

Responses to reviews of “Measurements of methane emissions from natural gas gathering facilities and processing plants: measurement methods” by J. R. Roscioli et al.

Comments by G. Allen

1)

From the beginning to the end I was looking for some results for the flux (aka emissions). They must have been calculated but they seem to be absent. I can only imagine this is by design? Section 6 mentions repeatedly that “emissions” are presented in the plots and there is discussion of relative emissions from different source types in the text. However, “emissions” results are actually presented as gas concentrations (often spatio-temporal plots of concentrations along a plume at ground level) and not a mass per unit time. Is the intention that some powder is being kept dry for the companion Mitchell paper submitted to Env. Sci. Tech? If so, then I looked for that paper but it is not yet accessible so I could not see results there. From what I read in this text, it seems that the Mitchell paper will be an attempt to scale up these measurements to the national scale (actually a bottom-up approach but based on local top-down measurement here). The absence of fluxes is an obvious flaw in the current manuscript and any reader would want to see those results here. I strongly recommend those are added here to do this paper the justice it deserves. It is the obvious and necessary place for those results. I see no reason how that would tread on the toes of a paper that tries to scale up, which is very separate. And to try to separate the flux results would be an inefficient way to present the data.

Response:

The authors appreciate the concern regarding the lack of emission rates in the manuscript. The manuscript by Mitchell et al (now available as an ASAP) aims to explore the full dataset from this measurement campaign, comprising 130 facilities across the country, along multiple analysis vectors (e.g. by facility throughput, horsepower, equipment onsite, capacity, etc). A third manuscript by Marchese et al (in preparation) aims to perform a national level scale-up. However, we agree with the reviewer that the paper would be strengthened by providing emission rates for the facilities represented by the figures in the paper.

The revised manuscript will report the emission rates for the facilities discussed in the paper. The text will be modified to report the average emission rate, with uncertainty. The captions of figures displaying individual measured plumes will also report the emission rates determined by those plumes.

2)

Where was this site? Do you have an anonymity agreement with the site/company? If so, I would accept that it isn't necessary to name it. But if not, it would be useful to know where the measurements were so readers of the companion paper (when accessible) can assess how representative it is for the purposes of scaling up to a national average.

Response:

The campaign consisted of flux measurements from 130 different sites, ranging from small gathering facilities to large processing plants. An anonymity agreement was maintained

between all measurement/analysis teams and industry partners. Part of this agreement included not providing location information. However, subsequent papers (by Mitchell et al and Marchese et al) will delve deeper into how to scale up from a limited 130-site dataset to a national average.

3)

The discussion of the problems with e.g. PBL convection and topography-induced turbulence as invalidators of the Gaussian plume assumption was interesting. Is there any way you can add more to the discussion of how these effects may influence the results of this case study (or more generally) as guidance for future studies? Is it something that could be folded into an error budget; and if so, how? More generally, what I think you could add here (as it is a technical guidance paper) would be some details of how to construct a robust and conservative error budget for tracer release flux calculation that others could follow. I don't think this is out of scope of this paper and it would really add to its impact.

Response:

The manuscript aims to explore the sources of error in the tracer release methodology. These effects are discussed in the context of Gaussian dispersion to provide "rule-of-thumb" understanding of the factors influencing codispersion in these settings. As discussed in the section titled "*Understanding and optimizing data quality*", these effects include wind speed, downwind distance, and spacing between tracer and emission source. Notably, ~1400 individual plumes were used to determine the flow rates from these facilities, for which the vast majority employed two tracers released simultaneously. This provided two critical pieces of information: 1) an internal check of the codispersion between tracer and CH₄, and 2) a direct quantification of the error associated with the measurement. This second piece of information is compiled in Table 2 in the manuscript, in the form of variances for each type of data analysis used in the study. Histograms of the actual data from which these variances were derived are shown in the supplemental information. This data is intended to serve as a conservative error budget for the tracer release method, as it encompasses a wide range of experimental conditions experienced over the course of the many-month field campaign.

By design, the two tracers were placed in such a way as to surround the emission vector. Therefore, the lack of dispersion due to spatial separation, wind shear, turbulence, and other effects is folded into the observed variance, without the need to use dispersion models. The one exception is CH₄ entrained in plumes rising from exhaust stacks. An attempt at quantifying the effects of plume rise upon the measured flow rates is discussed in the supplemental information section of this paper. The paper by Mitchell et al. combines this information with facility details (exhaust temperature, flow rate, composition, etc) in order to determine its effect upon the final measurement results.

Future work will aim to extract specific drivers of uncertainty (such as distance from site, tracer separation, etc), using several thousand dual tracer plumes obtained by the authors over several field campaigns (dating to 2012).

4)

Can you suggest any ways that the tracer system fluxes could be validated against another method? I realise that wasn't done here but how much need for this do you feel there is and could you suggest any ways to do this, e.g. eddy covariance, Lagrangian mass balancing, airborne measurement etc? A brief concluding discussion on this could be a nice addition.

Response:

One way in which the method can be tested against other measurement techniques has been demonstrated in the manuscript by Subramanian et al (Envir. Sci. Technol, **2015**, *49*, 3252-3261), where onsite leak measurements were performed simultaneously with tracer release. That study found agreement between onsite emission estimates and tracer estimates to within experimental uncertainty at all but two of the facilities visited. Another way to explore this using other methods would be combining the tracer release with airborne mass balancing methods. Recent advances in small-aircraft instrumentation allows for such a comparison, which are currently being explored.

