line-by-line comment 7:

line 73: spell out "FR".

Response to reviewer:

Have added to text

Changes to the manuscript:

At L77: flame resistant (FR)

Reviewer 1 Line-by-line comment 8:

line 80: the controlled release testing was performed with 85 to 95% by volume methane but if there is uncertainty in the methane concentration, this would translate to an error in the target emission rate. Which concentration was used to determine the target flow rate?

Response to reviewer:

At METEC the methane content of the natural gas in each release is measured by gas chromatography and accounted for in the known emission rate.

Changes to the manuscript:

At L85: At METEC the methane content of the natural gas in each release is measured by gas chromatography and accounted for in the known emission rate.

Reviewer 1 Line-by-line comment 9:

line 93: remove "relatively simple" as it is too subjective. The simplest method considered here is the HiFlow sampler.

Response to reviewer:

Have removed

Changes to the manuscript:

At L98: For the static chamber method a container of a known volume

Reviewer 1 Line-by-line comment 10:

line 95: it is not necessary to have a fan in a static chamber. Collier et al (2014) did not use a fan and Pihalatie et al. (2013) did not always use a fan.

Response to reviewer:

Acknowledged, have removed the sentence.

Reviewer 1 Line-by-line comment 11:

line 99: as mentioned above, Pihalatie et al. (2013) and Collier et al (2014) do not always use a fan. More importantly, their focus is on measuring agricultural soils with emission rates orders of magnitude below the range considered in this paper. The authors need to justify why the methods outlined in these papers are followed given that the application and emission rates are so different.

Response to reviewer:

Have changed the reference to Kang et al (2014; 2016) that did use the static chamber to measure emissions from oil and gas wells.

Reviewer 1 Line-by-line comment 12:

line 108: replace "chambers" with "chamber"

Response to reviewer:

Have replaced

Reviewer 1 Line-by-line comment 13:

line 110: was the chamber anchored to the ground as done in Collier et al (2014)? Following Collier et al. (2014), the anchoring needs to be done 1 day in advance. Also, why would the static chamber be more likely to be lifted off the ground compared to the dynamic chamber? Is the pump somehow anchoring the dynamic chamber?

Response to reviewer:

The chamber was made of two parts, a smaller lower part that was secured 4 cm into the soil and a larger upper part that was fixed to the lower part at the start of the experiment.

The 0.5 m^3 chamber was very large and easily caught the wind. The dynamic chamber was smaller and had the pump sitting on top.

Changes to the manuscript:

At L104: The chamber was constructed of two parts, a smaller lower part that was secured 4 cm into the soil and a larger upper part that was fixed to the lower part at the start of the experiment.

At L 115: larger (0.5 m^3) chamber

At L132: container (0.12 m^3) enclosing

Reviewer 1 Line-by-line comment 14:

line 141: did Riddick et al (2019) measure at similar rates as those tested in this paper?

Response to reviewer:

Yes, the emissions from abandoned wells reported by Riddick et al. (2019) ranged from background to 100 g CH_4 h^{-1}

Changes to the manuscript:

At L142: Methane emissions from abandoned wells have been quantified using this method between 4 µg CH_4 hr^{-1} and 100 g CH_4 hr^{-1} (Riddick et al., 2019a).

Reviewer 1 Line-by-line comment 15:

line 143: what is the criteria to determine if steady state has been reached?

Response to reviewer:

As stated at L148, the steady state was reached when the CH_4 concentration inside had become constant, as measured by the HXG-2D sensor.

Changes to the manuscript

At L148: The chamber was left until the CH4 concentration inside had become constant, as measured by a Sensit HXG-2D sensor (Sensit Technologies, Valparaiso, IN, USA).

Reviewer 1 Line-by-line comment 16:

line 152: is the industry standard set by some industry body? Or do the authors mean that this is common practice?

Response to reviewer:

Have deleted “industry standard”

Reviewer 1 Line-by-line comment 17:

line 169-170: how is the PGSC determined for a measurement determined? This is provided to some extent in the supporting information. However, given the large error in the GP method and the authors’ attribution of errors to the PGSC parameterization, it is important to discuss them here.

Response to reviewer:

The PGSC can either be calculated using the wind speed and a measure of the solar irradiance (Supplementary Material Section 1 Table S1) or using a sonic anemometer. Here, the former method was employed and the PGSC calculated from the wind speed (u, m s^{-1}) measured at 1.2 m and irradiance measured at the emission point (G, kW m^{-2}). Pasquill and Smith (1983) originally defined strong irradiance as sunny midday in midsummer in England and slight insolation to similar conditions in midwinter. Here we class strong irradiance as > 1 kW m^{-2}, Moderate irradiance 0.5 kW m^{-2} to 1 kW m-2 and Light irradiance as > 0.5 kW m^{-2}.

