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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

We thank the reviewer 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:
The paper presents controlled release experiments of multiple methane emission measurement methods that have been used to quantify emission rates of oil and gas sources. They considered the dynamic chamber, the hi flow sampler, the Gaussian plume method, and the backward Lagrangian stochastic models and to some extent, static chambers (see below for more on this). The contribution can be useful as methane monitoring is important for greenhouse gas emission reductions. However, below are some important revisions that are needed to make it easier for readers to understand the paper and the results.

Response to reviewer:
The controlled release experiments conducted at METEC show the static method to be inherently dangerous as we were unable to remove the chamber without the four-gas monitor, worn on the observer's collar, detecting CH_4 concentrations that exceeded the lower explosive limit, i.e. triggered the monitor's alarm. This poses a considerable risk to the observer. In addition to being an explosive risk, natural gas emitted from the subsurface could contain aromatic hydrocarbons and other toxic gases which could also collect to hazardous concentrations inside the static chamber and inhaled when removing the chamber. Therefore, we recommend that the static chamber method should not be used to quantify emissions from oil and gas infrastructure. While the reviewer feels that we should omit the static chamber method from the paper completely, we feel that its omission could appear to be a tacit acknowledgement that it is an acceptable method for quantifying emissions. Our observations show the static chamber method to be dangerous and want to clearly state that we do not recommend its use in the field. To make this point we have added the following to the manuscript.

Changes to the manuscript:
L248: "The method is inherently dangerous as we were unable to remove the chamber without the four-gas monitor, worn on the observer's collar, detecting CH_4 concentrations that exceeded the lower explosive limit, i.e. triggered the monitor's alarm."
L309: "Static chamber results are not presented as we were unable to remove the chamber without exposing the observer to an explosive environment."
At L 327: "This study investigates the utility, accuracy and precision of five methods"
At L329: "When the method has been shown to be no danger to the observer, we generate CH_4 emission estimates"
At L 333: "The static chamber method was found to be inherently dangerous, as the observer was unable to remove the chamber without being exposed to an explosive environment. As a result, the data from the static chamber experiments have not been presented in this study. Furthermore, the experiment conducted at METEC used processed natural gas where heavier/aromatic hydrocarbons and toxic gases have been removed. Gas emitted from abandoned oil and gas wells is unrefined and we advise that the static chamber method should not be used to quantify emissions of an unknown composition of natural gas as this

could expose the observer to high concentrations of toxic gas. Therefore, we recommend that one of the other methods presented here should be used to quantify emissions from abandoned oil and gas wells."

Reviewer 1 General comment 2:
There are several structural issues with the paper. Section 3.1 on the Method narrative, which is the first section of the results, would be best placed in the Methods or Discussion sections as no results are presented.

Response to reviewer:
Even though Section 3.1 contains no quantitative data, it presents qualitative information of the suitability of each method for deployment in the field. This helps to address objective 4: "Make recommendations on the suitability of each method for measuring emissions from relatively small point sources.". We feel that it is best placed in the results section. To highlight this, we have changed the name of the section to: "3.1 Method narrative – Qualitative observations of methods"

Changes to the manuscript:
At L 242: "**3.1 Method narrative – Qualitative observations of methods"**

Reviewer 1 General comment 3:
The presentation of the static chamber in the methods section (and the method narrative) is confusing as no results are shown. It would be easier for readers if the authors just limited the scope to the four methods that they analyzed. The authors could simply say that the four chosen methods fit the authors' objectives and move on.

Response to reviewer:
As noted above (General comment 1), we feel that the inclusion of the static method to this paper is essential. The method is inherently dangerous, should not be used to collect an unknown composition of natural gas at an unknown rate and another method should be used. We note that reviewer is confused by the omission of data and to address this we make our message very clear at the start of each results sections (3.1 and 3.2) and in the discussion.

