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Interactive comment on "Statistical atmospheric inversion of small-scale gas emissions by coupling the tracer release technique and Gaussian plume modeling: a test case with controlled methane emissions" *by* Sébastien Ars et al.

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Review of

"Statistical atmospheric inversion of small-scale gas emissions by coupling the tracer release technique and Gaussian plume modeling: a test case with controlled methane emissions"

by S. Ars et al.

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Discussion paper

Submitted by Scott C. Herndon, Joseph R. Roscioli and Tara I. Yacovitch, Aerodyne Research, Inc.

General Comments:

Overall, we think this is a rigorous methods paper and does a good job of describing a novel combination of tracer and dispersion methodologies. The descriptions of the experiments and methods are clear, and will be well understood by the community, both on the experimental side and on the dispersion side. We think this paper should be published after addressing the comments below.

1. Please cite some important related research that explores tracer mislocation effects:

Goetz et al. used a similar combined methodology in 2015 for tracer release experiments at wellpads. Goetz, J. D.; Floerchinger, C.; Fortner, E. C.; Wormhoudt, J.; Massoli, P.; Knighton, W. B.; Herndon, S. C.; Kolb, C. E.; Knipping, E.; Shaw, S. L.; et al. Atmospheric Emission Characterization of Marcellus Shale Natural Gas Development Sites. Environ. Sci. Technol. 2015, 49, (11), 7012; DOI: 10.1021/acs.est.5b00452.

Roscioli et al. Performed an extensive error analysis of the impact of tracer mislocation using dual-tracer release methodology. Additional methods of calculating the methane/tracer ratio are also described. Roscioli, J. R.; Yacovitch, T. I.; Floerchinger, C.; Mitchell, A. L.; Tkacik, D. S.; Subramanian, R.; Martinez, D. M.; Vaughn, T. L.; Williams, L.; Zimmerle, D.; et al. Measurements of methane emissions from natural gas gathering facilities and processing plants: measurement methods. Atmos. Meas. Tech. 2015, 8, (5), 2017; DOI: 10.5194/amt-8-2017-2015.

2. Please address some potential problems with the experimental measurements: Have experimentally measured mixing ratios been calibrated? Please briefly mention calibration procedure.

Line 370-375: The flow meter you refer to is an analog measurement of flow rate, so we don't think you have a time-resolved record of the flow rates. While you have a final

check of the release by mass difference, do you have any estimate of the variability of the flow rate over the course of the experimental measurements? This could be substantial, particularly with acetylene releases, which can vary as the cylinder cools. In such a case, the flow rate may appear the same on the flow meter, but the actual mass flow may be different.

Paragraph starting at 441: When multiple instruments are sampling in a mobile vehicle, they will often have different inlet times (lag due to air intake). This can be corrected based on experimental measurements, for example by delivering an excess of nitrogen or zero-air to the inlet tip, and monitoring the instrument responses. Has this been done experimentally? How does the time-shifting of data based on simulated plumes compare to the experimental lags?

3. Why are ideal tracer ratio experiments not producing acceptable results?

Config. 1 tracer release method estimates before Gaussian correction (e.g. Table 2, middle row) overestimate the true release rate by 19%, when it should produce accurate results without the need for correction (see discussion below for more detail). This discrepancy casts doubt on the biases of the remaining 3 configuration tests. The calibration of mixing ratios and the consideration of errors in the instantaneous flow rates (see above) should be considered.

Lines 584 – 590: Something seems wrong with this plume analysis. You are well above the detection limit (at least looking at Figures 4 and 5), so the explanation of why the tracer analysis is not accurate does not make sense. Configuration 1 should be the ideal case. Please rule out instrument calibration errors (both in measurement and release equipment), unit errors (temperature and pressure of measurement), and other experimental issues.

Dual-tracer release has been used in the past to quantify bias. When they are collocated, the emission rate of one tracer as derived from its downwind ratio to the other is found to agree extremely well with the known mass flow (much better than the 19%

error that is observed here). See Figure S4-4 of the supporting information for Allen, et al, Proc. Nat. Acad. Sci. (PNAS), vol 110, page 17768 for an example. It is also shown in the attached figure. There, the downwind ratio of two collocated tracers (C2H2 and N2O) is within 1% of the ratio of their mass flow rates (Figure S4-4c). If N2O were replaced by CH4, the scenario in Figure S4-4 would be experimentally identical to configuration 1 of this manuscript. The tracer release-derived CH4 flow rate would then be within 1% of the known flow rate. This level of agreement for collocated sources is routine in the field. Therefore, the 19% deviation observed here strongly suggests that there is an issue with the measurement method (instrumental), or tracer/methane flow rate.

If configuration 1 were viewed a "control", then the +19% bias indicates that other aspects of the measurements are +19% biased, not the tracer method inherently. In that case, it should be used as an bias offset – that is, the +29% bias for configuration 2 should actually be 29%-19% = 10%, for configuration 3 should be 17%-19% = -2%, and for configuration 4 should be 58%-19% = 39%.

Lines 605 – 613: Please better explain why this tracer ratio method is not working, given that earlier you say that position errors perpendicular to the wind should not have such a large effect. Was the wind varying? Also, the "configuration 4" panel in Figure 5 shows only one methane plume. Given that there are two CH4 sources, why don't you see two CH4 plumes, or at least a broad CH4 plume? We would expect the two CH4 plumes to look like a composite of the C2H2 and CH4 plumes in configuration 3, which has identical separation.