Changes to the manuscript:

At L191: The PGSC can either be calculated using the wind speed and a measure of the solar irradiance (Supplementary Material Section 1 Table S1) or using a sonic anemometer. Here, the former method was employed and the PGSC calculated from the wind speed (u, m s^{-1}) measured at 1.2 m and irradiance measured at the emission point (G, kW m^{-2}). Pasquill and Smith (1983) originally defined strong irradiance as sunny midday in midsummer in England and slight insolation to similar conditions in midwinter. Here we class strong irradiance as > 1 kW m^{-2}, Moderate irradiance 0.5 kW m^{-2} to 1 kW m^{-2} and Light irradiance as > 0.5 kW m^{-2}.

Reviewer 1 Line-by-line comment 18:

line 179: subscript 4 in "CH4".

Response to reviewer:

Have changed

Reviewer 1 Line-by-line comment 19:

line 189: provide approximate range of distances rather than saying "as close as possible".

Response to reviewer:

Have included a range of distances

Changes to the manuscript:

At L199: (between 1 and 10 m)

Reviewer 1 Line-by-line comment 20:

line 229: The Hi Flow sampler is logistically the simplest, not the static chamber.

Response to reviewer:

As above, have removed the sentence

Reviewer 1 Line-by-line comment 21:

line 250: are the authors suggesting that sources emitting aromatic hydrocarbons and hydrogen sulphide should not be used by the static chamber method or by all methods? This sentence is unclear.

Response to reviewer:

Have removed the sentence.

Reviewer 1 Line-by-line comment 22:

line 251: It's unclear what "our measurement data" is.

Response to reviewer:

Have changed the sentence

Changes to the manuscript:

At L 258: As such, we have not presented the measurement data collected during the static chamber experiments and strongly encourage the use of an alternative method.

Reviewer 1 Line-by-line comment 23:

line 254: missing word. proof read.

Response to reviewer:

Have added “time” to the manuscript.

Reviewer 1 Line-by-line comment 24:

line 276: how much does the Hi Flow instrument cost?

Response to reviewer:

The Hi Flow costs $35k.

Changes to the manuscript:

At L284: (costs $35,000)

Reviewer 1 Line-by-line comment 25:

line 282: how is safety assessed here? the oil and gas industry has specific safety standards and ways to assess/handle safety, and personnel are required to go through rigorous safety training. All oil and gas sites can be dangerous and carry safety risks. I suggest that the authors not make conclusions on safety conditions on sites, as that's beyond the scope of this work.

Response to reviewer:

Have removed the comment.

Dear Dr Chen,

We thank reviewer 2 for their comments. As suggested, we have amended the manuscript to address the reviewers' comments and have indicated changes to the manuscript in red text.

Please find our detailed responses below.

Reviewer 2 General comment 1:

The result section is fairly light and short in substance for a research paper, or at least by the standard that I would expect in AMT.

Response to reviewer:

All of the results are presented in the SI to help reduce the word limit and reduce the burden on the publication. The aim of this study was to produce simple ground-truth validation of quantification techniques and present the data is as simple a form as possible.

Reviewer 2 General comment 2:

The Conclusion section is structured in a very unusual way, with a new figure (Fig 3) and comparison against other research that were not introduced before. I suggest to split this Conclusions into a Discussion section and a more "classic" Conclusion section.

Response to reviewer:

Have amended the manuscript as suggested.

Reviewer 2 General comment 3:

L50 I suggest to moderate this statement in relation to the leak rate e.g. "cannot be used to quantify emission below XXX kg/h")

Response to reviewer: The sentence is correct as it stands, OGI cameras are not quantification instruments and con only be use for emission detections.

Changes to manuscript:

At L 50: While OGI cameras can be used for detecting emissions greater than 20 g CH_4 h^{-1} (Ravikumar et al., 2018; Stovern et al., 2020; Zimmerle et al., 2020), using this method for quantification remains in development with few studies published to date investigating the accuracy of emission rate estimates from OGI (Kang et al., 2022).

Reviewer 2 General comment 4:

L361 The use of the GP approach here is made with a single measurement in plume. If this context were explicitly stated I would agree with the statement. However mobile in situ measurements in (and across) the plume, even at distances shorter than 100m, would give much better results.

Response to reviewer:

Have clarified the text with your suggestion.

Changes to manuscript:

At L 369: This suggests that using the GP approach with a single measurement in the plume for distances less than 100 m, it is not correct to assume that repeat measurements will remove bias in the calculated average emission. It is currently unclear if mobile, in-situ measurements in and across the plume, even at distances shorter than 100m, would give much better results.

Editorial comments

Reviewer 2 Editorial comment 1:

L25 add against in: and AGAINST each other

Response to reviewer: As suggested, have added “against”

Reviewer 2 Editorial comment 2:

L50 difficulty

Response to reviewer: Have corrected

Reviewer 2 Editorial comment 3:

L75 add against in: and AGAINST each other

Response to reviewer: As suggested, have added “against”