Changes to the manuscript:
L248: "The method is inherently dangerous as we were unable to remove the chamber without the four-gas monitor, worn on the observer's collar, detecting CH_4 concentrations that exceeded the lower explosive limit, i.e. triggered the monitor's alarm."
L309: "Static chamber results are not presented as we were unable to remove the chamber without exposing the observer to an explosive environment."
At L 327: "This study investigates the utility, accuracy and precision of five methods"
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At L 333: "The static chamber method was found to be inherently dangerous, as the observer was unable to remove the chamber without being exposed to an explosive environment. As a result, the data from the static chamber experiments have not been presented in this study. Furthermore, the experiment conducted at METEC used processed natural gas where heavier/aromatic hydrocarbons and toxic gases have been removed. Gas emitted from

abandoned oil and gas wells is unrefined and we advise that the static chamber method should not be used to quantify emissions of an unknown composition of natural gas as this could expose the observer to high concentrations of toxic gas. Therefore, we recommend that one of the other methods presented here should be used to quantify emissions from abandoned oil and gas wells."

Reviewer 1 Line-by-line comment 1:
L27-28: The GWP for methane is wrong. It's ~30 for a 100 year time frame and is 86 for a 20 year time frame. Also, should cite the latest IPCC assessment report.

Response to reviewer:
As suggested, number changed and citation added

Reviewer 1 Line-by-line comment 2:
L95: add "methane" after "5,000 ppm"

Response to reviewer:
"CH_4" added

Reviewer 1 Line-by-line comment 3:
L109 and L118: One says at least three and then it says four samples. Sounds like redundant statements.

Response to reviewer:
Have deleted "at least three further"

Reviewer 1 Line-by-line comment 4:
L135: Would be useful to list the period of time needed for the CH4 concentrations to be stable.

Response to reviewer:
This varies as the time is a function of the emission rate and the flow of air through the chamber and the size of the chamber, so impossible to include a value.

Reviewer 1 Line-by-line comment 5:
L135: how did the authors determine that steady state has been reached?

Response to reviewer:
As stated at L150: "The chamber was left until the CH_4 concentration inside had become constant, as measured by a Sensit HXG-2D sensor (Sensit Technologies, Valparaiso, IN, USA)."

Reviewer 1 Line-by-line comment 6:
L139: which area of the chamber? Footprint?

Response to reviewer:
Changed "area" to "footprint"

Reviewer 1 Line-by-line comment 7:
L153: Add "rate" after "emission"

Response to reviewer:
"rate" added

Reviewer 1 Line-by-line comment 8:
L157: Is the Hi Flow sampler offered by Heath the same model as the Bacharach Hi Flow Sampler tested here? The names are different. On the Heath website, it doesn't mention the word "Bacharach".

Response to reviewer:
Apologies, this should be "(Bacharach, Pittsburgh, USA, www.mybacharach.com)". Bacharach are the only current manufacturer, Heath does not make a Hi Flow.

Reviewer 1 Line-by-line comment 9:
L158: "currently-available"

Response to reviewer:
Changed as suggested

Reviewer 1 Line-by-line comment 10:
L159: Connolly et al. (2019) used the Bacharach Hi Flow Sampler and it is not clear that this is the same as the one available from Heath.
Equation 3: all variables in the equation need to be defined.
2.3 Hi Flow. There are three high flow samplers mentioned here: the Bacharach, the Heath Hi Flow Sampler, and the new Hi Flow Unit by Vaughn et al. Which one was used here?

Response to reviewer:
The Bacharach Hi Flow was used in this study. We have added text to clarify this.

Changes to the manuscript:
At L160: "The Bacharach Hi Flow Sampler (Bacharach, Pittsburgh, USA, www.mybacharach.com) is the only currently-available Hi Flow sampler and was used in this study,"
At L169: "The Bacharach Hi Flow sampler used in this study was calibrated monthly as recommended by the manufacturer."

Reviewer 1 Line-by-line comment 11:
L165-167: the last sentence is redundant as it's a repeat of what is said two sentences ago.

Response to reviewer:
Deleted as suggested

Reviewer 1 Line-by-line comment 12:
L193: how much better would the results be if a sonic anemometer was used? Why wasn't it used?

Response to reviewer:
PGSCs are very granular and the classifications are cover a broad range of Monin-Obukhov lengths, as measured by the sonic anemometer, therefore this is not expected to be a large source of error. Due to power requirements, sonic anemometers are unlikely to be used in the field and, as such, a more basic approach is adopted.

Changes to the manuscript:
At L197: "Due to power requirements, sonic anemometers are unlikely to be used in the field and, as such, a more basic approach is adopted and"

Reviewer 1 Line-by-line comment 13:
L196: First "class" should be replaced with "classify". More importantly, what is the justification for using this classification. There is no reference here

Response to reviewer:
"classify" has been added, as has a citation.