4. Initial guesses for optimization

Paragraph at 276 & Line 474: Please explain how initial guesses completely independent of measurements can be done in practice, particularly for emission sources where the expected emission magnitudes may span many orders of magnitude (e.g. factor of 100, instead of 80%). This would be the case, for example, for certain oil and gas

emission experiments. Is it possible to use tracer release result (without any dispersion corrections) as the prior?

Specific Comments:

Lines 191 - 196: Other methods of calculating the ratio are commonly used, notably taking the slope of the plot of the methane vs tracer plume signals. This is generally found to be more precise than measuring the area under each plume, because it does not depend upon choice of background. See Roscioli et al: Roscioli, J. R.; Yacovitch, T. I.; Floerchinger, C.; Mitchell, A. L.; Tkacik, D. S.; Subramanian, R.; Martinez, D. M.; Vaughn, T. L.; Williams, L.; Zimmerle, D.; et al. Measurements of methane emissions from natural gas gathering facilities and processing plants: measurement methods. Atmos. Meas. Tech. 2015, 8, (5), 2017; DOI: 10.5194/amt-8-2017-2015.

Lines 225 : There is considerable debate in the atmospheric modeling community on the timescale appropriate for the canonical A-D stability classes (e.g. Pasquill-Gifford stability classes). The consensus seems to be more on the order of 10-15 minutes than 1-2 minutes. See, for example: Fritz, B. K.; Shaw, B. W.; Parnell, C. B. J. Influence of meteorological time frame and variation on horizontal dispersion coefficients in Gaussian dispersion modeling. Trans. ASABE. 2005, 48, (3), 1185; DOI: 10.13031/2013.18501

Line 302: "... spatial offset between the measured plume and the actual plumes due to the lag between the air intake and the concentration measurement." I recommend adding the parenthetical statement: (also known as inlet lag or inlet time)

Line 340: correct "serie" to "series"

Line 351: While the instrument reporting time is noted (2 seconds), the instrument response time is not. Furthermore, the data depicted for configuration 1 in Figure 5 suggests the time response for the two channels is not fully matched. Is the instrument reporting all mixing ratios simultaneously, or does the instrument sub-divide the

2 second interval to quantify each of the noted species.

Line 429: Is the 5th percentile of transect concentrations sufficient to determine baseline? Do the results change if the 2nd percentile, or 10th percentile is used?

Lines 561 to 567 and Figure 3: Why not show these results as a function of relative distance downwind, i.e. (distance between source and tracer)/(distance between site and measurement)? The distances in meters shown can not be easily generalized. I further suggest putting panels a) and b) on the same vertical scale, and adding gridlines at the same intervals for both.

All Figures: Please increase font size of all labels and numbers so that they are readable at the width of a normal sheet of paper.

Figure 4: Consider showing only Figure 5 (representative plume transects) instead of the whole data set. These full data results, depicted in Figure 4, might be better left to the supplementary info. Even better, consider publishing these results as a test dataset. If Figure 4 is to remain, then the vertical axes should be rescaled to see all of the plume intensities.

Figure 6: Figure 6 is the most important figure of the manuscript and needs to be reformatted for legibility. It is currently much too small. Consider perhaps a small cartoon drawing of "config. 1" and "config 4". To clarify the difference between the 2 sets of results shown. Please also label the meaning of the shaded area ("concentrations" plots) in the legend or figure title.

Line 565: Instruments with lower levels of detection than the one used here are available. Please alter this statement to reflect this fact: e.g.: "...signal to measurement noise ratio would likely be too small, using these instruments, to derive..."

Section 4.3, Lines 614: Please reference Goetz et al, who in 2015 used a similar approach to this.

Line 651: It is possible, (see Roscioli et al.) in some cases, to co-locate tracers and

emission vectors for real experiments. Please rephrase “which can hardly occur”

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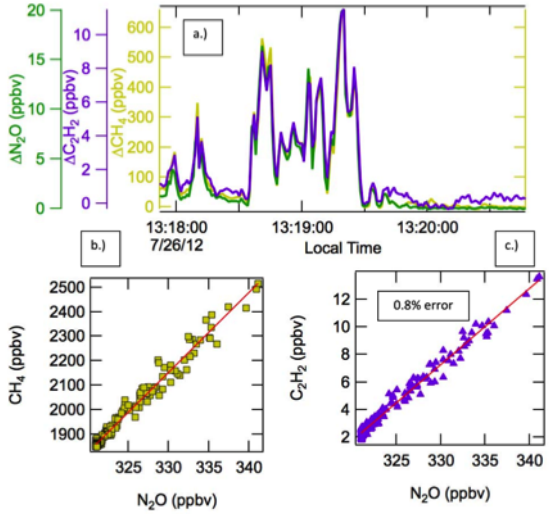

Figure S4-4 a.) Methane, acetylene and N_2O plumes observed downwind of a production site; tracers were co-located; b.) the average ratio of methane to N_2O in the plume, determined using second by second observations of methane and N_2O is indicated by the slope of the line; this ratio was used in Equation S4-2, with the known release rate for N_2O, to estimate methane emissions; c.) the average ratio of acetylene to N_2O in the plume determined using second by second observations of acetylene and N_2O in the plume is indicated by the slope of the line; the 0.8% error indicates the difference between the ratios of the observed concentrations in the plume and the ratios of the tracer release rates for this site.

Fig. 1.

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