In addition to comparing to other measurement methods, the intent of the dual tracer release technique is that the second tracer serves to directly quantify the error associated with the tracer method in general, as the second tracer can serve as a second “analyte” gas for which the flow rate can be determined and compared to the known flow. As a consequence, it serves as its own internal validation, and a meaningful expression of the tracer release uncertainty.

We will modify the manuscript to emphasize these points (i.e. “The use of the second ‘internal standard’ tracer then mitigates the need for a calibration of the tracer release method, or for benchmarking against other methods. Emissions rates determined by tracer release have, however, been compared to detailed onsite leak measurements in Subramanian et al. That study found that these two techniques usually agreed to within experimental uncertainty”)

1)

Change “emphases” to “emphasis” in the abstract.

Response:

This will be corrected in the manuscript.

Comments by anonymous reviewer

1)

To me the only major shortcoming in the current MS is the complete lack of the actual flux data. While I understand that the measured fluxes will be reported in other papers that are either submitted (Mitchell et al.) or still in preparation (Subramaniam et al.), I do think that out of the 130 facilities the authors sampled, flux data from a few (4-5) would be of great value. For example, the authors could consider adding the flux data from the case studies used for illustrating the different methods (e.g. in Figures 2 to 5).

Response

The authors appreciate the concern raised here. As discussed above, the revised manuscript will provide emission rates for the facilities discussed in detail in this paper, including per-plume emission rates, to give the reader a handle on the inherent scatter in the emission data.

2)

Page 12359; Lines 1-2: Please add the quantitative range of emission assessments e.g. xx-yy

Response

These will be included in the revision. Some reports focus on specific basins and geographic regions, rather than providing a national scale-up (as is intended in this study). Nonetheless, a range of “leak” rates (in terms of fraction of throughput) will be provided.

3)

Section 2.2: Top down approaches: What about inverse modeling?

Response

The reviewer raises the very good point that this is not well described in the manuscript. Inverse modeling was intended to be included in the paragraph discussing meteorological transport simulations. The issues with such modeling are a lack of accuracy due to uncertainty in meteorological measurements, as well as in physical modeling of the emission plume/advection process. The text will be modified as follows:

“Top-down estimates of regional emissions are also commonly performed using meteorological transport simulations in combination with a network of fixed sensors (McKain, 2012; Bullock and Nettles, 2014; Zavala-Araiza et al., 2014), or using inversion modeling coupled with dispersion or advection models. Such methods can leverage preexisting sensor networks with data available 24 h day⁻¹. However, the interpretation of sensor data for emissions measurements is highly dependent upon atmospheric modelling, with large uncertainties (Nehrkorn et al., 2010; Draxler and Hess, 1998, 1997).”

4)

Section 2.4 Page 12366 Line 1: “..stability classes A, B, C: : :” It would be good to add a few lines describing the stability classes A, B, C

Response

This is a very good point, and a table of stability class vs meteorological conditions will now be included in supplemental and referred to in the main text as

“As a rule of thumb, for typical mid-day atmospheric conditions (stability classes A, B, or C, as described in the supplemental information) and downwind distances (100–3000 m), the horizontal width of a plume that is propagating according to Gaussian dispersion is ~20–50% of the distance that it has traveled from its source.”

4)

Section 2.4 Line 21: typo “..end” instead of “and”

Response

This will be corrected to read “end”

5)

Page 12360; Line 13: Using bar for pressure would convey the compression factor more easily than psi

Response

The use of psi in this context was used, as it is the common unit employed by industry. The authors will revise this to read “...usually ~800 psi (~55 bar)”.

6)

Page 12368, Line 25: “of” is not required

Response

This will be corrected in the revised manuscript

7)

Page 12369, line 20: “true” wind speed should be explained to readers

Response

The reviewer raises a good point. The manuscript will be revised to read:

“...record true wind direction (i.e. horizontal wind direction relative to true north), ...”

8)

Page 12370, lines 23-28: please add the dynamic range of the calibrations and linearity of the instrumental response. By what magnitude might drift in instrumental response between calibrations affect the data and analyses?

Response

The range of these calibrations will be included in the revised text. As a rule, the linearity of the absorption-based instruments used here (both Aerodyne QCL and Piccaro CRDS instruments) is extremely linear across any realistic concentration range that may be measured downwind. Typical R^2 values for such calibrations are $R^2 > 0.99$.

The instrumental calibrations varied by less than 5% over the course of the measurement campaign. While these calibration values were included in the analysis to prevent bias, we note that their variation is much smaller than the empirically-determined uncertainty in the tracer release method.

9)

Page 12376, line 28: “e” missing in spelling of “measurement”

Response

This will be corrected in the revised manuscript.

10)

Page 12377, line 8: What caused the GC data to be unreliable?

Response

The source of this unreliability can come in several forms

- 1) The GC data is old (common for small stations)
- 2) The GC instrument is uncalibrated (ethane can be a difficult measurement on GC)
- 3) The measured emissions may represent flow from other sources onsite (condensate tanks, flares, etc), while the GC data is typically from the inlet or outlet composition
- 4) The GC data might be from another facility handling similar gas.

11)

Figure 3: The figure caption states that the lower left panel displays lack of correlation but the $r^2 = 0.95$! Is there a mistake?

Response

This is an excellent point – the reason why $R^2=0.95$ is because it represents the ethane/methane correlation. The fact that it is strongly correlated indicates that the mobile laboratory is measuring natural gas. However, it is not an indicator of correlation with tracers (shown in bottom center and right).

The revised manuscript will include the following last sentence in the Figure 3 caption:

“Note, however, the strong correlation between C_2H_6 and CH_4 (bottom left), indicating that the observed methane is derived from natural gas.”

Other Changes:

The Acknowledgements section should be modified to read:

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