Reviewer 1 Line-by-line comment 14:
L231: Does twice mean that it was conducted 6 times in total (3 x 2)?

Response to reviewer:
No, the sentence is a legacy of previous edits and is no longer required. It has been removed.

Reviewer 1 Line-by-line comment 15:
L238 onwards: The method narrative is not really a result. It should go in the Methods or Discussion. I think a lot of it could just be deleted.

Response to reviewer:
Even though Section 3.1 contains no quantitative data, it presents qualitative information of the suitability of each method for deployment in the field. This helps to address objective 4: "Make recommendations on the suitability of each method for measuring emissions from relatively small point sources.". We feel that it is best placed in the results section. To highlight this, we have changed the name of the section to: "3.1 Method narrative – Qualitative observations of methods"

Reviewer 1 Line-by-line comment 16:
L239: this paragraph and the following ones on the static chamber should go in the Methods. Better yet, as per my comment above, these paragraphs should just be deleted.

Response to reviewer:
As mentioned above, we find the result that the method is dangerous warrants its inclusion into the paper. This has been emphasized throughout.

Changes to the manuscript:

L248: “The method is inherently dangerous as we were unable to remove the chamber without the four-gas monitor, worn on the observer’s collar, detecting CH_4 concentrations that exceeded the lower explosive limit, i.e. triggered the monitor’s alarm.”
L309: “Static chamber results are not presented as we were unable to remove the chamber without exposing the observer to an explosive environment.”
At L 327: “This study investigates the utility, accuracy and precision of five methods”
At L329: “When the method has been shown to be no danger to the observer, we generate CH_4 emission estimates”
At L 333: “The static chamber method was found to be inherently dangerous, as the observer was unable to remove the chamber without being exposed to an explosive environment. As a result, the data from the static chamber experiments have not been presented in this study. Furthermore, the experiment conducted at METEC used processed natural gas where heavier/aromatic hydrocarbons and toxic gases have been removed. Gas emitted from abandoned oil and gas wells is unrefined and we advise that the static chamber method should not be used to quantify emissions of an unknown composition of natural gas as this could expose the observer to high concentrations of toxic gas. Therefore, we recommend that one of the other methods presented here should be used to quantify emissions from abandoned oil and gas wells.”

Reviewer 1 Line-by-line comment 17:
L246-248: the static chambers (referred to as non-steady-state chambers) can be vented or not vented as described in Livingston and Hutchinson, a source the authors here reference.

Response to reviewer:
The action described here is that the analyser outlet is left to the open air, therefore actively pumping air from the chamber (i.e. a dynamic chamber). The word “vented” maybe misleading and has been changed

Changes to the manuscript:
L252: “air from the analyzer exhaust is actively pushed outside the chamber”

Reviewer 1 Line-by-line comment 18:
L249: why would the gas coming out of an analyzer be necessarily lower concentration? The analyzer can be non-destructive.

Response to reviewer:
Gas is taken into the analyser, the concentration analysed and a finite time later (depending on the length of the tube and the specification of the instrument) the gas is passed back into the static chamber. We are not suggesting the analyser is destroying the CH_4, more the gas reintroduced to the chamber is of a lower concentration as it has been removed and recycled.

Reviewer 1 Line-by-line comment 19:
L252-253: Delete sentence beginning with "It is unlikely that gas will mix... ". This statement does not below in the results as there is no result that the authors are presenting to support this.

Response to reviewer:

Have deleted as suggested.

Reviewer 1 Line-by-line comment 20:
L279: What gas/air is being pumped? What is the composition of the gas/air and how could it affect results?

Response to reviewer:
The air is ambient drawn through 2 m of tubing and unlikely to affect the results as the source of emission is inside the chamber and far away from the gas vented from the chamber.

Changes to the manuscript:
At L289: "Another factor that could affect accuracy of measurement is the air being pumped into the chamber, care should be taken to ensure the inlet is apart from other CH_4 sources and far away from the chamber outlet."

Reviewer 1 Line-by-line comment 21:
L318: This sentence contradicts L295, which says bLs is the best.

Response to reviewer:
Acknowledged, this is misleading and have corrected the text.

Changes to the manuscript:
At L341: "but the dynamic chamber is more accurate".